



Full length article

First-principles determination of sub-lattice 3D vector magnetic fields in antiferromagnetic hematite

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ABSTRACT

In quantum materials, accurately predicting the full three-dimensional vector magnetic field inside materials is crucial for bridging theoretical magnetism and experimental observations. However, conventional approaches relying on the vector potential face inherent limitations of field strength arbitrariness due to gauge dependence. By integrating first-principles calculations with microscopic magnetostatic field modeling based on atomic-scale spin densities, this work quantitatively determines the 3D vector magnetic fields inside the unit cell of hematite (α -Fe₂O₃), without introducing a vector potential. This approach adopts gauge invariance and variational principle, to enable precise determination of field magnitudes. Applying this approach to antiferromagnetic hematite as a model system, this paper achieves the first clear visualization of its sublattice-resolved 3D vector magnetic field and quantitatively analyzes the internal field evolution across the Morin transition. It is found that although no region of the crystal in antiferromagnetic hematite exhibits a vanishing total magnetic field, each vector component spans a substantial volume where its magnitude approaches zero. Calculations yield a most probable internal flux density of 1.5 T below and 1.0 T above the Morin transition, showing close agreement (within 4%) with experimental values in both regimes. Notably, 99% of the crystal volume is subjected to magnetic flux densities below 17 T in both phases. Crucially, the ultrahigh effective magnetic field inferred from Mössbauer spectroscopy reflects a localized on-site scalar energy coupling, fundamentally distinct from the extended 3D magnetostatic vector field landscape. In contrast, the statistical characteristics of the total field from muon spin relaxation and rotation (μ SR) and the vector components from neutron scattering provide direct microscopic evidence. The morphology of the magnetic field isosurfaces evolves from reflecting long-range lattice order at low fields to tracing localized ionic orbitals at high fields (above 2 T). Furthermore, the consistency of the predicted surface fields with nitrogen-vacancy center magnetometry provides a complementary validation at the hematite's surface, solidifying the reliability of the approach across different spatial regimes. New results thus yield a unified framework that reconciles the apparent discrepancies in critical transition fields from various experiments, which offers fundamental insights for the understanding and control of magnetic structures at the atomic scale in quantum materials and devices.

1. Introduction

Significant advances in high-resolution probes have enabled atomic-scale structural characterization of quantum materials, achieving resolutions extending to the picometer regime [1]. Techniques like electron ptychography achieve resolutions below 20 picometers with sub-picometer phase precision, even in thick samples [2]. It is feasible to couple the Schrödinger equation with Maxwell's equations through gauge invariance and the variational principle (see Supplementary Material, Section 1.0). However, resolving magnetic fields at comparable resolution and accuracy remains challenging, as the quantum exchange–correlation energy of charges often dominates over magnetic interactions (see Supplementary Material, Section 1.1).

Recent breakthroughs in differential phase contrast scanning transmission electron microscopy (DPC-STEM) have enabled real-space visualization of magnetic fields in antiferromagnetic hematite (α -Fe₂O₃) at atomic resolution. This was achieved by isolating magnetic phase shifts and enhancing signal-to-noise ratios in field-free environments [3]. Complementary developments in time-dependent electron scattering simulations optimize the separation of weak magnetic signals from dominant electrostatic interactions, particularly in antiferromagnets [4]. Nevertheless, these techniques provide only uncalibrated projected images of magnetic field distribution (see Supplementary Material, Section 1.4.2).

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The relationship between magnetic structure and property at the atomic scale presents a critical yet unresolved question: How strong is the magnetic field within an antiferromagnetic lattice? A complete answer to this question has posed significant obstacles that must be overcome.

The first difficulty is revealing magnetic fields in 3D sub-lattice space with full 3-axis vector components. Neutrons and γ -rays cannot provide field distribution at high spatial resolution, whereas charged particles such as electrons and muons are dominated by electrostatic interaction. Although differential phase contrast techniques can be used, three-component magnetic fields in 3D space are difficult to unravel from surface or projection images via electron microscopy [3,4].

The second difficulty is piecing together a complete magnetic field map, which is consistent with all available measurements from various probes. Mössbauer studies estimate hyperfine fields at the iron nuclei in hematite above 50 T at low temperature and under ambient conditions [5,6]. At higher temperatures broader distributions in the field have been observed [7]. Mössbauer spectra associated with the spin-flop Morin transition have also shown a critical field of 6.75 T [8]. Muon spin rotation experiments yield a 0.8 T jump around the Morin transition [9]. According to a polarized-neutron study, the field dependent reflection curve indicates a cusp near 0.1 T and saturation above 1 T [10]. Although they span more than two orders of magnitude, the considerable differences in magnetic fields obtained by multiple detection methods have not been well understood yet.

The third difficulty lies in theoretical modeling and can be overcome through first principles plus 3D field calculations from the magnetic moment distribution. Under the local approximation of spin-density functional theory and the augmented-spherical-wave method incorporating spin-orbit coupling and noncollinear moment arrangements, the total energy correctly predicts the experimentally observed magnetic order of the ground state. Using the total energies of different magnetic configurations, the Néel temperature of hematite has been theoretically estimated [11]. An effective spin Hamiltonian allows one to determine parameters of isotropic and anisotropic Dzyaloshinskii-Moriya exchange interactions, so that the canting angles of magnetic moments in hematites are predicted to be in reasonable agreement with experimental data [12]. Based on ab initio calculations of the resulting weak noncollinear magnetization in addition to symmetric magnetic exchange, the absolute Dzyaloshinskii-Moriya interaction vector direction in hematites has been determined [13].

After relativistic formalisms for elastic electron scattering in a magnetostatic environment were examined, spin current effects and coupling to the vector potential were used to calculate the 2D magnetic field in α -Fe [14]. For a realistic description of magnetism in a solid, considering a stationary Pauli equation with a nonuniform magnetic field and corresponding vector potential in the Coulomb gauge, the layer-by-layer spin current density and magnetic fields of α -Fe are modeled from spin density functional theory, by using a Gordon decomposition and neglecting orbital currents [15]. To properly address the internal magnetostatics in magnetic solids, an exact connection between spin-density functional theory and current-density functional theory has been constructed [16].

To address these challenges, the author introduces a first-principles framework for determining the complete 3D vector magnetic field inside the unit cell of hematite. The approach combines segmented all-electron calculations, yielding a gauge-invariant spin density, with magnetostatic laws to compute the resulting magnetic fields. Hematite serves as an ideal model system: it is a ubiquitous antiferromagnet and a paradigm for multiscale magnetic studies spanning atomic to bulk dimensions [17] (see Supplementary Material, Section 1.4). Its corundum structure [18] exhibits characteristic Néel (950 K) and Morin (250 K) transitions [19,20], presenting intrinsic magnetic field contrast changes due to spin reorientation. With vanishingly small magnetic susceptibility and negligible stray fields, hematite is increasingly relevant for high-speed, high-density spintronic applications [3,21]. Furthermore,

magnetic analysis of hematite accounts exclusively for the dominant electron spin contributions, while the orbital magnetic moment and the magnetic moment arising from itinerant electrons were neglected, as Fe^{3+} ions in insulating crystalline hematite possess half-filled 3d orbitals and exhibit quenched orbital moments.

2. Computational methods

First principles self-consistent field calculations were carried out using CP2K with all-electron Gaussian-augmented plane-wave (GAPW) implementation [22], which employs a meta-generalized gradient approximation (meta-GGA) density functional to solve the Kohn-Sham equations of density functional theory in a periodic system without empirical parameters [23] (see Supplementary Material, Sections 1.2 and 2.0). To expand the charge density in plane waves, an energy cutoff of 350 Ry was used, and the convergence criterion over the maximum component of the wave function gradient was set to $1e-7$. To adequately correct for the strong electron correlation of the Fe 3d-electrons, the DFT+ U method is applied using an effective Hubbard parameter $U_{\text{eff}} = U - J = 5.0$ eV to obtain reasonable band structures [24]. All the DFT calculations were performed via the TPSS functional with pcseg-2 basis sets [25], which were downloaded from Basis Set Exchange platform [26]. Calculations of the spin moment were performed with $3 \times 3 \times 3$ supercells of primary trigonal unit cell in hexagonal axes to exclude artifacts of the periodic boundary conditions. Detailed basis set parameters and complete input files are provided in Sections 2.0.2 and 2.0.4 of the Supplementary Material, respectively.

DFT calculations output charge density and spin density data of $60 \times 60 \times 160$ elements with hexagonal cell parameters of $a_{\text{Hex}} = b_{\text{Hex}} = 5.03417 \text{ \AA}$ ($\angle ab = 120^\circ$), $c = 13.7321 \text{ \AA}$, and $a_{\text{Hex}} = b_{\text{Hex}} = 5.035128 \text{ \AA}$ ($\angle ab = 120^\circ$), $c = 13.7581 \text{ \AA}$ for the hematite lattice at 10 K and 300 K, respectively. To orthogonalize components of magnetic field, the primary trigonal unit cell in hexagonal axes is transformed into an orthogonal unit cell via two vertical diagonals of the $x_{\text{Hex}}y_{\text{Hex}}$ prismatic base plane as new x and y basis vectors. During the transformation, the z direction remains unchanged, but the area of the rectangular xy base plane is doubled from the original $x_{\text{Hex}}y_{\text{Hex}}$ prismatic base plane, so that $60 \times 120 \times 160$ elements of spin density data are used in space field calculation with cuboid volume of $8.719 \times 5.03417 \times 13.7321 \text{ \AA}^3$ and $8.7211 \times 5.035128 \times 13.7581 \text{ \AA}^3$ for the periodic cells at 10 K and 300 K, respectively.

Only the spin moment is taken as the internal field source to generate 3D vector magnetic fields for hematite, since neither the orbital magnetic moment nor the magnetic moment from itinerant electrons are quantitatively calculated because Fe^{3+} ions with half-filled 3d orbitals in insulating crystalline hematite are orbital quenched. Compared with electron spins, the spin moment effect of iron and oxygen nuclei on the magnetic field within the hematite lattice can be ignored, and nuclear spins are also neglected. The calculation process is illustrated in Fig. 1, where the main procedures are outlined in the following five steps (see Supplementary Material, Sections 1.3 and 2.2–2.4): (1) The continuous spin density $\sigma(\mathbf{r})$ obtained from first-principles calculations is discretized into localized magnetostatic dipole moments $\mathbf{m}(\mathbf{r})$, with sublattice antiferromagnetic (AFM) orientations either directly generated from the vector spin density or post-assigned to the scalar spin density, as denoted in Formula (a). (2) The real-space magnetic field $\mathbf{h}(\mathbf{r})$ is integrated via a microscopic magnetic dipole model, as indicated in Formula (b). To prevent unphysical calculation singularities at the origin (zero distance to the source magnetic dipole), a short-range exclusion cutoff radius of $R_c = 15 \text{ pm}$ is applied, perfectly adapting to the spatial grid resolution utilized in the DFT calculations. (3) The internal magnetic flux density $\mathbf{B}_{\text{int}}(\mathbf{r})$ in the 3D lattice space is evaluated via the fundamental constitutive relation (Formula (c)). The overall local magnetic field $\mathbf{B}$ is then formulated by rigorously superimposing $\mathbf{B}_{\text{int}}(\mathbf{r})$ with the externally applied field

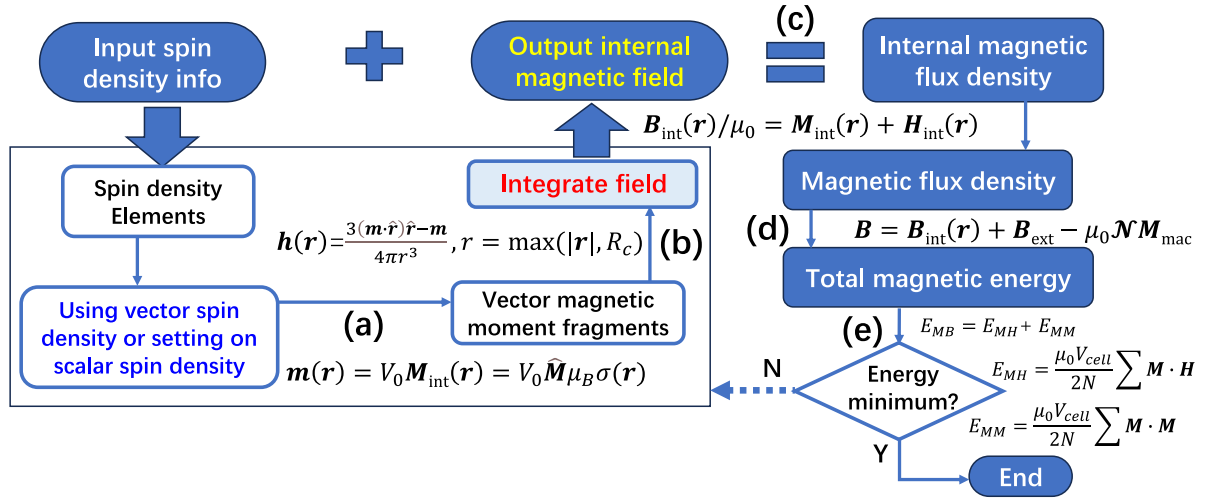


Fig. 1. Workflow for reconstructing the 3D vector magnetic field from first-principles spin density. The key computational steps and governing equations are detailed as follows: (a) Conversion of the continuous spin density $\sigma(\mathbf{r})$ into discrete magnetostatic dipole moments $\mathbf{m}(\mathbf{r}) = V_0 \mathbf{M}_{\text{int}}(\mathbf{r})$, where V_0 is the volume element, serving as the fundamental link between electronic structure and local magnetic models [27]. (b) Real-space integration of the magnetic field $\mathbf{h}(\mathbf{r})$ generated by the dipole moments, applying a short-range cutoff distance R_c to prevent unphysical calculation singularities at the origin. (c) Derivation of the internal magnetic flux density $\mathbf{B}_{\text{int}}(\mathbf{r})$ using the fundamental relation $\mathbf{B}_{\text{int}}(\mathbf{r})/\mu_0 = \mathbf{M}_{\text{int}}(\mathbf{r}) + \mathbf{H}_{\text{int}}(\mathbf{r})$. (d) Formulation of the overall local magnetic field $\mathbf{B}$, which rigorously superimposes the internal field $\mathbf{B}_{\text{int}}(\mathbf{r})$, the externally applied field $\mathbf{B}_{\text{ext}}$, and the macroscopic shape-dependent demagnetizing correction $-\mu_0 \mathcal{N} \mathbf{M}_{\text{mac}}$. (e) Evaluation of the thermodynamic magnetic energy (e.g., E_{MH} and E_{MB}) to provide feedback for optimizing spin orientation, consistent with established energy-mapping analysis principles for determining magnetic interactions from density functional theory [27].

and the macroscopic shape-dependent demagnetizing correction (Formula (d)). Although the external field and demagnetizing correction are included in the methodology, for the antiferromagnetic hematite considered here the macroscopic magnetization is negligible and no external magnetic field is applied. Therefore, the magnetic flux density calculated in this work is essentially governed by the internal field alone. To achieve a converged periodic distribution within the central unit cell, spin moments from a $3 \times 3 \times 3$ orthogonal supercell are utilized as the field source for bulk states, whereas a $3 \times 3 \times 1$ supercell is employed for mapping surface field distributions. (4) Guided by the principles of energy-mapping analysis [27], the thermodynamic magnetostatic energies within the unit cell are evaluated (Formula (e)). Specifically, the non-local spatial correlation term (E_{MH}), which captures environmental coupling and drives relative spin configurations, is rigorously distinguished from the purely local self-energy term (E_{MM}), which underpins single-ion anisotropy. Together, they constitute the total material-specific magnetostatic energy (E_{MB}). These energy terms serve as a quantitative feedback mechanism to verify the physical validity of the continuous field reconstruction and optimize the assigned spin orientations (for detailed derivations of the energy partitioning, see Supplementary Material, Section 1.3.4). (5) The preceding four steps are iteratively repeated until the thermodynamic energy minimum criterion is strictly satisfied.

During the preparation of input files for CP2K and the analysis of the output spin density, Multiwfn has been used [28]. After the calculated spin density or vector magnetic fields are ready, visualization and snapshots of the 3D isovalue surface for spin density or magnetic field data are obtained via the VMD (visual molecular dynamics) package [29].

To enable site-selective correlation with μSR measurements, the sublattice total magnetic induction $\mathbf{B}_i(\mathbf{r})$ was mapped jointly with the raw electronic charge density $\rho(\mathbf{r})$ obtained directly from DFT. The spatial correlation between the total magnetic field and the charge density—which defines the electrostatic potential landscape—was then quantified using two-dimensional overlap-count distributions plotted on a logarithmic scale.

A detailed introduction and step-by-step operation, together with a video chain and related input files with source codes, are provided in the supplementary multimedia materials to help readers understand and reproduce the main findings.

3. Results and discussion

3.1. Evaluation of spin density distribution in lattice

The calculation commences with the crystal structure and electronic distribution within the hematite lattice. Fig. 2a displays the primary trigonal unit cell in hexagonal axes of hematite, featuring a double-atomic-layer arrangement of Fe-O octahedra along the z -direction with alternating spin orientations (up-down-down-up). Below the Néel temperature [19], adjacent double atomic layers exhibit antiferromagnetic coupling while maintaining ferromagnetic alignment within each layer. Below the Morin temperature [20], spin moments orient along $\langle 001 \rangle_{\text{Hex}}$ in antiparallel configuration. At intermediate temperatures (between T_{Morin} and $T_{\text{Néel}}$), spins rotate toward $\langle 1\bar{1}0 \rangle_{\text{Hex}}$ directions with slight tilting [13,30].

The initial step in 3D field calculation involves generating self-consistent field optimized spin-dependent electronic structures (Fig. 2), utilizing experimental rhombohedral lattice parameters at 10 K and 300 K [31]. The density of states reveals spin polarization primarily in Fe d -orbitals and partially in O p -orbitals. A magnetic moment of $4.3 \mu_B$ per Fe atom is then obtained, consistent with the range of reported experimental values (4–6 μ_B) [31,32]. The calculated band gap of 2.1 eV aligns with experimental (2.14 eV) [33] and theoretical (2.2 eV) [34,35] estimates.

To examine orientation-dependent field variations, the full vector magnetic fields are computed for two characteristic spin configurations (OP-AFM and IP-AFM) within a transformed orthogonal unit cell. This cell is derived from the trigonal primitive cell in hexagonal axes (Fig. 2) by selecting two orthogonal diagonals of the $x_{\text{Hex}}y_{\text{Hex}}$ basal plane as new x and y axes. The cell volume consequently doubles from the original parameters in hexagonal axes ($a_{\text{Hex}} = b_{\text{Hex}} = 5.03417 \text{ \AA}$, $\angle ab = 120^\circ$, $c = 13.7321 \text{ \AA}$) to $8.719 \times 5.03417 \times 13.7321 \text{ \AA}^3$. Magnetic fields are computed using spin moments from a $3 \times 3 \times 3$ orthogonal supercell to ensure periodicity.

3.2. Total field in agreement with μSR

Fig. 3 presents the resolved 3D vector magnetic fields for both magnetic configurations at 10 K. The individual spatial components

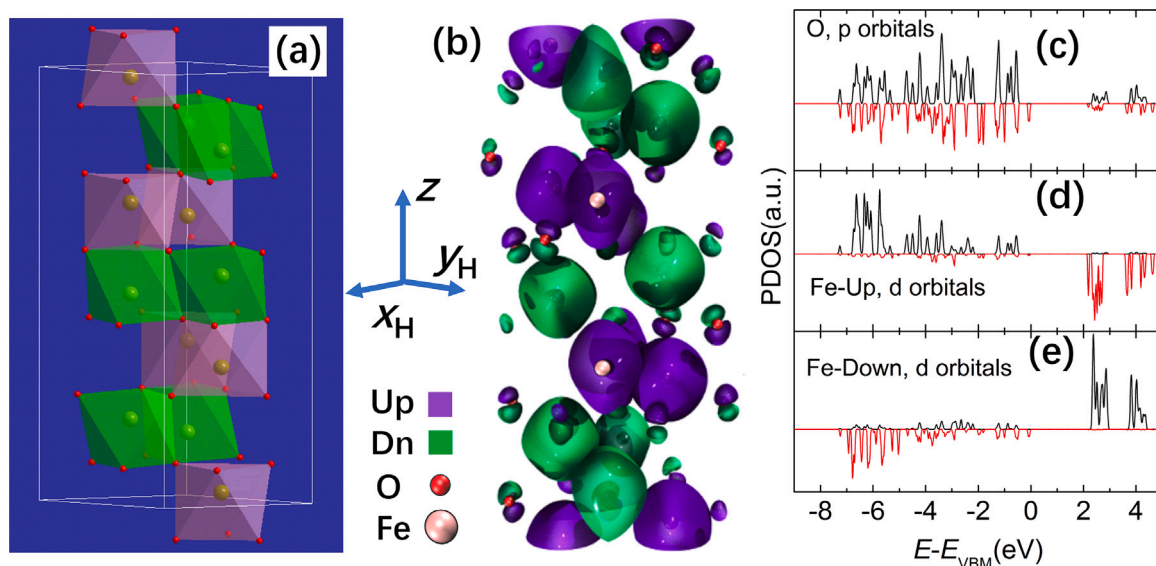


Fig. 2. (a) Trigonal primitive cell in hexagonal axes of hematite ($\alpha\text{-Fe}_2\text{O}_3$), with spin-up (violet) and spin-down (green) Fe-O octahedra. (b) Spin density isosurfaces at $\pm 0.007 \text{ \AA}^{-3}$ from DFT calculations. (c–e) Orbital-projected density of states at 10 K: (c) O p -orbitals, (d) spin-up Fe d -orbitals, (e) spin-down Fe d -orbitals (α/β electrons shown in black/red).

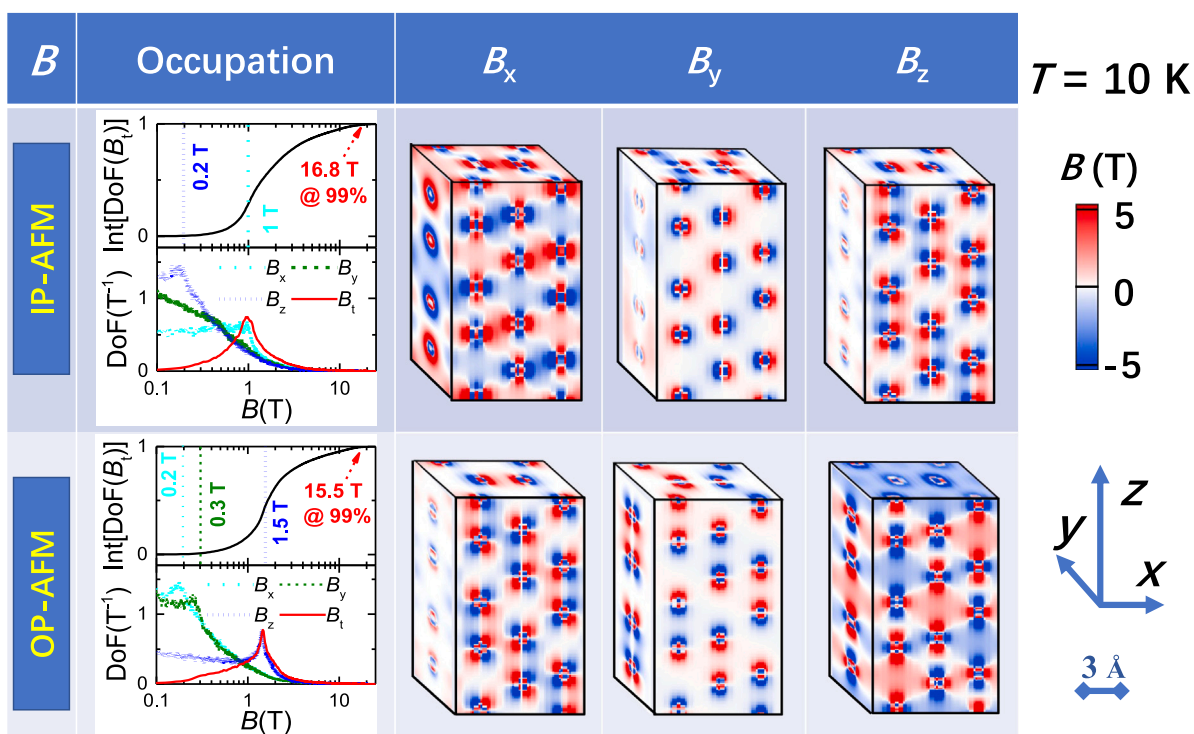


Fig. 3. 3D vector magnetic fields for out-of-plane (OP-AFM) and in-plane (IP-AFM) configurations at 10 K. Columns show B_x , B_y , B_z components and statistical distributions. Occupation column: Bottom panel shows field-dependent volume fraction density for components and total field (B_t); Upper panel displays cumulative distribution of B_t .

(B_x , B_y , B_z) and the total field magnitude B_t are displayed alongside their statistical volume-occupation distributions (see Supplementary Material, Section 2.5). The spatial distributions of the three magnetic field components were mapped onto the unit cell using a stereographic projection. For the low-temperature out-of-plane antiferromagnetic phase (OP-AFM), B_t exhibits a sharp peak at 1.5 T, indicating that this specific field strength dominates the fractional volume of the lattice—in notable agreement with zero-field muon spin rotation

(ZF- μ SR) experiments reporting local fields around 1.56 T [30,36–38]. Conversely, the in-plane canted phase (IP-AFM) above the Morin transition reveals a broader distribution with a primary peak centered near 1.0 T, reflecting the relatively uniform dispersion of B_x below 1 T. This broad profile is consistent with μ SR measurements above T_{Morin} ($\sim 0.7\text{--}0.96$ T) [36], where the flatter planar field topology enables muons to sample lower local fields. Notably, the effective magnetic dipole field measured via antiferromagnetic resonance (AFMR) is 1.026 T [39],

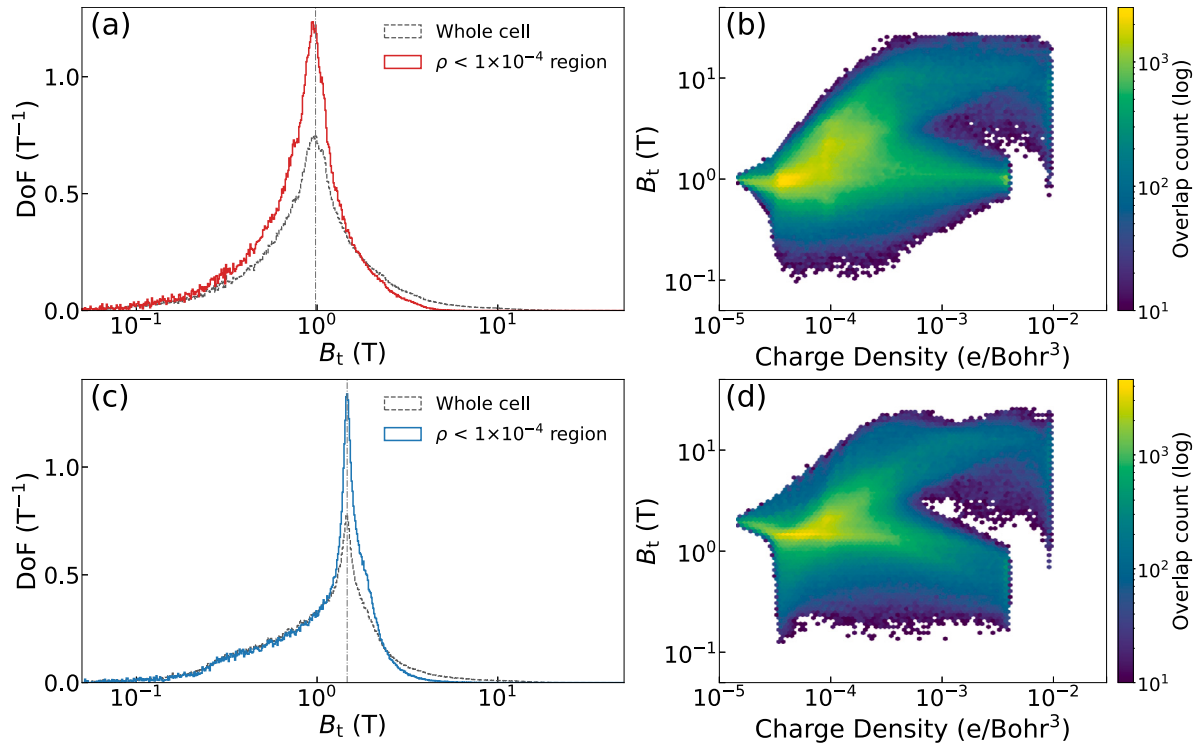


Fig. 4. Charge-density-resolved analysis of the internal vector magnetic field and cross-probe correlation with μ SR. (a, b) The in-plane antiferromagnetic phase (IP-AFM, $T > T_{\text{Morin}}$), corresponding to the IP-AFM configuration in Fig. 3: (a) One-dimensional probability distribution of the total magnetic field magnitude B_t comparing the whole-cell volume with the low-charge-density interstitial regions ($\rho < 1 \times 10^{-4} \text{ e/Bohr}^3$), both exhibiting the identical most probable field peak centered at 1.0 T. (b) Two-dimensional joint histogram of local electronic charge density ρ (e/Bohr^3) versus B_t (T), where the bin color denotes voxel counts on a logarithmic scale. The ridge of maximum probability density remains invariant across nearly four decades of electronic charge density (10^{-5} to 10^{-2} e/Bohr^3). (c, d) The low-temperature out-of-plane antiferromagnetic phase (OP-AFM, $T < T_{\text{Morin}}$), corresponding to the OP-AFM configuration in Fig. 3: (c) One-dimensional field distribution of B_t showing the sharp, dominant peak at 1.5 T for both the whole-cell and charge-density-filtered interstitial domains. (d) Two-dimensional joint histogram of ρ versus B_t for the OP-AFM phase, displaying the corresponding invariant ridge line across the low-to-intermediate charge density spectrum. This structural invariance demonstrates that across both magnetic phases, the most probable field of the whole unit cell intrinsically coincides with that of the interstitial low-charge-density channels, providing a rigorous physical foundation for validating μ SR local-field measurements via first-principles field distributions.

differing by less than 4% from calculated 1.0 T peak for the IP-AFM state.

Despite this straightforward numerical concordance, assigning the whole-cell volumetric field peak directly to the μ SR observable is methodologically premature. Implanted muons do not perform uniform Monte Carlo sampling throughout the crystal volume. Regardless of specific sign-dependent local interactions (e.g., electrostatic trapping of positive muons or atomic shell binding of negative muons), thermalized muons in insulating transition-metal oxides universally localize within deep electrostatic interstitial potential wells characterized by depleted interstitial valence electron density [37]. Therefore, an unbiased physical comparison with local μ SR measurements requires filtering the calculated 3D vector field to include solely the interstitial spatial domains accessible to the probe.

To address this site-selection effect, the author performed a systematic, charge-density-resolved analysis of the sub-lattice vector magnetic field, as summarized in Fig. 4. The two-dimensional correlation spectrum of local charge density $\rho(\mathbf{r})$ versus total field magnitude $B_t(\mathbf{r})$ (Fig. 4b) uncovers a striking topological feature of $\alpha\text{-Fe}_2\text{O}_3$: the ridge of maximum probability density along the B_t axis remains essentially invariant across nearly four decades of local charge density, extending continuously from 10^{-5} up to 10^{-2} e/Bohr^3 .

When the sampling volume is restricted strictly to the interstitial stopping domain ($\rho < 1 \times 10^{-4} \text{ e/Bohr}^3$, Figs. 4a and 4c), the resulting filtered field spectrum displays a peak position that is practically identical to that obtained from the unweighted whole-cell integration. In hematite, this equivalence is not a trivial artifact; rather, it originates from the spatial magnetostatic profile of the corundum lattice. The

high-symmetry interstitial channels correspond to broad magnetostatic saddle points, where local spatial gradients $\nabla B_t(\mathbf{r}) \approx 0$ produce an extensive volume fraction at the same characteristic field magnitude. Consequently, while the unweighted whole-cell histogram and the interstitial-filtered spectrum yield virtually identical peak values, the charge-density-filtered representation provides the physically rigorous foundation for validating μ SR measurements.

Because the implanted muon operates as a point-like, in-situ magnetometer, its spin precesses directly around the local vector induction field $\mathbf{B}_{\text{loc}}$ at its interstitial site with a Larmor frequency $\omega_\mu = \gamma_\mu |\mathbf{B}_{\text{loc}}|$. This measurement determines the absolute magnitude of the local field without reliance on secondary phenomenological models. By establishing that the characteristic interstitial field in hematite matches the predominant sub-lattice field landscape, this charge-density-resolved mapping provides a direct, quantitative bridge between first-principles electronic structure calculations and local magnetic probe experiments.

3.3. Features of component fields

Component-specific features are also resolved in Fig. 3. The volume fraction density distributions of B_x in OP-AFM and B_z in IP-AFM show a peak at 0.2 T, corresponding to low field cusp in magnetic field dependent neutron diffraction [10], and Mössbauer response versus magnetic field below 4 T [40], which is evidently a fine and exact reason from the view angle of the component magnetic field. For the volume fraction density of B_y , a weak peak is observed at 0.3 T in OP-AFM but no peak is observed in IP-AFM, which originates from

a greater volume fraction of near-zero magnetic field in the y component. The 3D field mapping and statistical analysis reveal a crucial distinction: while each component of the magnetic field passes through zero within finite volumes, the magnitude of the total field B_t remains strictly non-zero without exception.

Analysis of the 3D field maps reveals that the volumes of positive and negative fields for all three components are approximately equal and symmetric within the unit cell. Furthermore, consistent with the reciprocity principle, the distribution of the B_x component in the OP-AFM configuration is identical to that of the B_z component in the IP-AFM configuration. In contrast, the primary field components B_x in the IP-AFM state and B_z in the OP-AFM state, exhibit distinct distributions: the latter shows a clear positive-negative layering along the z -axis, whereas the former displays a uniform, interwoven mix of positive and negative fields. Moreover, the distributions of the B_y component in the two AFM states are not simply related by a global sign reversal, a consequence of the alternating spin configuration between adjacent double-atomic-layer along the z -axis in hematite.

3.4. High field region compared with mössbauer spectroscopy

The sub-lattice magnetostatic field behavior in the high-field regime (> 7 T) was evaluated against both macroscopic critical fields and microscopic local-probe measurements. Macroscopically, spin-flop transitions and magnon excitations induced by external fields typically occur around 7–10 T, and can reach up to 50 T under dynamic conditions [41,42]. Microscopic calculations reveal that 99% of the cell volume is permeated by internal fields below 15.5 T (OP-AFM) and 16.8 T (IP-AFM). Extreme fields exceeding 20 T occupy a minimal spatial fraction ($< 1\%$) and are strictly localized within a ± 5 pm vicinity of the Fe centers. This intense localization reflects the steep spatial gradients of the spin density approaching the atomic cores, though the finite grid resolution (~ 15 pm) of the DFT source limits sub-nuclear scale precision.

This spatial distinction is crucial for reconciling the sub-lattice vector field integration with ^{57}Fe Mössbauer spectroscopy, which universally reports massive effective fields of ~ 50 –54 T in $\alpha\text{-Fe}_2\text{O}_3$ [38]. Unlike interstitial probes (e.g., μSR) that directly sample the extended 3D magnetostatic vector fields, the Mössbauer effective magnetic field ($\mathbf{B}_{\text{eff}}$) is a superposition of the macroscopic local lattice field ($\mathbf{B}_{\text{loc}}$) and a dominant on-site hyperfine field ($\mathbf{B}_{\text{hf}}$) [43]. According to standard hyperfine interaction theory [43], $\mathbf{B}_{\text{hf}}$ decomposes into the Fermi contact field (B_s), the orbital field (B_L), and the on-site dipolar field (B_D). For the high-spin Fe^{3+} ($3d^5$) ions in hematite, the massive ~ 50 T observation is overwhelmingly governed by the Fermi contact term B_s , originating from the spin-polarization of core s -electrons by unpaired $3d$ valence spins [43].

Fundamentally, as formulated by Chen and Yang [43], the magnetic hyperfine Hamiltonian is a strict vector dot product ($H_M = -g\mu_N \mathbf{I} \cdot \mathbf{B}$), yet the experimental Mössbauer observable is a scalar energy eigenvalue (hyperfine splitting ΔE). This scalar energy is conventionally converted into an “effective magnetic field” scalar via $B_{\text{eff}} = \Delta E/(g\mu_N)$ [43]. Because the dominant Fermi contact term (B_s) arises from the spherically symmetric s -electron probability density at the nucleus, it manifests physically as an isotropic scalar energy coupling rather than a classical Maxwellian vector field traversing continuous space.

Therefore, reducing the complex Mössbauer energy splitting to a simple magnetic field scalar obscures the true three-dimensional vector magnetic field landscape of the lattice. The purely magnetostatic inter-site field ($\mathbf{B}_{\text{loc}}$) contributes only marginally (~ 1 T) to the total Mössbauer splitting. While Mössbauer spectroscopy serves as an unparalleled, ultra-sensitive indicator of localized atomic spin states and on-site electron correlations, the extended 3D vector magnetic field landscape dictating interstitial phenomena is accurately represented only by the lower-field, high-volume fraction continuum reconstructed in this study.

3.5. Isosurface visualization

Spatial field distributions are further visualized through 3D isosurfaces (Fig. 5 for global overview and Fig. 6 as a magnified view of the critical region of interest). Component fields (B_x , B_y , B_z) exhibit paired positive (violet) and negative (green) isosurfaces, while B_t (RMS magnitude) is strictly positive. Below 0.05 T, B_t isosurfaces are absent. At 0.35 T (OP-AFM) or 0.62 T (IP-AFM), fragmented isosurfaces coalesce into hexagonal grid layers parallel to the xy -plane, tilted at 72.5° to z . These layers evolve into “balloon”-like structures peaking at 1.5 T (OP-AFM) or 1.0 T (IP-AFM). Above 2.0 T, isosurfaces fragment into ellipsoidal enclosures around Fe nuclei, contracting with increasing field. The magnetic field isosurfaces transition from reflecting long-range order at low fields to representing localized orbitals at high fields, with the component isosurfaces showing orbital-like symmetries (Fig. 3 and Figs. 5–6).

The fine-grained visualization of three-dimensional vector magnetic field isosurfaces at the atomic scale is critically important, as it establishes an essential link between microscopic spin arrangements and emergent macroscopic magnetic behavior. Such visualization enables direct, quantitative comparison between first-principles predictions and experimental measurements from various local probes, thereby constructing a unified interpretation across experimental techniques. Furthermore, it reveals the precise magnetic landscape governing charge and spin dynamics in spintronic devices, guiding the rational design of quantum materials with tailored magnetic functionality.

3.6. Energy analysis for morin transition

Temperature effects on the spatial geometry are minimal: the cell volume changes by only 0.23% between 10 K and 300 K, yielding a negligible impact on the static field distributions for fixed spin configurations (Fig. 7a). However, the energy-mapping analysis (Fig. 7b) reveals a stark thermodynamic competition. Based on the partitioned first-principles magnetostatic energies, the low-temperature (10 K) preference for the out-of-plane antiferromagnetic (OP-AFM) order is overwhelmingly driven by the local single-ion anisotropy (E_{MM}). This local self-energy provides a stabilization of 1.5 meV/Fe relative to the in-plane configuration, decisively dominating over the non-local environmental coupling (E_{MH}) despite a minor thermodynamic penalty in the latter’s continuous dipole–dipole interactions. As the temperature increases above T_{Morin} , thermal fluctuations progressively destabilize this local single-ion anisotropy. This suppression allows the resilient non-local magnetostatic coupling (E_{MH}) and entropic contributions to take precedence, thereby driving the first-order Morin transition to the in-plane antiferromagnetic (IP-AFM) phase at 300 K (see Supplementary Material, Section 3.6).

The integration of magnetostatic energy analysis within a first-principles framework is pivotal for achieving a quantitative and predictive understanding of magnetic materials. Unlike phenomenological models, this approach calculates the long-range dipole–dipole interaction energy directly from the fundamental, atomically-resolved spin densities, placing it on an absolute energy scale consistent with other key contributions like exchange and anisotropy. By unraveling the total energy into its fundamental components, it allows us to definitively identify the physical origin of magnetic phenomena, accurately predict thermodynamic phase transitions, and rationally guide the design of materials with targeted domain structures and hysteresis properties. Therefore, first-principles magnetostatics transforms our understanding from a descriptive to a predictive level, enabling the engineering of magnetic behavior from the atomic scale up.

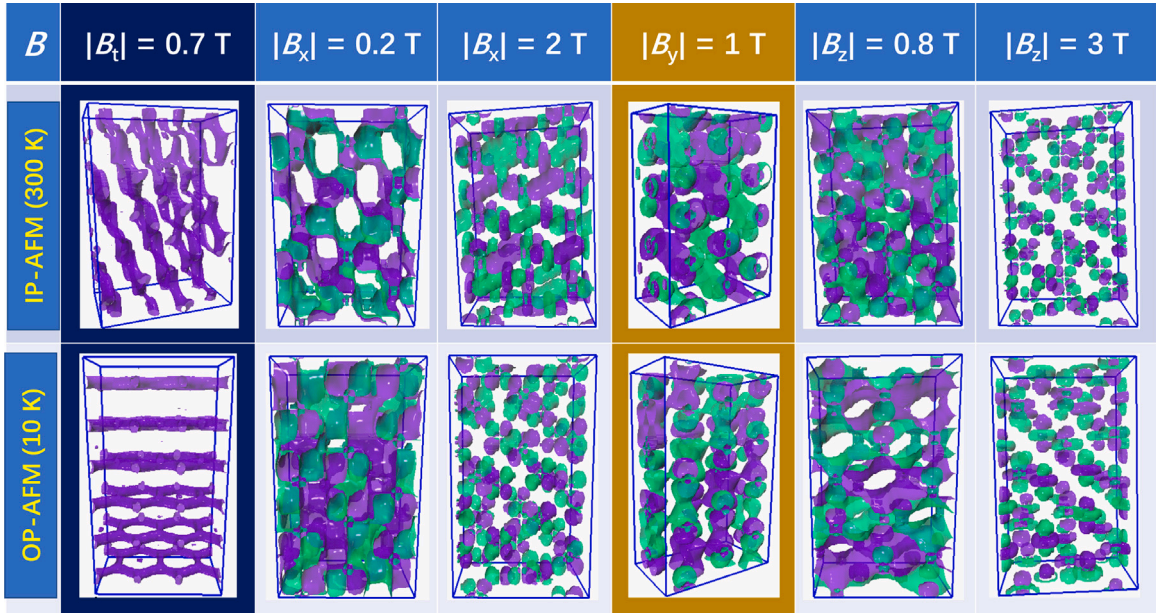


Fig. 5. 3D isosurfaces of magnetic field components (B_x , B_y , B_z) and total field (B_t) at selected field strengths. The visualization viewport for all 3D isosurfaces is defined by a rectangular bounding box of $8.7 \times 5.0 \times 13.7 \text{ \AA}^3$. Top row: OP-AFM phase at 10 K. Bottom row: IP-AFM phase at 300 K. Component fields show positive (violet) and negative (green) isosurfaces; B_t (RMS magnitude) is strictly positive.

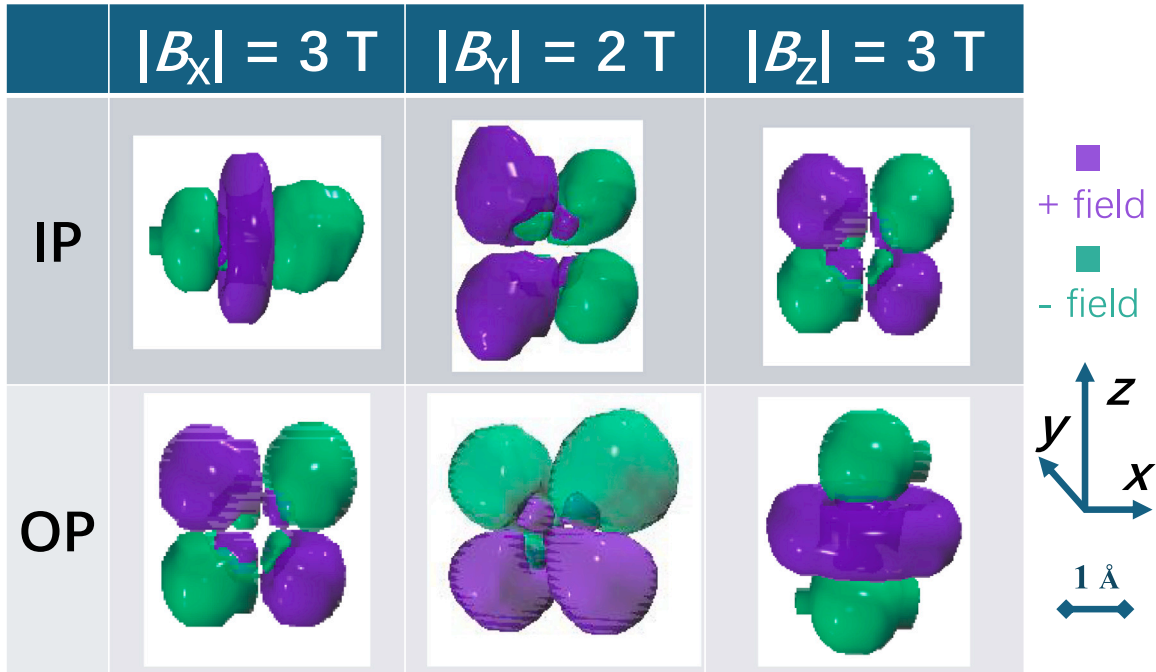


Fig. 6. Local 3D isosurfaces of magnetic field components: (a,c,e) IP-AFM at 300 K; (b,d,f) OP-AFM at 10 K. (a–b) B_x isosurfaces, (c–d) B_y isosurfaces, (e–f) B_z isosurfaces. Orbital-like symmetries emerge with minor crystal field distortions.

3.7. Surface field decay validated by diamond magnetometry

Surface field simulations (Fig. 7c–f) demonstrate rapid decay within 0.5 nm of surface Fe atoms, slowing beyond 1 nm. Predicted surface fields at the height of 5 nm ($B_z^{\text{OP-AFM}} = 0.5 \text{ mT}$ at 10 K; $B_x^{\text{IP-AFM}} = 0.4 \text{ mT}$ and $B_z^{\text{IP-AFM}} = 0.05 \text{ mT}$ at 300 K) quantitatively match nitrogen-vacancy (NV) center measurements [44]. Based on calculation of both distribution of before and after surface relaxation, which profoundly reshapes the magnetic field decay in hematite, results show it drives

a two-stage attenuation profile and amplifies the field strength by factors of 2–5 beyond 1 nm (see Supplementary Material, Section 3.6). Beyond the relaxation-driven attenuation captured by first-principles calculations, real hematite surfaces host complex magnetic domains due to competition of surface stress and magnetostatic anisotropy, resulting in long-range fields (e.g., 0.05 mT B_z at 500 nm). Accurately modeling these large-scale distributions for complex factors from first principles remains a computational challenge.

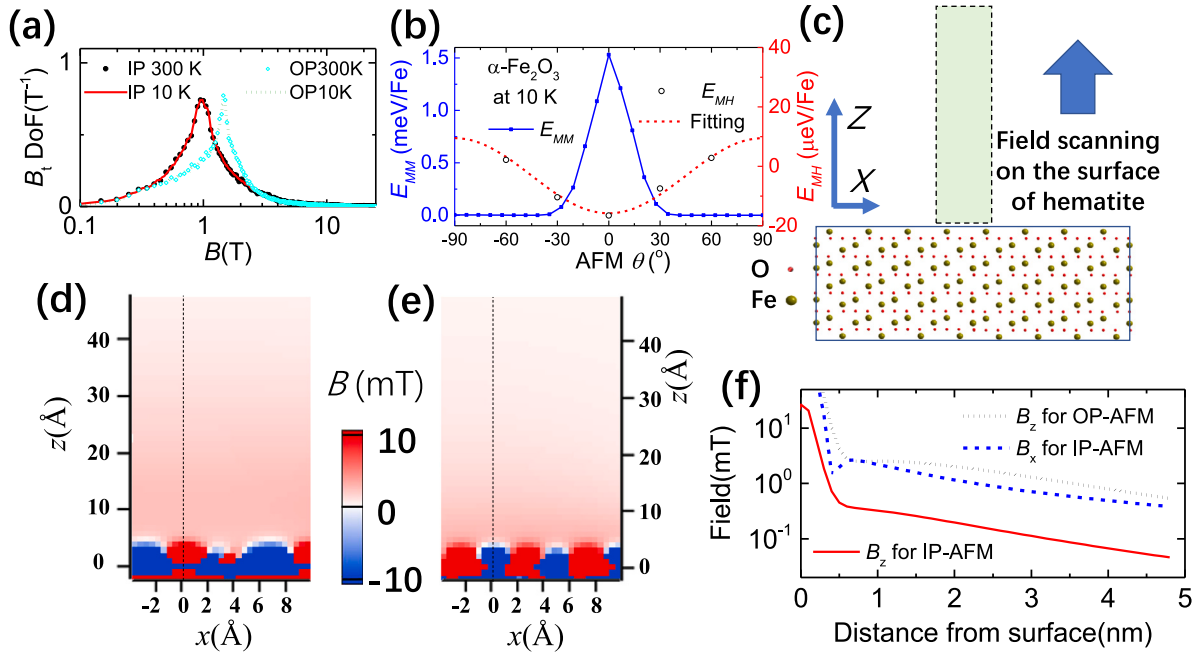


Fig. 7. (a) B_z distributions for OP/IP-AFM at 10/300 K. (b) Magnetostatic energy vs. spin orientation angle θ (scatters: simulation; curves: sinusoidal fits). (c) Measurement schematic for (001) surface fields. (d–e) Simulated surface field maps: (d) B_z^{OP} at 10 K, (e) B_x^{IP} at 300 K. (f) Field decay profiles from surface Fe atoms (black dotted line: B_z^{OP} at 10 K; blue dashed line: B_x^{IP} at 300 K; red line: B_z^{IP} at 300 K).

Diamond nitrogen-vacancy (NV) magnetometry offers a significant advantage for the absolute, vectorial measurement of magnetic fields at the nanoscale. It directly quantifies both the magnitude and orientation of the magnetic field vector by optically detecting the Zeeman shift of the NV center's spin levels, providing a direct and calibration-free measurement. This capability is perfectly complemented by first-principles calculations, which can map the full 3D vector field distribution across a material's surface and into the adjacent vacuum. The powerful synergy lies in bridging the internal and external magnetic landscapes: the calculations predict how the intricate internal magnetic structure of magnetic materials, governed by its atomic spin ordering, manifests as a detectable stray field outside the surface.

3.8. Evaluation of non-collinear spin configurations and cross-code uncertainties

In hematite (α -Fe₂O₃), the magnetic ground state undergoes a well-known spin reorientation across the Morin transition temperature ($T_M \approx 260$ K). Below T_M , the system stabilizes in an out-of-plane collinear antiferromagnetic (OP-AFM) state with Fe³⁺ moments aligned strictly parallel and antiparallel along the rhombohedral [111] axis (hexagonal [0001] c -axis). Above T_M , the spins rotate into the basal (111) plane (IP-AFM), where the relativistic Dzyaloshinskii–Moriya interaction (DMI) induces an extremely subtle out-of-plane canting. This canting angle, typically $\psi \approx 0.05^\circ$ – 0.15° (~ 1 – 3 mrad), gives rise to a macroscopic weak ferromagnetic moment of $\sim 2 \times 10^{-3} \mu_B/\text{Fe}$.

To construct the intra-cell magnetostatic vector field distributions, the primary framework utilizes real-space scalar spin densities calculated via all-electron Gaussian and plane-wave (GAPW) functionals in CP2K, onto which sub-lattice directional quantization vectors are post-assigned. This approach provides the sub-angstrom spatial resolution required to resolve sharp electron localization around the Fe 3d shells, while circumventing the prohibitive computational overhead and well-known electronic band-gap underestimations associated with standard local-density non-collinear implementations.

The impact of the finite canting angle ψ on the local magnetic induction $\mathbf{B}(\mathbf{r})$ can be quantitatively evaluated via a first-order perturbation

to the sub-lattice magnetostatic kernel:

$$\mathbf{B}(\mathbf{r}, \psi) = \mathbf{B}^{(0)}(\mathbf{r}) + \delta \mathbf{B}_{\text{cant}}(\mathbf{r}, \psi) \quad (1)$$

where $\mathbf{B}^{(0)}(\mathbf{r})$ represents the unperturbed collinear basal-plane field (~ 1.0 – 1.5 T in the interstitial channels), and the canting-induced deviation field scales directly with the uncompensated transverse moment components:

$$\delta \mathbf{B}_{\text{cant}}(\mathbf{r}, \psi) = \frac{\mu_0}{4\pi} \sum_i \psi m_i \left(\frac{3[(\mathbf{r} - \mathbf{r}_i) \cdot \hat{\mathbf{z}}](\mathbf{r} - \mathbf{r}_i)}{|\mathbf{r} - \mathbf{r}_i|^5} - \frac{\hat{\mathbf{z}}}{|\mathbf{r} - \mathbf{r}_i|^3} \right) \quad (2)$$

Given $\psi \approx 10^{-3}$ rad, the magnitude of $|\delta \mathbf{B}_{\text{cant}}|$ is strictly bounded within 1.5–3.5 mT across the entire unit cell. This deviation accounts for less than 0.3% of the characteristic local field strength, which remains well below the experimental resolution limits and intrinsic linewidths of local probes such as zero-field μSR (~ 10 – 30 mT).

Furthermore, a comparative analysis between the CP2K scalar-density scheme and the Quantum ESPRESSO (QE) fully relativistic non-collinear scheme reveals differences in absolute intensities but remarkable consistency in spatial topologies (detailed in Supplementary Material, Section 3.7). Specifically, the local Fe magnetic moments and the corresponding most probable magnetic field calculated by QE are approximately 30% higher than those derived from the CP2K collinear IP-AFM configuration, whereas the difference in the calculated band gaps (~ 2.0 eV) remains insignificant. As discussed in Section 3.7, this intensity inconsistency originates primarily from the different computational setups: the QE non-collinear calculation utilizes a GGA functional with a pseudopotential basis, whereas the CP2K scalar spin-density calculation employs a meta-GGA functional with an all-electron basis, which provides superior accuracy. Crucially, despite this overall scaling difference in magnitude, the spatial nodal topologies, field-dependent volume fraction distributions, and high-field isosurface contours remain virtually identical between the two methodologies.

4. Conclusion

In summary, this work establishes a general framework for determining the full 3D vector magnetic field in quantum materials by

integrating first-principles calculations with a microscopic magneto-static model based on atomic-scale spin densities derived fractional Bohr magnetons without requiring a vector potential. Application to hematite (α -Fe₂O₃) enables the first sublattice-resolved visualization of the vector field and a quantitative description of its evolution across the Morin transition. Key findings include the absence of any vanishing total field in the crystal, a most probable flux density of 1.5 T (below) and 1 T (above) the transition, and the confinement of over 99% of the crystal volume to fields below 17 T in both phases.

Fundamentally, the predicted internal field distributions show consistent agreement with bulk probes—specifically, the total field statistics from μ SR and the vector components resolved by neutron scattering. Further support comes from nitrogen-vacancy center magnetometry at the surface, confirming the transferability of the predictions across spatial regimes. These results reconcile long-standing discrepancies in reported critical transition fields and explicitly delineate the extended 3D magnetostatic vector continuum from the localized on-site scalar energy couplings inferred via Mössbauer spectroscopy.

The present study thus establishes a unified, gauge-independent first-principles paradigm for computing atomic-scale magnetic fields with quantitative accuracy. While this framework has been rigorously validated for systems where microscopic magnetic moments are predominantly governed by quasi-static pure spin densities (such as the orbitally quenched high-spin Fe³⁺ in hematite), extending this paradigm to systems with significant orbital currents or dynamic spin currents will require further theoretical integration. To address these complex mechanisms, efforts to incorporate current-density functional theory (CDFT) into the present framework are currently underway. Ultimately, this work not only resolves material-specific controversies in hematite but also opens a robust pathway toward the predictive modeling of complex field distributions across a broad range of quantum magnets, spintronic devices, and magnetic-field-controllable molecular clusters at the picometer scale.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary material related to this article can be found online at <https://doi.org/10.1016/j.commatsci.2026.115184>.

Data availability

Data will be made available on request.

Step-by-step operation procedures for obtaining sub-lattice 3D vector magnetic fields (Original data) (HepData)

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Supplementary Materials for “First-Principles Determination of Sub-Lattice 3D Vector Magnetic Fields in Antiferromagnetic Hematite”

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1. Overview of molecular field theories

1.0 Quantum field framework for microscopic magnetism

1.0.1 Classical magnetostatics

In classical electromagnetism, the magnetic field has two primary and widely recognized sources: electric currents and magnetic moments arising from the intrinsic magnetization of matter. The source–field relationships for these are established via the Biot–Savart law and the formula for the magnetic dipole field, respectively. In the framework of Maxwell's equations, as illustrated in Fig.1.1, the roles of the current source and the magnetization source are subtly distinct; they appear separately in the curl equation for the magnetic field and in the auxiliary

equations describing the magnetic properties of media.

The explicit form of the magnetic dipole and the equation for the magnetic field produced by a dipole do not appear directly in Maxwell's equations. This is because Maxwell's equations focus primarily on the dynamic relationship between electric and magnetic fields. In purely magnetostatic studies where time-varying effects are absent, these elements must be introduced separately. Alternatively, they can be effectively incorporated through intermediate quantities, such as the intrinsic magnetic vector potential, thereby enabling an equivalent conversion between magnetization sources and current sources^[1].

The theory of magnetostatics for macroscopic fields has been extended into micromagnetism, where magnetization continues to be treated as a classical vector field, consistent with macroscopic approaches^[2]. For most micromagnetic problems—especially at scales above 100 nm—the continuum approximation remains valid, with deviations generally staying within a few percent. This approximation, however, fails when the characteristic size of the magnetic structure approaches interatomic distances. At such sub-nanometer or atomic scales, intrinsic material parameters like magnetization and anisotropy can no longer be described by a single set of uniform bulk values. Consequently, applying bulk properties in these regions becomes a relatively crude approximation.

For over a century, a central issue in microscopic electromagnetism has been **whether continuum field equations apply at atomic scales, given the inherent non-uniformity in magnetic-field and magnetic-moment distributions.**

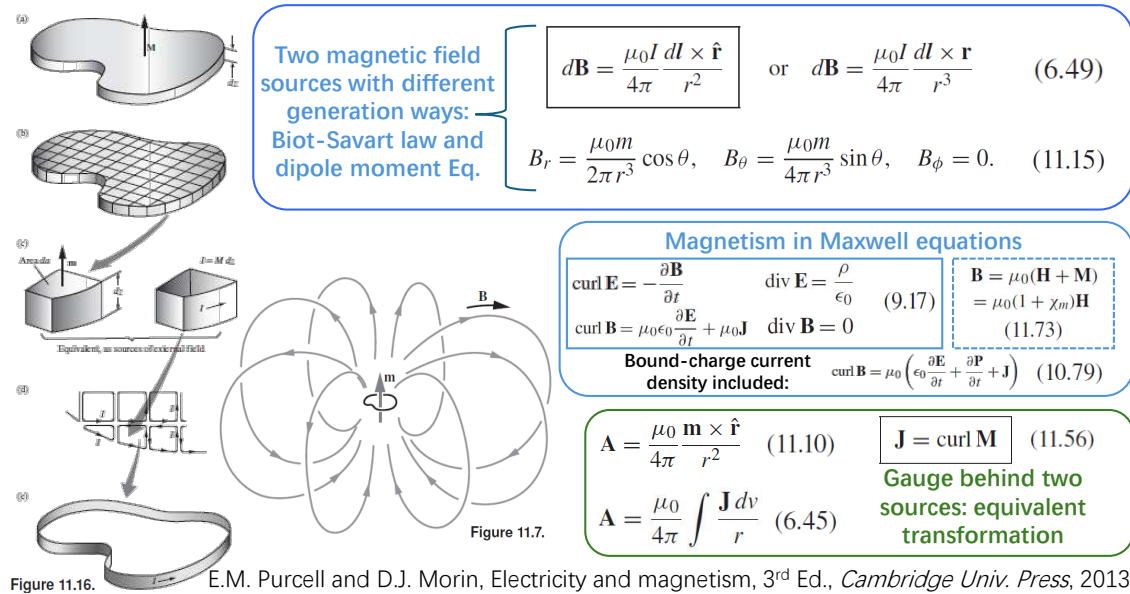


Fig.1.1 Classical theory for electromagnetism in a textbook^[1].

1.0.2 Quantum theory of electrons

At the atomic scale, the motion of matter is governed by the laws of quantum mechanics. Fig.1.2 illustrates a foundational journey in the theoretical description of electrons in matter, marking key transitions from fundamental quantum mechanics to practical computational frameworks for condensed-matter magnetism.

In 1926, Schrödinger published in succession the stationary and time-dependent wave equations for electrons. This represented a revolutionary shift from the classical description of continuous trajectories to one in which microscopic fluctuations are characterized by a probability-density related wave function^[2]. However, solving the Schrödinger equation directly for systems containing many interacting electrons proved analytically and computationally intractable—the celebrated “many-body problem.”

Nearly four decades later, a major breakthrough was achieved by Kohn and

Sham in 1965. Their formulation of density functional theory (DFT) demonstrated that the ground-state properties of an interacting electron system can be obtained by solving a set of effective single-particle Schrödinger equations^[3]. The key insight was that all quantum mechanical many-body effects—exchange and correlation—could be incorporated into a single, density-dependent potential. This replaced the intractable many-body wave function with the much simpler electron density as the central variable, thereby making realistic calculations for molecules and solids feasible.

By 1976, the theory had been promptly extended to spin-density-functional theory (SDFT)^[4]. In this formulation, the equations separate for spin-up and spin-down electrons according to their spin densities. By generalizing the charge density to a pair of spin densities and solving them separately, DFT became applicable to magnetic materials, atoms, and molecules—systems in which spin polarization plays an essential role.

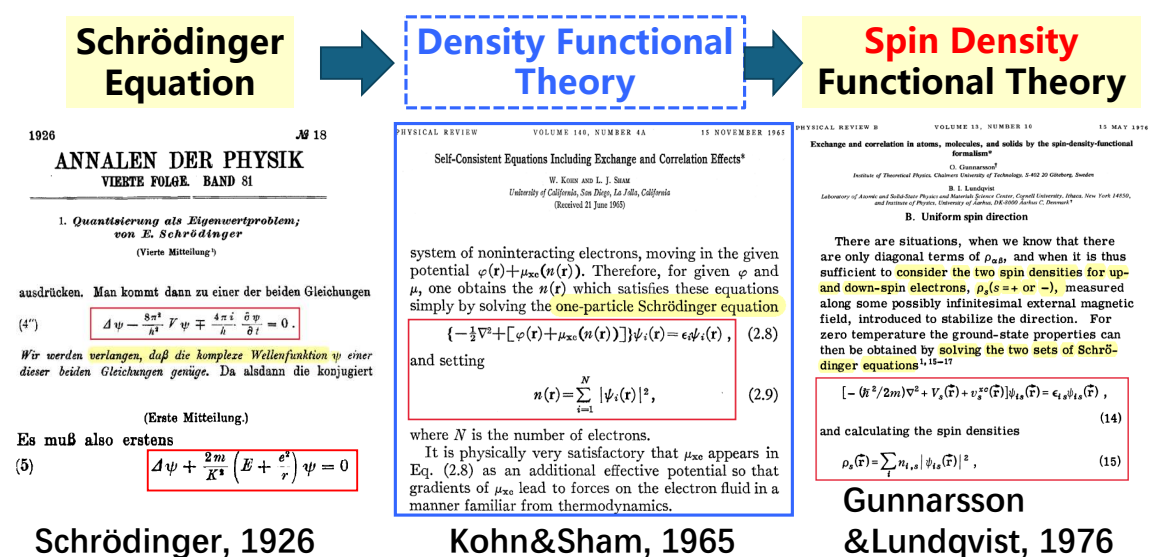


Fig.1.2 Quantum theory of electrons from Schrödinger's equation^[2] to Kohn-Sham equation^[3] and spin-density functional theory^[4].

1.0.3 Magnetic field and its sources at the atomic scale

Integrating the microscopic Schrödinger equation with the macroscopic Maxwell's equations to study electromagnetic fields, charge distributions, and their interactions at the quantum scale was once an unimaginably formidable task. Fortunately, the giants of quantum mechanics identified gauge invariance and the variational principle as key guiding concepts. Quantum mechanics incorporates both the principle of gauge invariance, which mandates that physical predictions remain unchanged under transformations of the electromagnetic potentials, and the variational principle, which states that the expected energy of any trial wavefunction is an upper bound to the true ground state energy. These principles ultimately enabled the quantization of the electromagnetic field and electron spin, transforming them from abstract ideas into the theoretical cornerstone of quantum electrodynamics. This achievement marked a crucial step in the development of quantum field theory.

Fig.1.3 traces a pivotal conceptual arc in theoretical physics: the forging of a consistent quantum theory that unifies the macroscopic description of electromagnetic fields with the microscopic behavior of electrons. The journey centers on two guiding principles: gauge invariance and the variational principle.

The cornerstone was laid by Hermann Weyl in 1918^[5], whose work on unifying gravitation and electricity introduced the profound idea of gauge invariance—the concept that certain transformations of the electromagnetic potential should leave the fundamental physics unchanged. By replacing the original real-scale

transformation with an imaginary phase transformation in 1929, this principle was later recognized as the milestone for constructing self-consistent theories of microscopic fields^[6]. The decisive step toward a quantum theory of electromagnetic field is embodied in Dirac's 1927 paper. By treating the field variables as quantum-mechanical operators satisfying commutation relations, he successfully described the emission and absorption of radiation by atoms^[7]. Crucially, this established light quanta (photons) as the excitations of the quantized field, seamlessly bridging Maxwell's macroscopic electromagnetism with Einstein's photon concept. His method inherently respected the underlying gauge structure of the classical theory.

The synthesis and rigorous formalization of these ideas in a relativistic framework is captured in the excerpt from Wolfgang Pauli's 1941 review^[8]. This paper offers profound insights and a clarifying exposition, notably on the exclusion principle and the author's work on the spin–statistics theorem within the framework of relativistic quantum field theory. It provided a critical foundation for the subsequent development of quantum field theory and renormalization.

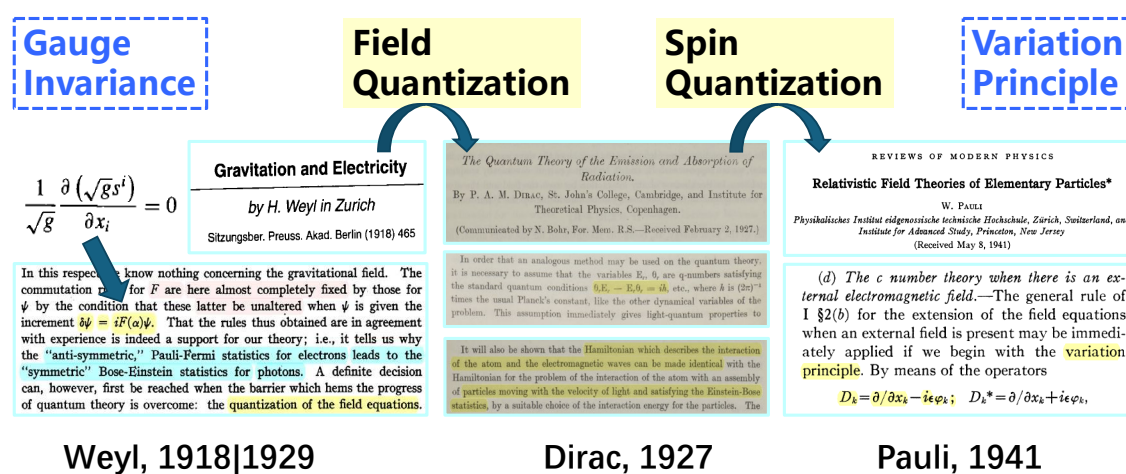


Fig.1.3 The foundational papers of quantum field theory: unifying fields and matter through gauge invariance^{[5][6]}, field quantization^{[6][7]}, and the variational principle^[8].

1.0.4 Sub-lattice magnetostatics in condensed matter

As shown in Fig.1.4, to connect the macroscopic electromagnetic field and microscopic electronic properties, which are governed by Maxwell's equations and the Schrödinger equation respectively, requires establishing the theoretical framework of quantum field theory. Gauge invariance and the variational principle serve as two crucial pillars for bridging the two. From fundamental particles to crystals, in order to accurately describe the static magnetic field distribution in sub-lattice level, it is not only necessary to transform the single-point Lagrangian into a spin density functional that accounts for full-electron and well-defined exchange-correlation effects, but also to convert the total spin quantum into a spin magnetic moment expressed as fractional Bohr magnetons via the spin density distribution. As an important frontier in condensed matter theory, the dynamic interplay between microscopic fields and matter is poised to be revealed with unprecedented precision. This advancement will illuminate fundamental processes such as nonequilibrium phase transitions, ultrafast energy transfer, and emergent collective phenomena. However, significant challenges remain in realizing this goal.

Based on high-precision basis sets and functionals for spin-density functional calculations, the key innovation of this work lies in deriving fractional Bohr magneton moments and distributed 3D vector magnetic field from the spin density (the system of equations shown in Figure 1 of the main text). This preserves the fundamental principles of gauge invariance and the variational principle, and supports the

establishment of a sub-lattice magnetostatic theory that self-consistently aligns the distributed magnetic field with the corresponding magnetic moments.

To verify the effectiveness of the method, this work systematically calculates the magnetic field distribution within the unit cell of hematite and analyzes the influence of various factors on the results, so as to open a new avenue for the design of molecular magnetic circuits.

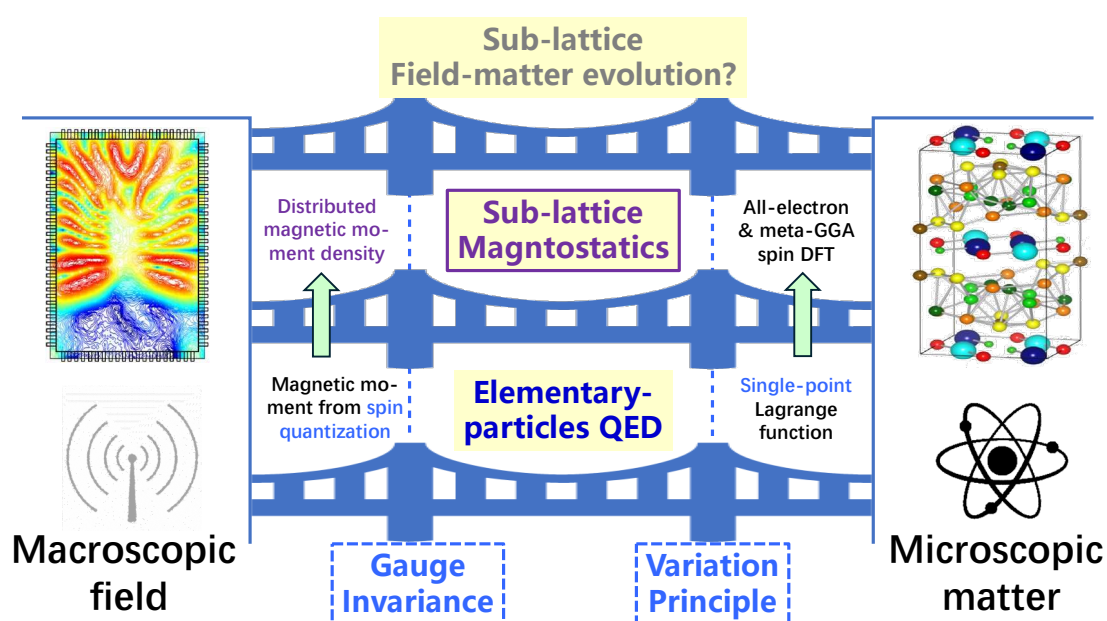


Fig.1.4 The landscape of emerging theories in quantum field theory, with gauge invariance and the variational principle as the two foundational pillars connecting macroscopic electromagnetic fields and microscopic electronic properties. Sub-pictures showing complex macroscopic field and microscopic matter are from internet^[10].

1.1 Molecular field: from phenomenology to quantum interaction

1.1.1 Semi empirical estimation

For magnetic materials with spontaneous magnetization above room temperature, based on empirical thermodynamic energy, the minimum internal

magnetic field could be estimated 450 T as shown in the following equation^[9]. It is too high to be the predominant mechanism which gives rise to the alignment of the magnetic moments in ferromagnetic materials at room temperature.

$$H = \frac{kT_{RT}}{\mu_0 \mu_B} \cong \frac{1.38 \times 10^{-23} \times 300}{1.2566 \times 10^{-6} \times 9.274 \times 10^{-24}} = 3.55 \times 10^8 (A/m) \sim 4.5 \times 10^2 (T) \quad 1.1$$

1.1.2 Weiss molecular field

As shown in Fig.1.1, Weiss proposed that there is a molecular field of electrostatic origin proportional to the magnetization (similar to "van der Waals pressure")^[2]. Along this idea, Néel extended the molecular field theory for ferrimagnetic matters using total magnetization, by superimposing antiparallel moments^[3]. The corresponding linear coefficients are equivalently associated to Currie and Néel temperature for ferromagnetic and ferrimagnetic materials, respectively.

Weiss molecular field: original idea and description in the English textbook.

P. Weiss, *J. Phys.*, **6**, 661-690(1907)

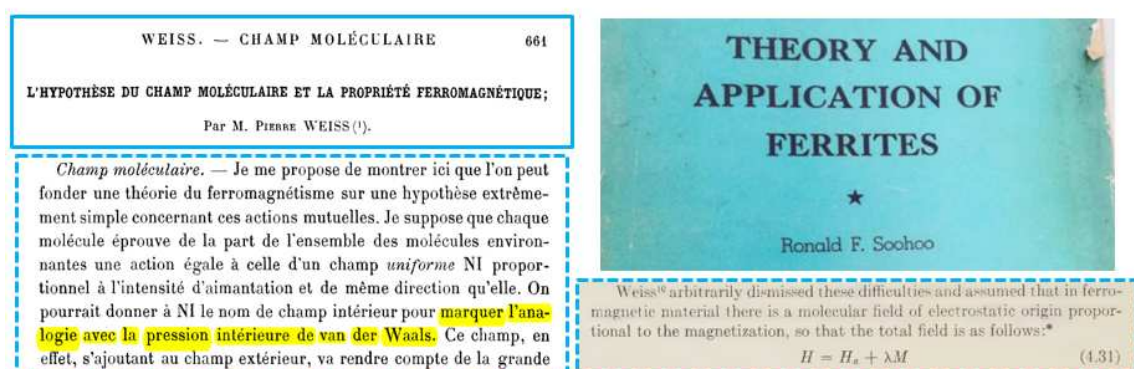


Fig.1.5 The original paper of molecular field theory^[12] and its description in a textbook^[11]

1.1.3 Heisenberg exchange interaction

As shown in Fig.1.2, Heisenberg claimed that Weiss's molecular forces are

attributed to a quantum mechanical exchange phenomenon^[4]. The magnetic interactions have been totally disregarded because they are much weaker than exchange interaction.

Heisenberg exchange interaction:

replacing molecular field to mainly contribute molecular forces

Zur Theorie des Ferromagnetismus.

Von **W. Heisenberg** in Leipzig.

Mit 1 Abbildung. (Eingegangen am 20. Mai 1928.)

Die Weiss'schen Molekularkräfte werden zurückgeführt auf ein quantenmechanisches Austauschphänomen; und zwar handelt es sich um diejenigen Austauschvorgänge die in letzter Zeit von Heitler und London mit Erfolg zur Deutung der homöopolaren Valenzkräfte herangezogen worden sind.

$$J_E = \int d\tau_1 d\tau_2 \dots d\tau_n (\psi_1^1)^2 (\psi_2^2)^2 \dots (\psi_n^n)^2 \left\{ \sum_{k>l} \frac{e^2}{r_{kl}} + \sum_{\substack{\alpha, \beta \\ \alpha > \beta}} \frac{e^2}{r_{\alpha\beta}} - \sum_{\substack{k, l \\ k \neq l}} \frac{e^2}{r_{kl}} \right\}. \quad (2)$$

Die magnetischen Wechselwirkungen lassen wir wegen ihrer Kleinheit ganz unberücksichtigt. Trotzdem werden sich vermöge der Austauschprozesse die Spinnomente sämtlicher Elektronen teils parallel, teils antiparallel stellen. Nimmt man die grundlegende Annahme des Pauli-Prinzips hinzu, daß die Eigenfunktionen des Gesamtsystems antisymmetrisch in sämtlichen Elektronen sein sollen, so gehört zu jedem

W. Heisenberg, *Z. Physik*, **49**, 619-636(1928)

Weiss's molecular forces are attributed to a quantum mechanical exchange phenomenon; These are the processes of exchange that Heitler and London have recently successfully used to interpret the homopolar valence forces.

We **disregard the magnetic interactions** completely because of their smallness. Nevertheless, the **exchange processes will be able to place the spin moments** of all electrons partly parallel, partly antiparallel.

Fig.1.6 The original paper of Heisenberg exchange interaction^[4]

1.1.4 Single-site molecular field models

A general spin Hamiltonian describing the magnetic properties of a material with localized electrons can be expressed as the following equation^[5].

$$\hat{H}_{\text{spin}} = \sum_{i<j} J_{ij} \hat{S}_i \cdot \hat{S}_j + \sum_{i<j} \vec{D}_{ij} \cdot (\hat{S}_i \times \hat{S}_j) + \sum_i A_i S_{iz}^2 + \sum_{i<j} K_{ij} (\hat{S}_i \cdot \hat{S}_j)^2 \quad 1.2$$

where the first term represents the symmetric exchange interaction, the second term the antisymmetric exchange interaction, the third term the single-site anisotropy, and the last term the biquadratic interaction. The symmetric exchange interaction is usually known as the Heisenberg exchange interaction, and the antisymmetric exchange interaction as the DM interaction.

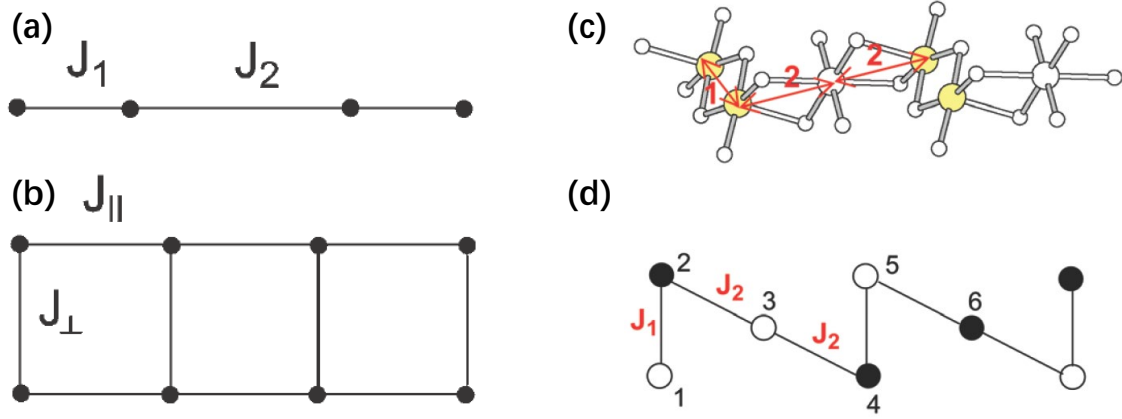


Fig.1.7 Single-site exchange interactions from linear (a) and simple square lattice (b) to 3D network (c) and complex topology (d) ^[5]

Besides spin-spin exchange interactions among electrons, starting with the Dirac theory for the electron, the Fermi contact form of the Hamiltonian for the interaction of a nucleus with a single electron could be written as ^[6].

$$H = -gg_I\mu_B\mu_N\{(8\pi/3)\delta(\mathbf{r})\mathbf{I} \cdot \mathbf{S} + r^{-3}\mathbf{I} \cdot (\mathbf{L} - \mathbf{S}) + 3r^{-5}(\mathbf{I} \cdot \mathbf{r})(\mathbf{S} \cdot \mathbf{r})\} \quad 1.3$$

where $\mathbf{L}$, $\mathbf{S}$, and $\mathbf{I}$ represent, electron orbital, electron spin, and nuclear-spin angular momentum operators, respectively; μ_B and μ_N are the Bohr and nuclear magnetons; and g and g_I are the electronic and nuclear spectroscopic splitting factors. The delta-function term, which is nonzero only for s electrons, for which case the last two terms for dipolar interaction, are zero.

Hamiltonian of the equation may be interpreted as arising from the interaction of an effective magnetic field, B_e , with the nuclear-spin magnetic moment. Therefore, the hyperfine effective magnetic field is obtained after dividing by the nuclear magneton.

$$\mathbf{B}_e = (8\pi/3)g\mu_B\mathbf{S}|\psi(0)|^2 \quad 1.4$$

where $|\psi(0)|^2$ is the s electron's density at the nucleus.

Single-site models are rather useful for total energy calculation and uniform property prediction, which have a clear physical picture to simplify magnetic interactions, as shown in Fig.1.3. Nevertheless, molecular field understanding is roughly based on a phenomenological description, and detailed local distribution of magnetic fields around magnetic ions remains unsolved.

1.1.5 Spin current density and vector potential derived field

As mentioned previously, Weiss molecular field and single-site molecular field are both based on phenomenological theory. Although ~ 55 T strong magnetic field in irons can be interpreted by Mössbauer experiments^[6], Heisenberg suggested that the magnetic interactions are neglectable because they are much weaker than exchange interaction^[4].

In contrast to phenomenological single-site models, distributed molecular field has been calculated on the basis of density function theory and spin current density with the vector potential used^{[7][8]}. As shown in Fig.1.4, the spin current density and vector potential derived molecular field in bcc Fe is so strong that the Weiss molecular field theory would seem to be correct. However, the distribution pattern and field values in two articles are too inconsistent with each other to be right simultaneously. Since there are infinitely many ways to choose the phase of the vector potential, known as the gauge choice, different field predictions may be generated when the local spin current density are calculated using an unequal gauge as that in DFT calculation. To solve this problem, one need to develop integrated

DFT programs particularly for molecular field simulation, or calculate field using gauge invariant magnetic moment method after spin density is produced by a normal DFT. The current author mainly treats the later in this work.

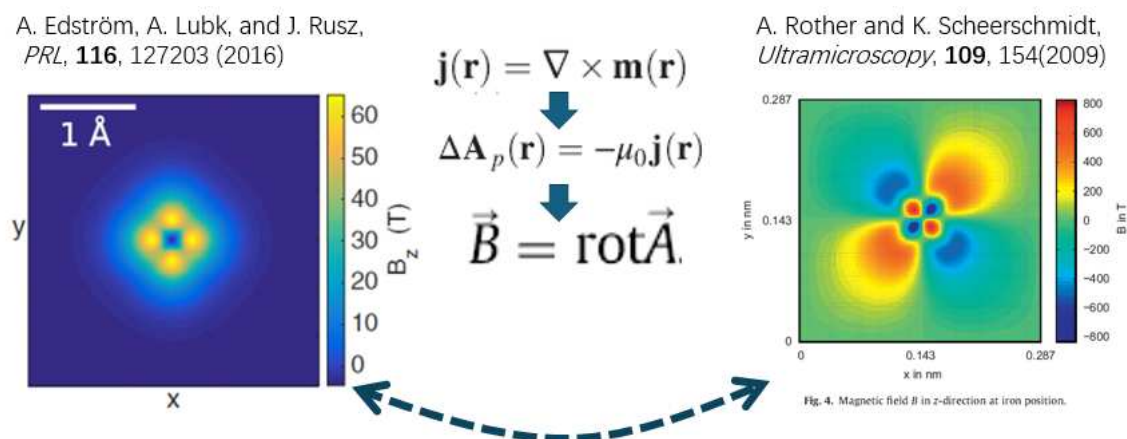


Fig.1.8 Comparison of magnetic field B_z distribution at iron position within the bcc unit cell^{[7][8]}. Left and right pictures are both derived from spin current density and vector potential method as shown in the middle, but quantitative values and patterns seem inconsistent.

When reexamining the molecular field which had been disregarded in Heisenberg's quantum exchange model, we should straightly answer whether the magnetic field inside the magnetic molecules is weak or how strong it is. In the main text, the current author takes two steps in sequence, from first principle calculating magnetic sources of moment, to generation of magnetic field from microscopic dipole moments without vector potential. The first step is widely used spin DFT, while the other step is independently developed in this work. The theoretic basis of these two steps is introduced in the next two sections.

1.2 First principles of calculating microscopic magnetic properties in solid

1.2.1 Density-functional theory derived spin density^[9]

The electronic many-body problem in solid is defined to find the solutions of the static Schrödinger equation for a system of N interacting electrons,

$$\hat{H}\Psi_j(\mathbf{x}_1, \dots, \mathbf{x}_N) = E_j\Psi_j(\mathbf{x}_1, \dots, \mathbf{x}_N) \quad 1.5$$

where the total Hamiltonian includes kinetic energy, potential energy and electron–electron interaction. Here, the Born-Oppenheimer approximation is already employed to allow the electronic part to be solved with the nuclear positions as parameters. Due to the substantial disparity between the electronic and nuclear masses, the two sets of degrees of freedom have essentially been decoupled.

Calculating the full Ψ to obtain wavefunction based properties seems like a terribly wasteful approach if N is large. To relieve us of this duty, DFT can help to obtain all properties of a many-body system exactly, without having to solve the many-body Schrödinger equation. Hohenberg–Kohn theorem ensures that interacting N -electron system with a given particle–particle interaction there exists a one-to-one correspondence between the external potential and the ground-state density $n_0(r)$.

In Kohn-Sham (KS) DFT, we have the same one-electron density as the true system by superimposing some key interaction items.

$$\begin{aligned}
E_{v_0}[n] &= T[n] + W[n] + \int d^3r n(\mathbf{r})v_0(\mathbf{r}) \\
&= T_s[n] + \int d^3r n(\mathbf{r})v_0(\mathbf{r}) + \frac{1}{2} \int d^3r \int d^3r' \frac{n(\mathbf{r})n(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} + E_{xc}[n]
\end{aligned} \tag{1.6}$$

In this way, the calculation time is much shorter than that of a traditional direct approach, and so much larger systems (several hundred atoms) can be routinely handled. In principle, this approach produces the exact ground-state energy and density, but in practice, one must approximate a small (but vital) contribution, called the exchange correlation (XC) energy. The quality of the results depends on the quality of this approximation. Much of modern DFT research is devoted to developing such approximations, usually termed XC functionals, since they are extremely important for properties such as the binding energy of matter.

In practice, the Kohn–Sham equations are almost always implemented in their spin-resolved form. For an N-system with $N = N_{\uparrow} + N_{\downarrow}$, the total ground-state density becomes a sum of spin-up and spin-down densities,

$$n_0(\mathbf{r}) = n_{0\uparrow}(\mathbf{r}) + n_{0\downarrow}(\mathbf{r}) = \sum_{\sigma=\uparrow,\downarrow} \sum_{j=1}^{N_{\sigma}} |\varphi_{j\sigma}(\mathbf{r})|^2 \tag{1.7}$$

and the Kohn–Sham spin orbitals follow from

$$\left(-\frac{\nabla^2}{2} + v_{s\sigma}[n_{\uparrow}, n_{\downarrow}](\mathbf{r})\right) \varphi_{j\sigma}(\mathbf{r}) = \varepsilon_{j\sigma} \varphi_{j\sigma}(\mathbf{r}). \tag{1.8}$$

The Kohn–Sham effective potential is given by

$$v_{s\sigma}[n_{\uparrow}, n_{\downarrow}](\mathbf{r}) = v_{\sigma}(\mathbf{r}) + \int d^3r' \frac{n(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} + v_{xc\sigma}[n_{\uparrow}, n_{\downarrow}](\mathbf{r}), \tag{1.9}$$

where the XC energy and potential are defined as functionals of the individual spin densities

$$v_{xc\sigma}[n_{\uparrow}, n_{\downarrow}](\mathbf{r}) = \frac{\delta E_{xc}[n_{\uparrow}, n_{\downarrow}]}{\delta n_{\sigma}(\mathbf{r})}. \tag{1.10}$$

The spin-resolved Kohn–Sham formalism is more general than the spin-independent version, since it includes spin-dependent external potentials $v_{\sigma}(\mathbf{r})$. In

most applications, however, the external potential is simply the Coulomb potential of the nuclei, and under these circumstances the spin-independent and the spin-dependent Kohn–Sham formalism is in principle equivalent. In practice, the functional dependence of E_{xc} on the spin-up and spin-down densities provides more flexibility and is better suited for the construction of approximations.

The choice of functional strongly depends on the chemical system at hand. Due to the diversity of bonding situations in inorganic chemistry, ranging from covalently bonded isolated molecules to ionic crystals and metal clusters, a uniformly and usefully accurate approximate DFT description for all these systems is not yet available. The meta-GGA functional TPSS performs quite similarly to PBE. Hybrid functionals predict bonds to be slightly too long, but generally in very good agreement with experiment. Complexes of 3d elements are better described by GGA/meta-GGA functionals while hybrid functionals yielded somewhat smaller deviations from the experiment for 4d and 5d element compounds.

Generalized gradient approximations are a class of xc functionals which have the following general structure:

$$E_{xc}^{GGA} = \int d^3r e_{xc}^{GGA}(n_{\uparrow}(\mathbf{r}), n_{\downarrow}(\mathbf{r}), \nabla n_{\uparrow}(\mathbf{r}), \nabla n_{\downarrow}(\mathbf{r})). \quad 1.11$$

To achieve more flexibility, meta-GGAs include additional elements:

$$E_{xc}^{MGGA} = \int d^3r e_{xc}^{MGGA}(n_{\uparrow}, n_{\downarrow}, \nabla n_{\uparrow}, \nabla n_{\downarrow}, \nabla^2 n_{\uparrow}, \nabla^2 n_{\downarrow}, \tau_{\uparrow}, \tau_{\downarrow}). \quad 1.12$$

In addition to the Laplacians of the spin densities, meta-GGAs also include the Kohn–Sham orbital kinetic-energy densities. Unlike some GGAs that rely heavily on empirical tuning, many meta-GGAs are parameterized primarily using ab initio data,

aiming for a more systematic approach. As a result, meta-GGAs often give better predictions for the band gaps of semiconductors and other materials compared to GGAs. However, meta-GGAs require more computational resources than simpler GGAs, which can lead to slower convergence with respect to basis set size and grid fineness in numerical calculations.

1.2.2 Magnetic field related properties^[10]

The situation is complicated when investigating microscopic magnetic fields. An electric field interacts directly with the charged particles (electrons and nuclei) and adds a potential energy term to the Hamiltonian operator. A magnetic field, however, interacts with the magnetic moments generated by the movement of the charged particles, that is a magnetic perturbation changes the kinetic energy operator in the Hamiltonian. The generalized momentum operator is defined by $\pi = p - qA$, where q is the charge and A is the vector potential associated with the magnetic field H , with the latter being given as the curl of the vector potential:

$$\mu_0 H = \nabla \times A . \quad 1.13$$

Only the kinetic energy of the electrons is considered within the Born–Oppenheimer approximation and the generalized momentum becomes ($q = -1$) :

$$\pi = p + A . \quad 1.14$$

The vector potential is not uniquely defined since the gradient of any scalar function may be added (the curl of its gradient is always zero). For an external magnetic field, it is conventional to write:

$$\mathbf{A}_{ext}(\mathbf{r}) = \mu_0 \mathbf{H}_{ext} \times (\mathbf{r} - \mathbf{R}_G) . \quad 1.15$$

Here $\mathbf{R}_G$ is referred to as the gauge origin, that is the center of the vector potential. One may verify by explicit calculation that the curl of $\mathbf{A}_{ext}$ in Equation then gives $\mathbf{H}_{ext}$.

The spin associated with an electron acts as a magnetic dipole, giving rise to a vector potential $\mathbf{A}_e$ and an associated magnetic field:

$$\mathbf{A}_e(\mathbf{r}) = -\frac{g_e \mu_B}{c^2} \frac{\mathbf{s}_i \times (\mathbf{r} - \mathbf{r}_i)}{|\mathbf{r} - \mathbf{r}_i|^3} \quad 1.16$$

$$\mu_0 \mathbf{H}_e(\mathbf{r}) = \frac{g_e \mu_B}{c^2} \left[\frac{\mathbf{s}_i}{|\mathbf{r} - \mathbf{r}_i|^3} - \frac{(\mathbf{r} - \mathbf{r}_i)((\mathbf{r} - \mathbf{r}_i) \cdot \mathbf{s}_i)}{|\mathbf{r} - \mathbf{r}_i|^5} - \frac{8\pi}{3} \mathbf{s}_i \delta(\mathbf{r} - \mathbf{r}_i) \right] . \quad 1.17$$

The $\mathbf{H}_e$ expression similarly determines the magnetic field at position $\mathbf{r}$ due to an electron at position $\mathbf{r}_i$.

The electron spin related interactions can be described by two-electron spin-dipolar, two-electron Fermi contact, and two-electron part of the spin-orbital.

$$\mathbb{H}_{ee}^{SD} = \mathbf{s}_i \cdot \mathbf{P}_{ee}^{SD} \cdot \mathbf{s}_j = \frac{g_e^2 \mu_B^2}{2c^2} \mathbf{s}_i \cdot \frac{\mathbf{r}_{ij}^t \mathbf{r}_{ij} \mathbf{1} - 3 \mathbf{r}_{ij} \mathbf{r}_{ij}^t}{r_{ij}^5} \cdot \mathbf{s}_j \quad 1.18$$

$$\mathbb{H}_{ee}^{FC} = \mathbf{s}_i \cdot \mathbf{P}_{ee}^{FC} \cdot \mathbf{s}_j = -\frac{4\pi g_e^2 \mu_B^2}{3c^2} \delta(\mathbf{r}_{ij}) (\mathbf{s}_i \cdot \mathbf{s}_j) \quad 1.19$$

$$\mathbb{H}_{ee}^{SO} = \mathbf{s}_i \cdot \mathbf{P}_{ee}^{SO} = -\frac{g_e \mu_B}{2c^2} \cdot \mathbf{s}_i \cdot \frac{\mathbf{r}_{ij} \times \mathbf{p}_i + 2 \mathbf{r}_{ij} \times \mathbf{p}_j}{r_{ij}^3} \quad 1.20$$

When both electron spin and an external magnetic field are considered, there is a mixed $\mathbf{A}_{ext} \cdot \mathbf{A}_e$ term:

$$\mathbb{H}_{ee}^{DS} = \mu_0 \mathbf{H}_{ext} \cdot \mathbf{P}_{ee}^{DS} \cdot \mathbf{s}_i = -\frac{g_e \mu_B}{2c^2} \mu_0 \mathbf{H}_{ext} \cdot \frac{\mathbf{r}_{iG}^t \mathbf{r}_{iG} \mathbf{1} - \mathbf{r}_{iG} \mathbf{r}_{iG}^t}{r_{iG}^3} \cdot \mathbf{s}_i . \quad 1.21$$

This electronic diamagnetic shielding operator contributes to the $\mathbf{g}$ tensor.

1.2.3 Gauge including atomic orbitals^[10]

In a standard perturbation formalism, the spin-spin coupling constant contains

four contributions arising from the diamagnetic spin-orbit operator and combinations of the paramagnetic spin-orbit, Fermi-contact, and spin-dipole operators. Due to the interaction between an unpaired electron spin and a nuclear spin, the hyperfine coupling constant should also be considered. The Fermi-contact operator is a δ -operator measuring the electron density at the nuclear position, and it is clear that standard basis sets are inadequate for representing such a point property and that a number of s-functions with (very) large exponents are required. The incorrect (zero) slope at the nucleus for GTOs could potentially also be a problem, but in practice it has been found not to be the case. The paramagnetic spin-orbit operator is also involved in the chemical shielding constant and, as for spin-spin coupling, also requires an additional tight p-function. The spin-dipole operator is found to require tight p-, d-, and f-functions. Because spin-spin coupling constants are very sensitive to the quality of the wave function near the nuclei, it is difficult to contract the basis sets without destroying the inherent accuracy.

The natural choice of basis functions for (non-hybrid) DFT calculations on periodic systems are periodic plane waves. This basis can be systematically improved by adding functions with ever shorter wavelengths (thus higher energy), up to a cut-off energy. Magnetic properties derived from an exact wave function are independent of the gauge origin, but this is not necessarily the case for approximate wave functions. Having a property depending on the choice of origin is clearly undesirable, but this can be remedied by defining so-called London or Gauge

Including Atomic Orbitals (GIAO). By including a complex factor in each basis function, it can be shown that the magnetic property becomes independent of the gauge origin, although the wave function itself now depends on the gauge origin.

The gauge origin for an external magnetic field is arbitrary, while the nuclear position is the natural gauge origin for nuclear magnetic moments. The chemical shielding constant is a second-order tensor property, where the isotropic part defined as one-third of the trace, is the observable quantity in solution phase spectra. While standard energy-optimized basis sets often provide quite reasonable values for shielding constants, it is possible to reduce the basis set error by employing specific optimized basis sets. One widely used basis set for designing specific for magnetic properties was DFT energy-optimized pcseg-n, where a detailed analysis indicated that including a single tight p-function and slightly relaxing the contraction significantly reduces the basis set error. Although optimized at the DFT level, they have been shown to also perform better than other alternatives at the MP2 level and are available for all elements up to Kr and in qualities up to quintuple zeta.

1.3 Theoretical derivation of 3D vector magnetic fields

1.3.1 Magnetic moment from spin density

DFT calculated spin density indicates a fractional number of electrons per unit volume with a net polarization. Therefore, magnetostatic dipole moment $\mathbf{m}$ of spins in each volume element V_0 is then considered directly from spin density, associated with a magnetization direction (higher magnetic moments are rarely encountered

and are not considered in the current paper).

$$\mathbf{m}(\mathbf{r}) = V_0 \mathbf{M}_{\text{int}}(\mathbf{r}) = V_0 \hat{\mathbf{M}} \mu_B \sigma(\mathbf{r}) \quad 1.22$$

Here the internal magnetization $\mathbf{M}_{\text{int}}$, defined as the magnetic moment per unit volume, is proportional to the spin density, with coefficient of unit volume and Bohr magneton. The corresponding unit vector $\hat{\mathbf{M}}$ need presetting or post optimization to describe the polarized direction of the spins.

The formula 1.22 corresponds to the formula (a) of Fig.1 in the main text.

1.3.2 Microscopic current density induced magnetic moment^[11]

A dynamic system of charges with charge density $\rho(\mathbf{r})$ gives rise to a current density:

$$\mathbf{J}(\mathbf{r}) = \rho(\mathbf{r}) \mathbf{v}(\mathbf{r}) \quad 1.23$$

where $\mathbf{v}(\mathbf{r})$ is the velocity distribution. The magnetic moment due to this current density is given as

$$\mathbf{m} = \frac{1}{2} \int_V (\mathbf{r} - \mathbf{R}_{Go}) \times \mathbf{J}(\mathbf{r}) d\mathbf{r} = \frac{1}{2} \int_V \rho(\mathbf{r}) (\mathbf{r} - \mathbf{R}_{Go}) \times \mathbf{v}(\mathbf{r}) d\mathbf{r} . \quad 1.24$$

The magnetic moments can be shown to be independent of the gauge origin $\mathbf{R}_{Go}$ as a direct consequence of the absence of magnetic monopole moments.

In the main text, only the spin moment is considered the sole source of the internal magnetic field generating the 3D vector magnetic fields in hematite. This simplification holds because contributions from orbital-induced current densities and itinerant electrons are negligible. Hematite, an insulating crystalline material with half-filled 3d orbitals, exhibits quenched orbital angular momentum, precluding

significant orbital or electronic contributions. However, in materials containing free charge carriers or possessing non-zero orbital angular momentum, the current density-induced magnetic moment must be explicitly accounted for when calculating internal magnetic moments or fields.

1.3.3 Calculate magnetic field from magnetic moments

The classical expression for the magnetic field generated by an element of magnetic dipole moment is

$$\mathbf{h}(\mathbf{r}) = \frac{3(\mathbf{m} \cdot \hat{\mathbf{r}})\hat{\mathbf{r}} - \mathbf{m}}{4\pi r^3}, r = \max(|\mathbf{r}|, R_c) . \quad 1.25$$

Here $\hat{\mathbf{r}}$ is the unit vector pointing from the moment towards the point of calculating the field, and R_c is a cut-off distance to avoid calculation singularity by dealing with a collection of the moment with a defined spatial extent. Practically, the R_c is chosen as comparable to element size of magnetic moments. Although originally derived for macroscopic magnetic sources, this approach remains valid in the field of quantum molecular magnetism^[12].

Then the total internal magnetic field produced by adding up the contributions from all the individual magnetic dipoles in the system.

$$\mathbf{H}_{\text{int}}(\mathbf{r}) = \sum \mathbf{h}(\mathbf{r}) \quad 1.26$$

Therefore, the internal magnetic flux density is given by

$$\mathbf{B}_{\text{int}}(\mathbf{r})/\mu_0 = \mathbf{M}_{\text{int}}(\mathbf{r}) + \mathbf{M}_{\text{int}}(\mathbf{r}) \quad 1.27$$

The total magnetic field is obtained by combining the intrinsic internal field with the externally applied field and the macroscopic demagnetizing contribution:

$$\mathbf{B} = \mathbf{B}_{\text{int}}(\mathbf{r}) + \mathbf{B}_{\text{ext}} - \mu_0 \mathcal{N} \mathbf{M}_{\text{mac}} \quad 1.28$$

Here, $\mathbf{B}_{\text{int}}(\mathbf{r})$ is the position-dependent internal field generated by the material's own magnetic structure, $\mathbf{B}_{\text{ext}}$ is the externally applied magnetic field. The term $\mu_0 \mathcal{N} \mathbf{M}_{\text{mac}}$ represents the demagnetizing field arising from the macroscopic magnetization $\mathbf{M}_{\text{mac}}$, where μ_0 is the vacuum permeability and $\mathcal{N}$ is the dimensionless demagnetization tensor determined by the sample geometry. Thus, the total field probed locally, for example by a muon, is the sum of the local internal field, the external field, and a shape-dependent demagnetizing correction.

The formula 1.25, 1.27 and 1.28 correspond to formula (b), (c) and (d) of Fig.1 in the main text, respectively.

1.3.4 Energy evaluation

Based on the magnetization, magnetic field, and magnetic flux density, thermodynamic energy can be obtained

$$E_{MH} = -\frac{\mu_0 V_{\text{cell}}}{2N} \sum \mathbf{M} \cdot \mathbf{H} \quad 1.29$$

$$E_{BH} = -\frac{V_{\text{cell}}}{2N} \sum \mathbf{B} \cdot \mathbf{H} = -\frac{\mu_0 V_{\text{cell}}}{2N} \sum (\mathbf{M} \cdot \mathbf{H} + \mathbf{H} \cdot \mathbf{H}) = E_{MH} + E_{HH} \quad 1.30$$

$$E_{MB} = -\frac{V_{\text{cell}}}{2N} \sum \mathbf{M} \cdot \mathbf{B} = -\frac{\mu_0 V_{\text{cell}}}{2N} \sum (\mathbf{M} \cdot \mathbf{H} + \mathbf{M} \cdot \mathbf{M}) = E_{MH} + E_{MM} \quad 1.31$$

Here the $\mathbf{B} \cdot \mathbf{H}$ energy term captures the total magnetic energy of a unit cell, including the energy of the magnetic field in free space, while the $\mathbf{M} \cdot \mathbf{H}$ energy term isolates the contribution from the material itself. Crucially, it is necessary to distinguish the physical distinctness between the $\mathbf{M} \cdot \mathbf{H}$ and $\mathbf{M} \cdot \mathbf{M}$ terms. The $\mathbf{M} \cdot \mathbf{H}$ term represents the non-local spatial correlation between localized moments and their surrounding

magnetic environment, making it highly sensitive to relative spin configurations and the primary thermodynamic driver for providing feedback to optimize spin orientation, as detailed in equation 1.22. In contrast, the $\mathbf{M} \cdot \mathbf{M}$ term captures the purely local self-energy. While less sensitive to relative spin arrangements, $\mathbf{M} \cdot \mathbf{M}$ plays a pivotal role when mapping to single-ion anisotropy (SIA)^[13], which plays a crucial role in understanding the magnetocrystalline anisotropy.

Furthermore, evaluating $\mathbf{M} \cdot \mathbf{H}$ via real-space integration over the continuous spin density represents a fundamental electrodynamic upgrade to the conventional magnetic dipole-dipole (MDD) interaction models discussed in standard energy-mapping analyses^[13]. While traditional MDD evaluations rely on point-dipole approximations that fundamentally obscure intense sub-lattice static magnetic gradients, continuous $\mathbf{M} \cdot \mathbf{H}$ formulation rigorously captures both the long-range macroscopic dipolar coupling and the extreme short-range near-field magnetic energy at the atomic scale. By combining these local and non-local contributions, one obtains the total material-specific energy $\mathbf{M} \cdot \mathbf{B}$, as described in equation 1.31, which comprehensively governs the magnetocrystalline anisotropy.

The formula 1.31 corresponds to formula (e) of Fig.1 in the main text.

1.4 Motivation and highlights for hematite

1.4.1 Hematite: an ancient yet cutting-edge quantum material

Hematite, also known as iron ore or $\alpha\text{-Fe}_2\text{O}_3$, is a mineral that has been used by humans for thousands of years. From ancient civilizations to modern society,

hematite has played a crucial role in the development of our world^[14]. Recent research has shown that hematite is not just an ordinary mineral, but also a cutting-edge quantum material with fascinating properties^{[15][16]}.

Antiferromagnets, with opposite magnetic moments, have zero net magnetic moment, making them immune to external fields and ideal for spintronic devices. Their rapid spin dynamics, unmatched by ferromagnets, are crucial for high-speed applications. Additionally, antiferromagnets enable long-range spin transport by eliminating scattering centers, a limitation in ferromagnetic materials. These advantages highlight the potential of antiferromagnets in advancing spintronics and related fields^[15].

Two of the key advantages of using hematite as an antiferromagnetic material are its high biocompatibility and high Néel temperature (~ 950 K). To tailor hematite for a wide range of spintronic applications at the nanometer-scale or even the picometer-scale, it is imperative to conduct advanced micromagnetic research in the atomic scale. This is essential for comprehensively understanding the mechanisms underlying its distinctive quantum properties. It also enables the development of innovative methodologies to harness these attributes effectively.

1.4.2 Internal magnetic fields remain puzzling

Quantitatively characterizing the distribution of the magnetic field within hematite at the atomic scale presents a complex challenge. As illustrated in Figure 1.5a, atomic-resolution differential phase contrast scanning transmission electron

microscopy has successfully visualized the real-space magnetic field distribution in hematite in a projection manner^[16]. However, this technique does not provide information on the strength of the magnetic field.

Various experimental methods have been employed to estimate the magnetic field strength in hematite at the atomic scale, yielding disparate results:

- 1) Mössbauer Studies: These studies have estimated hyperfine fields at the iron nuclei in hematite to be above 50 T at low temperatures and in atmospheric conditions at room temperature^[17].
- 2) Polarized-Neutron Studies: The field-dependent reflection curve obtained from these studies indicates magnetic fields in the order of 0.1 T^[18].
- 3) Muon Spin Rotation Experiments: These experiments have reported magnetic fields ranging from 0.6 to 1.6 T^[19].

The significant discrepancies in the magnetic field strengths obtained through these various detection methods have not been fully understood. This highlights the need to reconcile these differences and to establish a comprehensive understanding of the magnetic field distribution within hematite at the atomic scale.

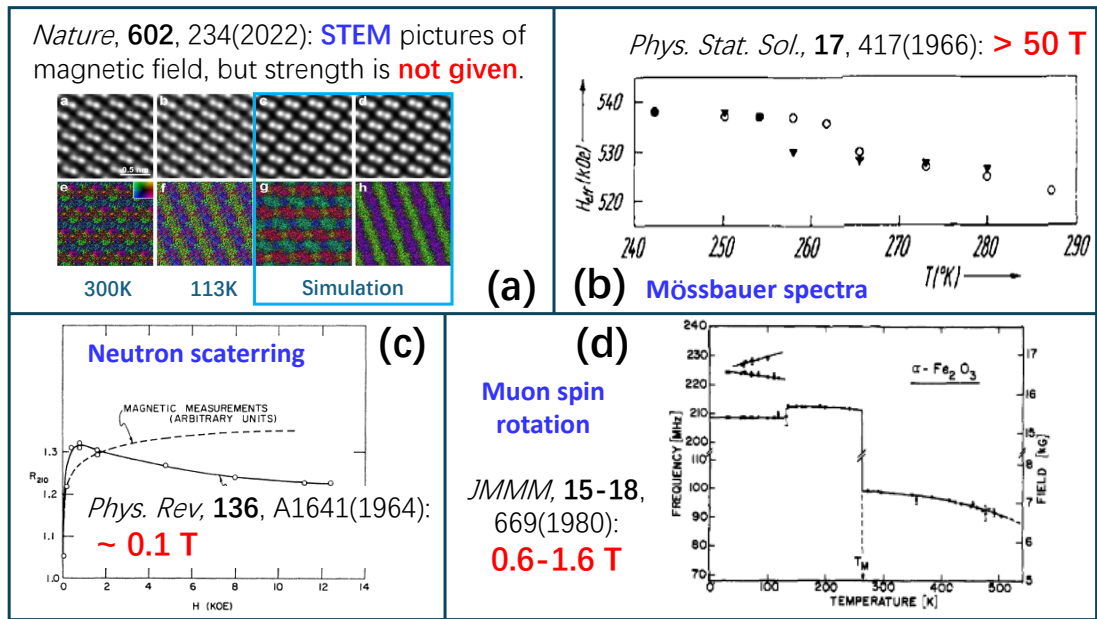


Fig.1.9 Comparison of internal magnetic fields for the hematite: (a) unknown from scanning TEM^[26], (b) more than 50 T from Mössbauer studies^[27], (c) 0.1 T from neutron scattering^[28], and (d) 0.6–1.6 T from muon spin rotation^[29].

1.4.3 Key contributions of this work

This work establishes a reasonable pathway to calculate the full 3D vector magnetic field distribution from first principles. It is essential to develop a theoretical framework capable of accurately predicting magnetic field strengths comparable to those obtained from direct experimental measurements, such as muon spin rotation and neutron scattering. This unified approach integrates theoretical predictions with experimental observations, providing a comprehensive understanding of magnetic field distribution within quantum materials.

In the main text, the current author employs a two-step approach: first, calculating the magnetic moments from first principles, and second, generating the magnetic field from microscopic dipole moments of fractional Bohr magnetons

without requiring a vector potential. The first step utilizes the widely applied spin density functional theory (DFT), while the second step is independently developed in this work. By combining first-principles calculations with field calculations based on spin density and associated magnetic dipole moments at the atomic scale, this study obtains a detailed 3D vector magnetic field distribution of hematite, revealing the following key insights.

- 1) For the first time, sub-lattice 3D vector magnetic fields are clearly visualized for hematite.
- 2) Using the proposed method, the internal magnetic fields of hematite are quantitatively analyzed before and after the Morin transition, demonstrating excellent agreement with experimental results from muon spin resonance and neutron scattering. This highlights the effectiveness of the approach in unifying different experimental techniques and resolving the discrepancy of over two orders of magnitude in hematite's internal magnetic field.
- 3) Some unexpected new findings about hematite include: No regions of zero magnetic field exist within the crystal, although the three components of the magnetic field exhibit a finite zero-field volume. The most likely magnetic flux densities above and below the Morin temperature are determined to be 1 T and 1.5 T, respectively, with less than 1% of the cell volume exceeding 17 T in both cases.

Key novel contributions of this work have been illustrated in an abstract graphic in Fig.1.10, helping readers comprehend its main meaning in a short amount of time.

Hematite: pathway to picometer spintronics

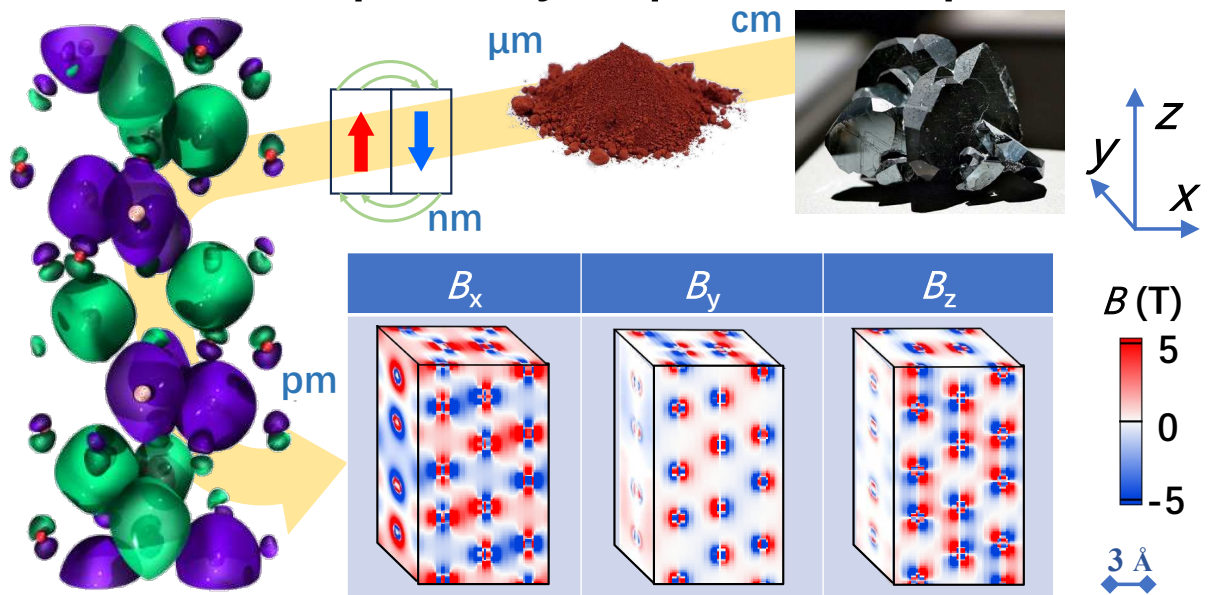


Fig.1.10 The graphical abstract for this work spans from the centimeter to the picometer scale, capturing increasingly rich quantum phenomena.

2. Operational procedures

This section introduces step-by-step operational procedures for obtaining 3D vector magnetic fields within a material, particularly hematite, and then visualizing those fields. Corresponding to the introduction in this section, on the one hand, the supplementary materials include a presentation .PPTX file with video demonstrations, and on the other hand, the input files for each step are included in the compressed files of the supplementary materials.

2.0 Preparing data for DFT calculation

The setup of the input data for Density Functional Theory (DFT) calculations performed in CP2K is first described, which forms the foundation for determining

magnetic properties for crystalline quantum materials.

2.0.1 Get a CIF file

To get crystallography CIF (Crystallographic Information File) files, several methods can be used. As shown in Fig.2.1, available structures are downloaded from crystallography databases; similar structures to known .cif files are obtained through simple modifications in a text editor; while completely unknown structures may be inferred by means such as X-ray diffraction tools.

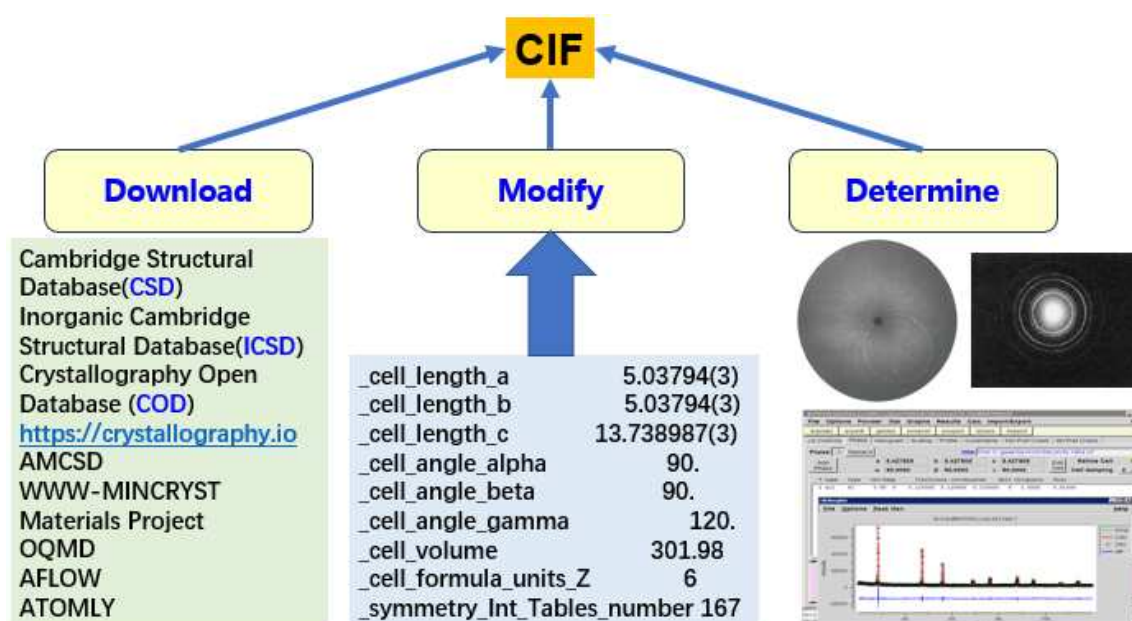


Fig.2.1 Several methods of obtaining crystallography CIF files

2.0.2 Prepare basis set like pcseg-2 for magnetism

Preparing a basis set like pcseg-2 is straightforward from the Basis Set Exchange (BSE) website (<https://www.basissetexchange.org/basis/pcseg-2/format/cp2k/>). After ensuring the appropriate version (e.g., pcseg-2 for a specific element or a general set), download the Basis Set. Open the downloaded file to ensure it contains

the expected basis set information. The file should include the exponents, coefficients, at least for Fe and O elements shown below. When encountering any issues, readers can consider reaching out to the BSE community or forums for assistance.

```
#-----
# Basis set: pcseg-2
# Description: Segmented contracted version of the pc-2 basis
# Role: orbital
# Version: 1 (Data from Frank Jensen (2014))
#-----
# Oxygen pcseg-2 (11s,6p,2d,1f) -> [4s,3p,2d,1f]
O pcseg-2
  10
1 0 0 7 1
  0.135887E+05      0.665492E-03
  0.203847E+04      0.514222E-02
  0.464041E+03      0.263472E-01
  0.131339E+03      0.101500E+00
  0.425340E+02      0.285877E+00
  0.150429E+02      0.500000E+00
  0.546616E+01      0.368211E+00
1 0 0 2 1
  0.100915E+02      -0.100084E+00
  0.110429E+01      0.500000E+00
1 0 0 1 1
  0.448082E+00      1.0000000
1 0 0 1 1
  0.175325E+00      1.0000000
1 1 1 4 1
  0.608220E+02      0.908253E-02
  0.140332E+02      0.645878E-01
  0.418244E+01      0.241502E+00
  0.142490E+01      0.500000E+00
1 1 1 1 1
  0.478741E+00      1.0000000
1 1 1 1 1
  0.146257E+00      1.0000000
1 2 2 1 1
  0.220000E+01      1.0000000
1 2 2 1 1
```

0.650000E+00	1.0000000
1 3 3 1 1	
0.110000E+01	1.0000000

Iron pcseg-2 (19s,14p,8d,2f,1g) -> [6s,4p,3d,2f,1g]

Fe pcseg-2

16	
1 0 0 10 1	
0.124866E+07	0.556470E-04
0.187039E+06	0.432457E-03
0.425683E+05	0.226917E-02
0.120562E+05	0.950693E-02
0.393233E+04	0.337679E-01
0.141914E+04	0.102683E+00
0.553832E+03	0.255373E+00
0.226129E+03	0.481641E+00
0.961317E+02	0.500000E+00
0.420443E+02	0.181434E+00
1 0 0 4 1	
0.337941E+03	-0.217335E-01
0.103114E+03	-0.104953E+00
0.161551E+02	0.500000E+00
0.676652E+01	0.492509E+00
1 0 0 2 1	
0.107404E+02	-0.156319E+00
0.174932E+01	0.500000E+00
1 0 0 1 1	
0.723737E+00	1.0000000
1 0 0 1 1	
0.147147E+00	1.0000000
1 0 0 1 1	
0.503270E-01	1.0000000
1 1 1 8 1	
0.483551E+04	0.528807E-03
0.114553E+04	0.459172E-02
0.370070E+03	0.249702E-01
0.141266E+03	0.947891E-01
0.591370E+02	0.264344E+00
0.261088E+02	0.484617E+00
0.119731E+02	0.500000E+00
0.538806E+01	0.201124E+00
1 1 1 4 1	
0.293038E+02	-0.192403E-01
0.439039E+01	0.266421E+00

	0.206040E+01	0.500000E+00
	0.952685E+00	0.415150E+00
1 1 1 1 1		
	0.412454E+00	1.0000000
1 1 1 1 1		
	0.115000E+00	1.0000000
1 2 2 6 1		
	0.138418E+03	0.345330E-02
	0.411431E+02	0.263915E-01
	0.153511E+02	0.109884E+00
	0.618032E+01	0.290275E+00
	0.261339E+01	0.463287E+00
	0.107371E+01	0.500000E+00
1 2 2 1 1		
	0.413063E+00	1.0000000
1 2 2 1 1		
	0.140918E+00	1.0000000
1 3 3 1 1		
	0.355300E+01	1.0000000
1 3 3 1 1		
	0.789000E+00	1.0000000
1 4 4 1 1		
	0.180000E+01	1.0000000
#	End of Basis set: pcseg-2	

2.0.3 Generate a raw .inp file from CIF

To create an input file for the CP2K program from a CIF file, it is recommended to follow these steps using Multiwfn:

- 1) Launch Multiwfn and load the structural file (Fe2O3.cif in the 0-cif_BS-preparing directory of the supplementary multimedia package).
- 2) Access the CP2K input file generation feature. Navigate to the main function 100, select the sub-function 2, which is for generating input files for various quantum chemistry program, and choose option 25 specifically for creating a CP2K input file.

- 3) Specify the output file path. Input the path where the CP2K input file to be saved.
- 4) Set calculation parameters. Use the available options to set up calculation parameters. This includes choosing the method, basis set, and other relevant settings.
- 5) Generate the input file. Select option 0 to generate the CP2K input file.

In the supplementary presentation file, specifically on slide 4 of "_sop-Manual_with_video-chain.pptx," an operational video is embedded to serve as an alternative reference.

Note: Multiwfn's interface for creating CP2K input files is continually being improved to accommodate updates and additional features of CP2K, so readers should always use the latest version of Multiwfn to ensure compatibility with the latest CP2K version.

2.0.4 Refine .inp file as required

Open the generated CP2K input file and adjust any parameters that need to be tailored to magnetic properties calculation, such as plane wave cut-off energy, spin configurations, XC functional, output specifications, etc.

Readers can find an example of *Fe2O3.inp* in the *1-cp2k_inp* directory of the supplementary multimedia package, which is shown below.

```
!*****Head of Input*****
&GLOBAL
  PROJECT alpha-Fe2O3
  PRINT_LEVEL MEDIUM
  RUN_TYPE ENERGY
&END GLOBAL

&FORCE_EVAL
```

```

METHOD Quickstep
&SUBSYS
  &CELL
    A    5.03417000    0.00000000    0.00000000
    B   -2.51708500    4.35971911    0.00000000
    C    0.00000000    0.00000000   13.73210000
    PERIODIC XYZ #Direction of applied PBC (geometry aspect)
  &END CELL
  &COORD
    Fe      0.00000000    0.00000000    1.98199999
    O       4.29412047    1.35813659   10.38743775
    Fe      0.00000000    0.00000000   11.86791701
    O      -1.75499997    3.03974912    3.46247925
    O       1.75499997    3.03974912   10.38743775
    O       0.78412053    1.35813659    3.46247925
    O      -0.97087944    4.39788571   10.38743775
    O       3.50999994    0.00000000    3.46247925
    FeX     2.53912050    4.39788571    4.94295851
    FeX     2.53912050    4.39788571    8.90695849
    Fe      2.53912050    1.46596190    6.59863899
    O       1.75499997    2.82409850    1.15415975
    Fe      2.53912050    1.46596190    2.63463901
    O       3.32324103    0.10782531    8.07911825
    O       1.75499997    0.10782531    1.15415975
    O       3.32324103    2.82409850    8.07911825
    O       4.10736156    1.46596190    1.15415975
    O       0.97087944    1.46596190    8.07911825
    FeX     2.53912050    1.46596190    9.55959751
    FeX     2.53912050    1.46596190   13.52359749
    Fe      0.00000000    2.93192381   11.21527799
    O      -0.78412053    4.29006040    5.77079875
    Fe      0.00000000    2.93192381    7.25127801
    O       0.78412053    1.57378721   12.69575725
    O      -0.78412053    1.57378721    5.77079875
    O       0.78412053    4.29006040   12.69575725
    O       1.56824106    2.93192381    5.77079875
    O      -1.56824106    2.93192381   12.69575725
    FeX     0.00000000    2.93192381    0.32631951
    FeX     0.00000000    2.93192381    4.29031949
  &END COORD
  &KIND O
    ELEMENT O
    BASIS_SET pcseg-2
    POTENTIAL ALL

```

```

&BS
&ALPHA
NEL 2
L 1
N 2
&END ALPHA
&BETA
NEL 2
L 1
N 2
&END BETA
&END BS
&END KIND
&KIND FeX
ELEMENT Fe
BASIS_SET pcseg-2
POTENTIAL ALL
&DFT_PLUS_U
L 2 #Quantum number of angular momentum the atomic orbitals to +U. 2=d, 3=f
U_MINUS_J [eV] 5 #Effective on-site Coulomb interaction parameter #U(eff) = U - J
&END DFT_PLUS_U
&BS
&ALPHA
NEL 4 -2
L 2 0
N 3 4
&END ALPHA
&BETA
NEL -6 -2
L 2 0
N 3 4
&END BETA
&END BS
&END KIND
&KIND Fe
ELEMENT Fe
BASIS_SET pcseg-2
POTENTIAL ALL
&DFT_PLUS_U
L 2 #Quantum number of angular momentum the atomic orbitals to +U. 2=d, 3=f
U_MINUS_J [eV] 5 #Effective on-site Coulomb interaction parameter #U(eff) = U - J
&END DFT_PLUS_U
&BS
&ALPHA

```

```

NEL -6 -2
L 2 0
N 3 4
&END ALPHA
&BETA
NEL 4 -2
L 2 0
N 3 4
&END BETA
&END BS
&END KIND
&END SUBSYS

&DFT
BASIS_SET_FILE_NAME BASIS_BSE_pcseg-2_FeO
POTENTIAL_FILE_NAME POTENTIAL
# WFN_RESTART_FILE_NAME alpha-Fe2O3-multiwfn-RESTART.wfn
CHARGE 0 #Net charge
# MULTIPLICITY 1 #Spin multiplicity
UKS
PLUS_U_METHOD MULLIKEN #The method used in DFT+U. Can also be Lowdin
&QS
EPS_DEFAULT 1E-10 #This is default. Set all EPS_xxx to values such that the energy
will be correct up to this value
METHOD GAPW
&END QS
&POISSON
PERIODIC XYZ #Direction(s) of PBC for calculating electrostatics
PSOLVER PERIODIC #The way to solve Poisson equation
&END POISSON
&XC
&XC_FUNCTIONAL
&MGGA_X_TPSS
&END MGGA_X_TPSS
&MGGA_C_TPSS
&END MGGA_C_TPSS
&END XC_FUNCTIONAL
&END XC
&MGRID
CUTOFF 350
REL_CUTOFF 50
&END MGRID
&SCF
MAX_SCF 128

```

```

EPS_SCF 1.0E-07 #Convergence threshold of density matrix during SCF
# SCF_GUESS RESTART #Use wavefunction from WFN_RESTART_FILE_NAME
&OT
  PRECONDITIONER FULL_ALL #Usually best but expensive for large system.
  MINIMIZER DIIS #CG is worth to consider in difficult cases
  LINESEARCH 2PNT #1D line search algorithm for CG. 2PNT is default
  ALGORITHM STRICT #Algorithm of OT. Can be STRICT (default) or IRAC
&END OT
&MIXING #How to mix old and new density matrices
  METHOD BROYDEN_MIXING #PULAY_MIXING is also a good alternative
  ALPHA 0.1 #Default. Mixing 40% of new density matrix with the old one
  NBROYDEN 8 #Default is 4. Number of previous steps stored for the actual
mixing scheme
&END MIXING
&END SCF
&PRINT
&MO
  ENERGIES T
  OCCUPATION_NUMBERS T
  &EACH
    QS_SCF 0
  &END EACH
&END MO
&DOS
&END DOS
&PDOS
  NLUMO -1
  COMPONENTS
&END PDOS
&MO_MOLDEN #Exporting .molden file containing wavefunction information
  NDIGITS 9 #Output orbital coefficients if absolute value is larger than 1E-9
  GTO_KIND SPHERICAL #Spherical-harmonic type of basis functions
&END MO_MOLDEN
&ELF_CUBE
  STRIDE 1 #Stride of exported cube file
&END ELF_CUBE
&E_DENSITY_CUBE
  STRIDE 1 #Stride of exported cube file
&END E_DENSITY_CUBE
&MULLIKEN
  PRINT_ALL F #If T, then printing full net AO and overlap population matrix
&END MULLIKEN
&MOMENTS
  PERIODIC T #Use Berry phase formula (T) or simple operator (F), the latter

```

normally applies to isolated systems

&END MOMENTS

&END PRINT

&END DFT

&PROPERTIES

&LINRES

RESTART

&LOCALIZE

MAX_ITER 20

&PRINT

&LOC_RESTART

&END

&END

RESTART

&END

ENERGY_GAP 2.0

PRECONDITIONER FULL_KINETIC

MAX_ITER 2500

&CURRENT

GAUGE R_AND_STEP_FUNCTION

ORBITAL_CENTER WANNIER

CHI_PBC T

&END CURRENT

&NMR

&PRINT

&CHI_TENSOR

FILENAME NMR_x

&END CHI_TENSOR

&SHIELDING_TENSOR

COMMON_ITERATION_LEVELS 10

FILENAME NMR_shielding

&END

&END

&END

&EPR

&PRINT

&G_TENSOR

ADD_LAST NUMERIC

COMMON_ITERATION_LEVELS 0

SOO_RHO_HARD T

&XC

```

&XC_FUNCTIONAL TPSS
&END XC_FUNCTIONAL
&END XC
FILENAME ESR-g
&END G_TENSOR
&END PRINT
&END EPR
&END
&END
&END FORCE_EVAL
!*****End of Input*****

```

2.1 Calculate spin density with CP2K

As shown in Fig.2.2, based on the Fe2O3.inp and available shell script, spin density file can be obtained after normal completion of CP2K with version above 2023.1.

In the supplementary presentation file, specifically on slide 6 of "***_sop-Manual_with_video-chain.pptx***," an operational video is embedded to serve as an alternative reference.

Note: Refer to the CP2K User's Manual or tutorials for detailed information on how to setup and use CP2K. If encountering issues, consider reaching out to the CP2K community through forums or mailing lists.

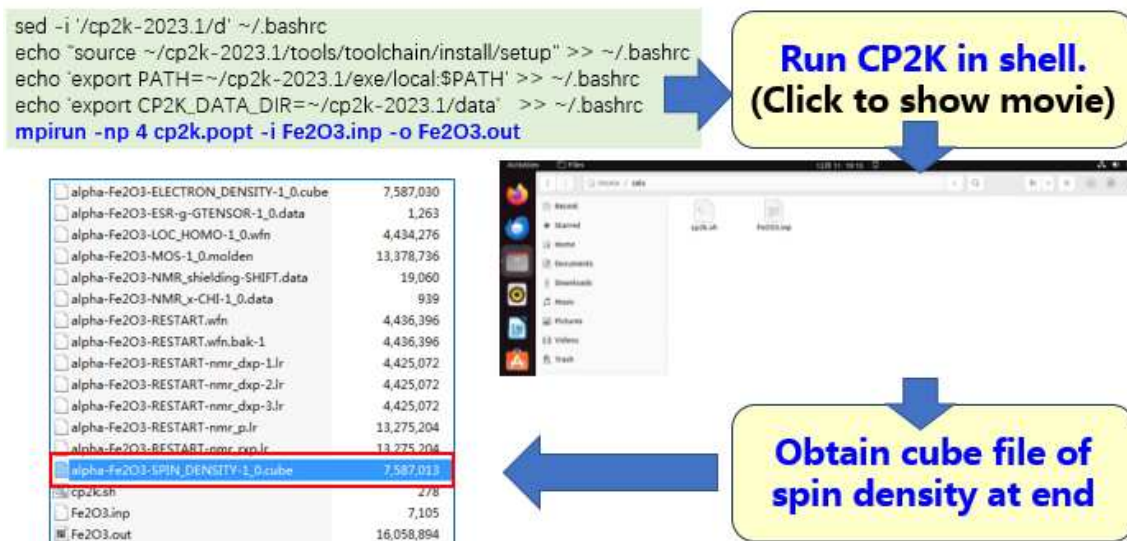


Fig.2.2 Key sub steps for running CP2K and obtain the spin density result

2.2 Transform spin density to moment file

2.2.1 Spin density to array of magnetic moment

To transform spin density from output of DFT program to magnetic moment distribution file for 3D vector magnetic field calculation, it is recommended to proceed following steps using the *cubeReading.llb* tool in the multimedia supplementary package:

- 1) Load the main entrance program, as shown in Fig.2.3, and input a spin density cube file, with an example of available format in the supplementary package.
- 2) Input lattice parameters and cut-off coefficient of output magnetic moments. The cutting coefficient "*abThreshold*" is important for calculation cost, which is discussed in the section 3.1. Higher "*abThreshold*" value may shorten the calculation time due to omission of lower magnetic moments, but it is not recommended to set a value above 0.4, so that the calculated magnetic field distribution makes sense.

3) For rhombohedral lattice like hematite, it is better to calculate and present magnetic fields in an orthorhombic lattice, so the original lattice should be transformed and extended with rotation plus translation operation. Therefore, the default values for "*Rotation_XY*", "*Translation*", "*plus RSTE*" and "*E-double RH2OR*" are recommended fixed.

4) Run the program by clicking the running icon in the upper left corner, select the "Save" icon if necessary. An operation video has been embedded in slice 7 of the supplementary multimedia file *_sop-Manual_with_video-chain.pptx*.

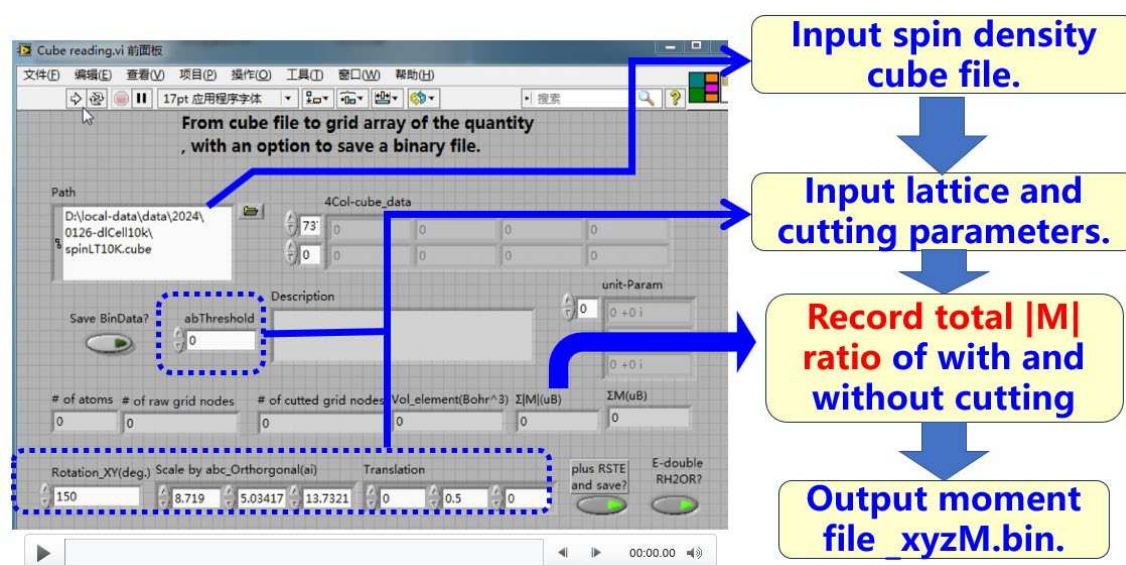


Fig.2.3 Key sub steps for transforming spin density to array of magnetic moment

2.2.2 Moment array to H calculation script

Before generating magnetic field from dipole moments, calculation scripts need to be prepared. It is recommended to proceed following steps using the *xyzM2H-CalScript.llb* tool in the multimedia supplementary package:

1) Load the main entrance program, as shown in Fig.2.4, and input a moment

distribution file, with an example of available format in the supplementary package.

2) So as to implement formula 1.22 or formula (a) of Fig.6, input an antiferromagnetic pitch angle, where 90 degrees for out-of-plane spin configuration and 0 degree for in-plane configuration. The current transform tool program is designed for collinear AFM, where positive and negative moments are set with opposite directions. This open-source tool allows for the extension to complex spin configurations by enabling the definition of non-collinear angles beyond anti-parallel alignment.

3) Input orthogonal lattice parameters and supercell configuration. The default supercell script “-1 1 -1 1 -1 1” indicates low or up limit of calculation range ($X_{low}=-1$ $X_{up}=1$ $Y_{low}=-1$ $Y_{up}=1$ $Z_{low}=-1$ $Z_{up}=1$), which corresponding to a proper $3 \times 3 \times 3$ supercell as discussed in the section 3.2. The calculated magnetic field distribution in the center cell of $3 \times 3 \times 3$ supercell has no obvious difference from that of $9 \times 9 \times 9$ supercell (with $X_{low}=-4$ $X_{up}=4$ $Y_{low}=-4$ $Y_{up}=4$ $Z_{low}=-4$ $Z_{up}=4$), but can reduce computer cost by 27 times.

4) Run the program by clicking the running icon in the upper left corner, select the “Save” icon if necessary. An operation video has been embedded in slice 8 of the supplementary multimedia file *_sop-Manual_with_video-chain.pptx*.

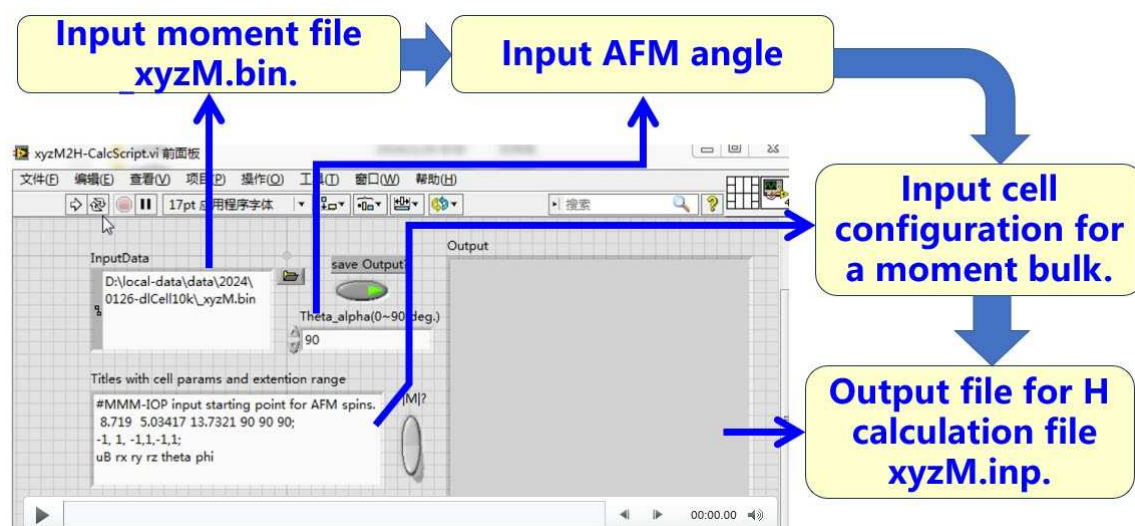


Fig.2.4 Key sub steps for generating magnetic field calculation script

2.3 Virtual measurement of magnetic fields

To generate magnetic field from dipole moments, this is the main step and usually takes above 24 hours to complete in a PC. It is recommended to proceed following steps using the *multiThreadYslices-FileInput.llb* tool in the multimedia supplementary package:

- 1) Copy the calculation script file *###.inp* and the setting file *###.ini* into a working directory. As shown in Fig.2.5, the default cut-off radius is 0.15 Å, which matches the spin density cell from DFT calculation, as discussed in the section 3.3.
- 2) Load the main entrance program and specify the working directory. Other data does not need to be entered manually, as they can be automatically read from the setting file. The output 3D vector magnetic field data for each Y-slice will be saved in the same directory.
- 3) Run the program by clicking the running icon in the upper left corner. The magnetostatic energy is calculated by implementation of section 1.3.3. An operation

video has been embedded in slice 9 of the supplementary multimedia file *_sop-Manual_with_video-chain.pptx*.

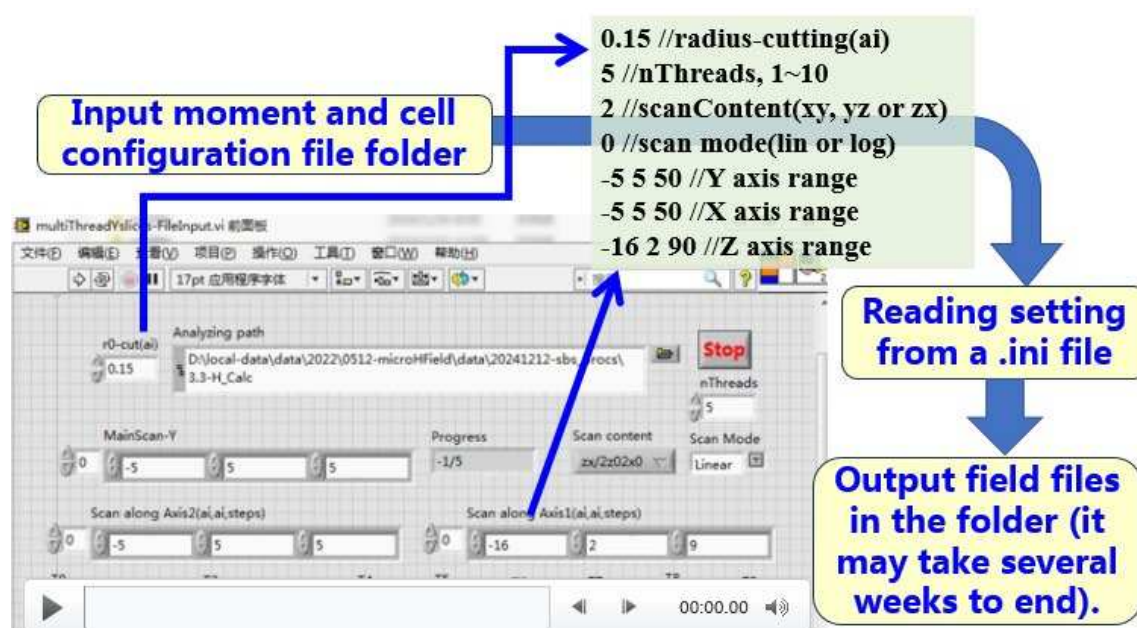


Fig.2.5 Key sub steps for calculating 3D vector magnetic field from array of magnetic moments

2.4 Analysis of 3D vector magnetic fields

2.4.1 Spin moment preparing for energy calculation

To analyze magnetic fields properly, it is necessary to adjust magnetic field with magnetic moment. The moment distribution file for calculating magnetic fields may be not well arranged after cutting off and lattice transformation (rhombohedral to orthogonal), so the *FileDataSliceVisualizing-preparing.llb* tool is developed to check and sort the magnetic moments from the raw moment file without cutting.

First, as shown in Fig.2.6, input the two files generated in step 2.1 (section 2.2.1), with the “*abThreshold*” parameter set to 0. Next, click the run icon in the upper left

corner to execute the program. Once the program completes, the *_M-trueCube_9Lines.bin* file will be automatically generated for use in step 4.2 (section 2.4.2).

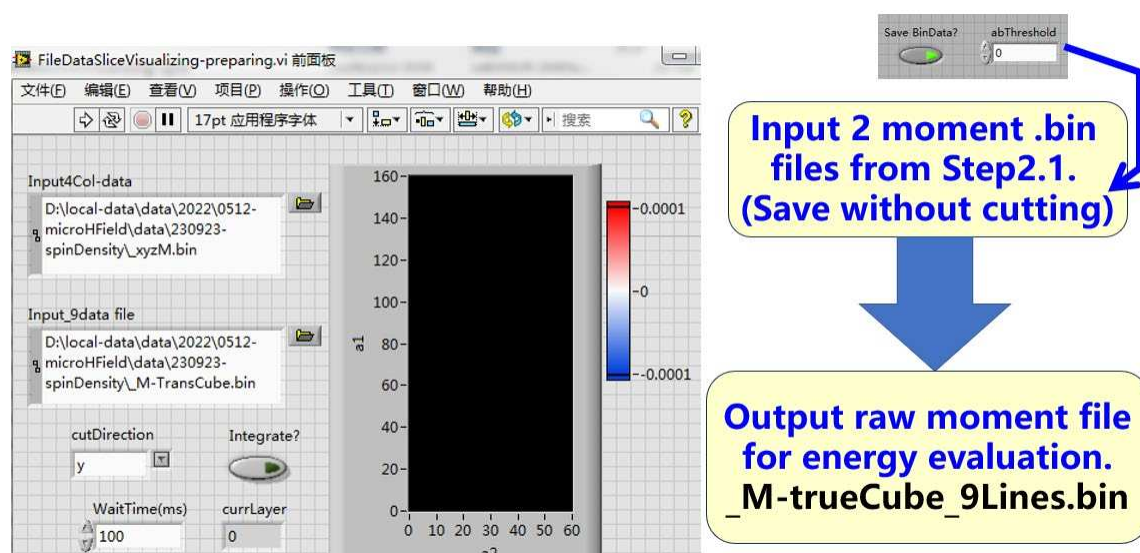


Fig.2.6 Key sub steps for preparing magnetic lattice data for further analysis

2.4.2 Unified field analysis including magnetostatic energy calculation

This is a key step to computer magnetic flux density and magnetostatic energy based on magnetic moment and field distribution. To analyze the magnetic field calculation results in a unified way, it is recommended to proceed following steps using the *BHmerging.llb* tool in the multimedia supplementary package:

- 1) Input directories of raw moment file and working folder of field calculation in the previous step. Next, as shown in Fig.2.7, input the moment ratio for correcting the field strength loss due to the moment cutting, as discussed in the section 3.1.
- 2) Input the pitch and azimuth angle for AFM spin configuration and spatial range, which should be consistent to that in section 2.2.2.

3) Run the program by clicking the running icon in the upper left corner. The magnetostatic energy is calculated by implementation of section 1.3.4. Once the program completes, the batch files of results for presentation will be automatically generated for use in the visualization steps, including magnetic fields, flux density, dipole moment, and energy. An operation video has been embedded in slice 11 of the supplementary multimedia file *_sop-Manual_with_video-chain.pptx*.

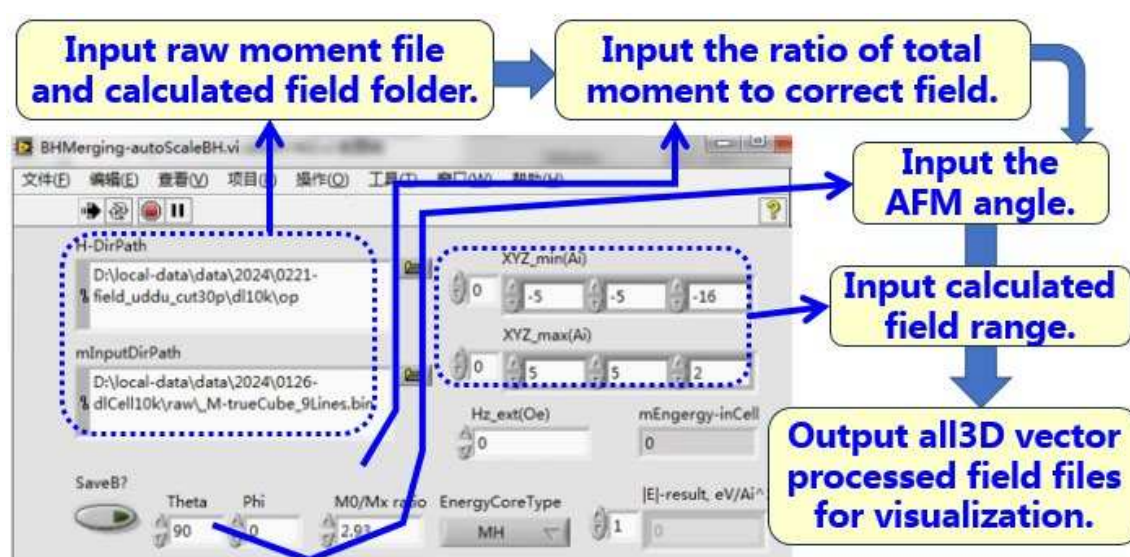


Fig.2.7 Key sub steps for combination analysis including energy calculation

2.5 Visualization of results

2.5.1 Statistical visualization

The magnetic field distribution can be analyzed and visualized by the *BinaCube-histoDemo.llb* tool in the multimedia supplementary package:

1) As shown in Fig.2.8, input data file generated from the previous step 4.2 (section 2.4.2).

- 2) Input the number of counting windows, and keep the “ $M0/M1$ ” ratio fixed at 1 if this parameter has already been correctly input in the previous step.
- 3) Run the program by clicking the running icon in the upper left corner. An operation video has been embedded in slice 12 of the supplementary multimedia file *_sop-Manual_with_video-chain.pptx*.

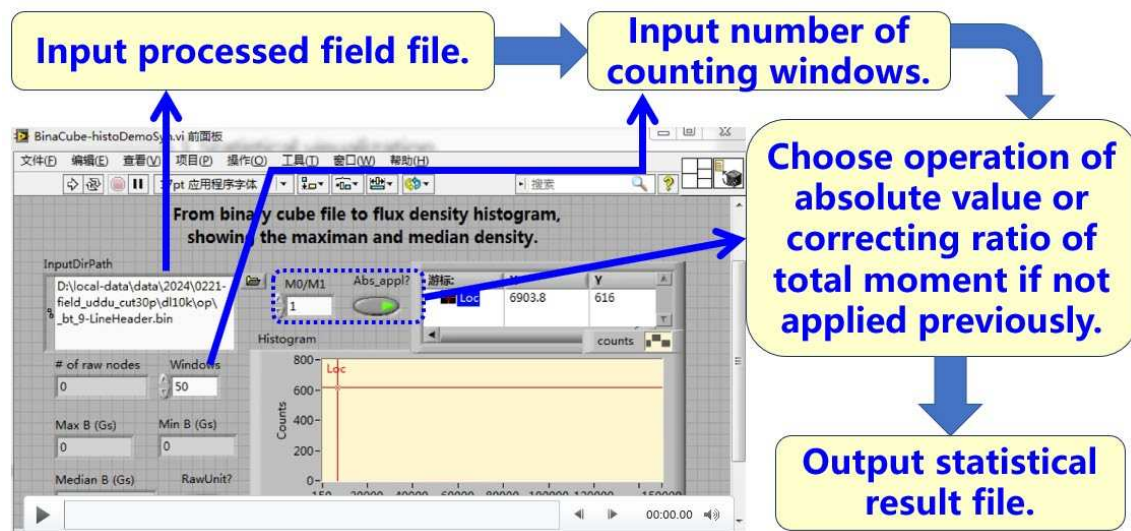


Fig.2.8 Key sub steps for statistical analysis of magnetic field distribution

2.5.2 Slice-by-slice visualization

The magnetic field distribution can be visualized in x/y/z cutting slices by the *FileDataSliceVisualizing-1slicePresent.vi* tool in the multimedia supplementary package:

- 1) As shown in Fig.2.9, input data file generated from the previous step 4.2 (section 2.4.2).
- 2) Input the cut direction and slice number.
- 3) Run the program by clicking the running icon in the upper left corner. The

magnetic field can be shown in 2D slices. An operation video has been embedded in slice 13 of the supplementary multimedia file *_sop-Manual_with_video-chain.pptx*.

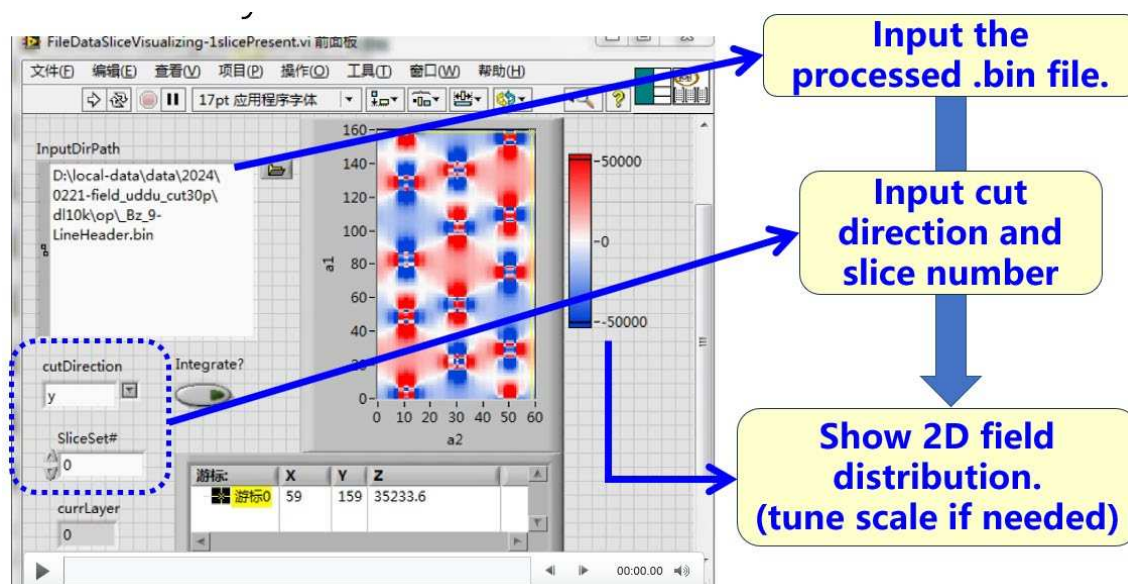


Fig.2.9 Key sub steps for tomography visualization of 3D vector magnetic fields

2.5.3 Isosurface 3D visualization

The magnetic field distribution can be visualized in isosurface fashion by the VMD (Ver. 1.9.3) tool after transformed by the *bin2cub.llb* tool in the multimedia supplementary package:

- 1) As shown in Fig.2.10, input data file generated from the previous step 4.2 (section 2.4.2), to transform the data format from binary to text of .cub for visualization in VMD. Run the program by clicking the running icon in the upper left corner.
- 2) Copy the transformed .cub file into the VMD main directory (*vmd.rc* and *showcub.vmd* files in the multimedia supplementary package should be copied together in the same directory, if they are not there).

3) The magnetic field can be shown in 3D isosurface fashion in VMD with a command “*cub xxx ###*” in the shell window, where “xxx” is the .cub filename without extension and ### is an isosurface value for visualization. An operation video has been embedded in slice 13 of the supplementary multimedia file *_sop-Manual_with_video-chain.pptx*.

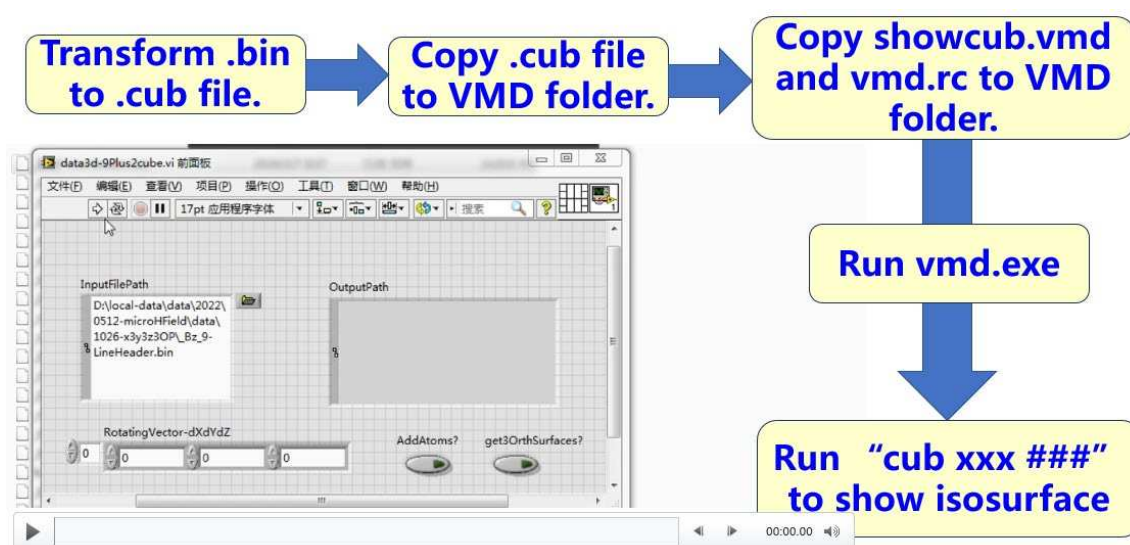


Fig.2.10 Key sub steps for visualizing 3D isosurface of vector magnetic fields

3. Notes on related approximations

3.1 Cut-off magnetic moment

As described in section 2.2.1, there is a parameter of cutting magnetic moment to reduce computation cost. How does the cut-off approximation influence the calculation results?

Neglecting small magnetic moments in magnetic field calculations will lead to a reduction in magnetic field strength; however, the magnetic field can be compensated proportionally based on the ratio of the total sum of the absolute

values of magnetic moments before and after truncation. The test results are shown in Table 3.1 and Fig.3.1, which prove that the field distribution keeps unaffected when the cutting percentage is no more than 30% ($abThreshold \leq 0.3$). When the cutting percentage is 40%, the distribution curve and peak value can be obviously changed for hematite.

The spin density is inhomogeneously distributed in the hematite lattice, and most space is filled with spin density below $0.001 \text{ \AA}^{-3}$, contributing negligible moments for magnetic field calculation in lattice. Therefore, major features of magnetic field distribution can be estimated by considering moments higher than some threshold. A smaller number of moments will inevitably cause underestimation of magnetic field strength, if the total amount of omitted moments is substantial. Nevertheless, research indicates that the ratio of the remaining moment before and after cutting, defined as $\alpha = M_0 / \Sigma |M|$, can be used to compensate magnetic fields when the cut-off percentage does not exceed 30%, by multiplying the calculated field strength by α .

Cutting moments before calculation is practically useful because it reduces the computation time by over 50 times. The comparison tests were performed on a PC equipped with a 4-core Intel i-6500 CPU, which has a theoretical peak performance of approximately 102 GFLOPS (Giga Floating Point Operations Per Second) in a 64-bit program. The number of moment elements was reduced from 1,152,000 to 19,268, and the computation time decreased from *176* days to 2.8 days. In Table 3-1, the computation time and peak value marked with left and right asterisks (*)

indicate that these values were extrapolated from data in other rows.

Table 3.1 Calculated peak magnetic field dependence on moment distribution after several cut-off setting

Cutting percentage	10 K					300 K	
	$\Sigma M , \mu_B$	$\alpha=M_0/\Sigma M $	# per cell	Time(Days)	$\alpha*B_p(T)$	$\Sigma M , \mu_B$	$M_0/\Sigma M $
0	$M_0=36.477$	1	1152000	*176*	*1.467(2)*	$M_0=36.4872$	1
0.07	27.2664	1.3378	74128	11.3	1.465	27.2690	1.3380
0.16	20.9898	1.7378	40848	5.6	1.468	20.9790	1.7392
0.2	18.5589	1.9655	33162	5.2	1.466	18.5631	1.9656
0.25	15.5584	2.3445	25590	3.7	1.470	15.5775	2.3423
0.3	12.4495	2.9300	19168	2.8	1.467	12.4492	2.9309
0.4	3.2854	11.1027	4534	0.63	1.430	3.3220	10.9835

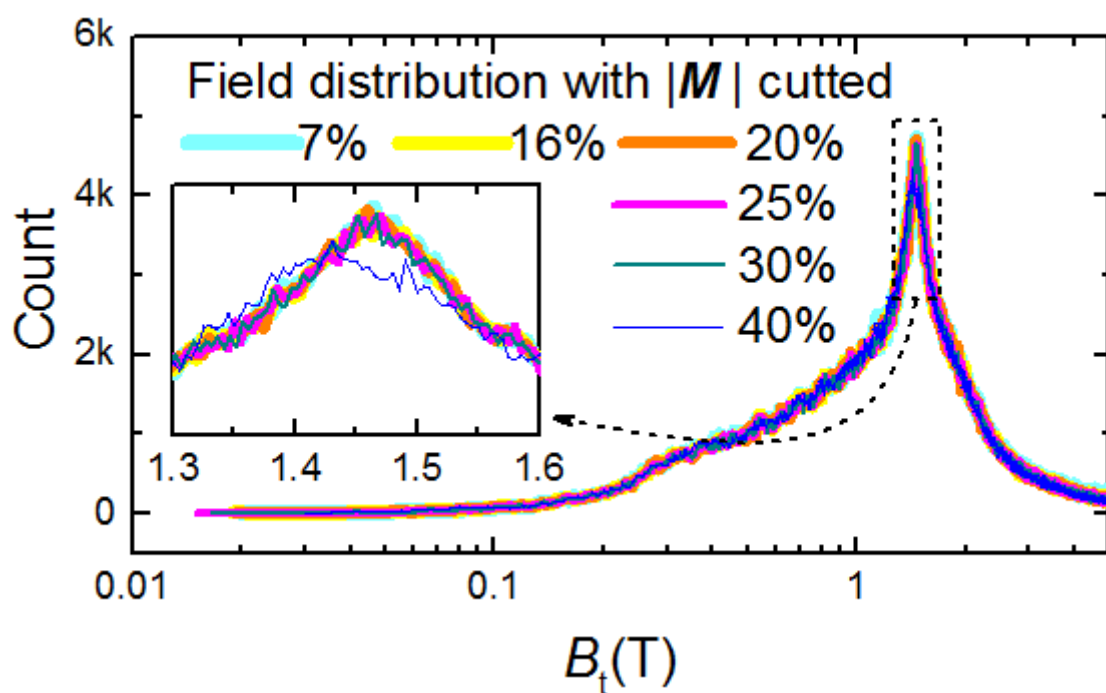


Fig.3.1 Magnetic field distribution dependence on different cut-off percentage of magnetic moments

3.2 Size of supercell

The size of the supercell is another parameter to consider in this work. To calculate magnetic fields in an orthogonal cell, four supercell configurations with

same distributed moments in the unit cell are compared. To specify the size of the supercell without considering the detailed distribution of moments, a single moment was set to $4.9 \mu_B$ at each Fe^{3+} ion position. As shown in Fig.3.2, while a $1 \times 1 \times 1$ supercell is inadequate for resolving the internal field distribution, a $3 \times 3 \times 3$ supercell yields results for the central cell that show negligible differences from those obtained with larger supercells.

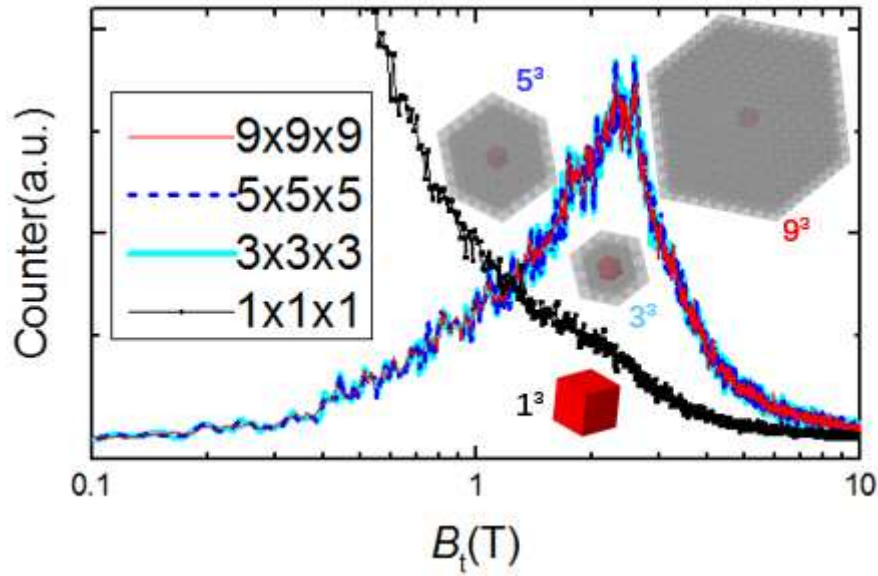


Fig.3.2 Magnetic field distribution dependence on size of supercell

3.3 Cut-off radius

In the formula 1.25 or formula (b) of Fig.6 in the main text, there is a cut-off radius parameter R_c , which should be properly set to obtain satisfying results. Various R_c in a wide range have been investigated. Based on formula 1.29, an index of energy fineness (ratio of total absolute $\mathbf{M} \cdot \mathbf{H}$ energy to overall $\mathbf{M} \cdot \mathbf{H}$ energy in the orthogonal unit cell) has been used to distinguish the cut-off effect. As shown in Fig.3.3, $R_c \sim 10$

pm gives a best fineness, which is comparable to the size of moment element for calculation (5~20 pm). Lower R_c produces too high and inhomogeneous field inside the moment element, while higher R_c may give too low and over-smooth field, so that the fineness is not satisfactory.

For Formula 1.29, which includes a distance variable in the denominator, there is a mathematical singularity when the distance approaches zero. However, in physical problems, this singularity can be resolved by maintaining the validity of the point approximation. In practical scenarios, when a test probe is near the field source, the source volume can be divided into smaller parts, allowing the probe to be sufficiently far away to safely use this formula. If the elemental source size is comparable to R_c , the singularity can be avoided both mathematically and physically.

To rigorously validate the numerical stability of the Biot--Savart real-space integration framework, the current author performed systematic convergence tests on two critical operational parameters: the spatial grid resolution and the integration truncation radius R_c . As illustrated in Fig.3.4(a), the computed field probability distributions exhibit significant aliasing and flattening artifacts at coarse grid resolutions (30 pm, 100 pm, and 1 nm). However, the distributions demonstrate strict numerical convergence for grid resolutions of 15 pm and finer (e.g., 1 pm and 0.1 pm), indicating that a 15 pm voxel step size is fully sufficient to capture the steep spatial gradients of the internal field without losing structural fidelity. Furthermore, Fig.3.4(b) maps the asymptotic behavior of the most probable field (B_{peak}) as the integration boundary R_c expands. The calculated B_{peak} converges to a stable,

physically meaningful value once R_c exceeds approximately 100 pm. These tests rigorously justify the chosen grid density and truncation parameters used in the main text, ensuring that the reported 3D field topologies are independent of the discrete integration boundaries.

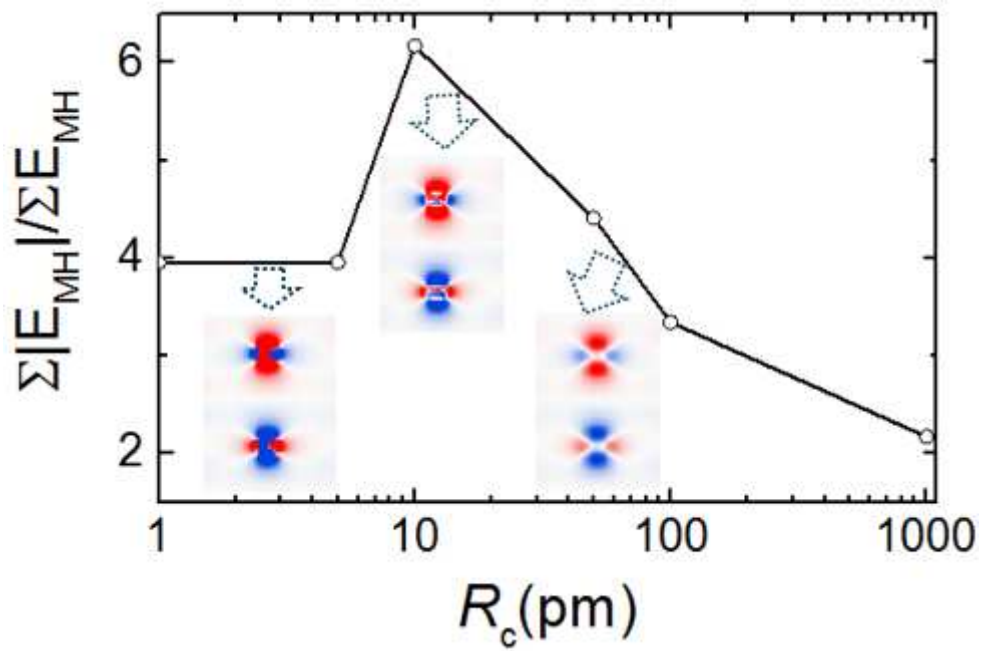


Fig.3.3 “Fineness” dependence on cut-off radius of magnetic dipole interaction

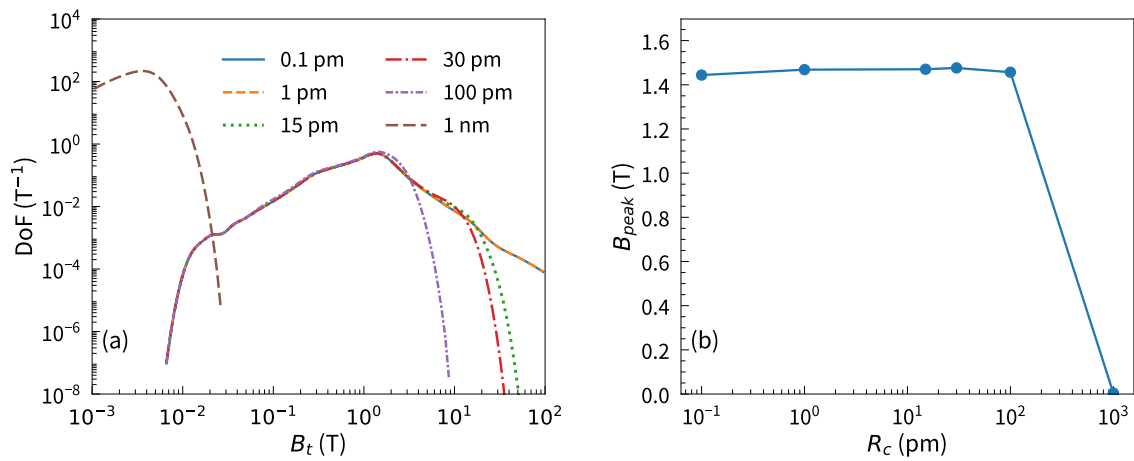


Fig.3.4 Numerical convergence analysis of the sub-lattice magnetostatic field reconstruction. (a) Probability density distributions (Density of Field, DoF) of the total magnetic field B calculated across different spatial grid resolutions ranging from 0.1

pm to 1 nm. Grid resolutions of 15 pm and finer (1 pm, 0.1 pm) yield perfectly overlapping distribution curves, establishing 15 pm as the strict numerical convergence threshold. (b) The most probable magnetic field (B_{peak}) as a function of the real-space integration truncation radius R_c . The peak field stabilizes at its asymptotic continuum value (~ 1.46 T) once R_c exceeds 100 pm.

3.4 Comparison of spin configurations

Since the simplest spin structure of hematite contains four Fe atoms, the current author considers four magnetic sublattices for its low-temperature crystallographic structure and begin by comparing the following magnetic configurations: $\uparrow \downarrow \downarrow \uparrow$, $\uparrow \downarrow \uparrow \downarrow$, $\downarrow \downarrow \uparrow \uparrow$, $\uparrow \uparrow \uparrow \uparrow$ and no spin order, where $\uparrow$ and $\downarrow$ designate up- and down-spin directions with respect to the z-axis. The total energies of these configurations have been shown in Table 3.2, in full agreement with experiment and earlier DFT calculations: the total energy is lowest for the $\uparrow \downarrow \downarrow \uparrow$ configuration (shown in Fig. 1a of main text), the other two antiferromagnetic spin configurations possess higher energy than that of $\uparrow \downarrow \downarrow \uparrow$, but lower than that of ferromagnetic order^[30]. The other two antiferromagnetic configurations are separated in energy from the ground state by about 156 and 1660 meV per Fe atom. The ferromagnetic configuration lies 1711 meV per Fe atom above the ground state.

Table 3.2 Converged energy of self-consistent field calculation for typical spin configurations

Spins	E (Hartree/cell)	ΔE_1 (meV/Fe)	ΔE_2 (eV/nm ³)
↑ ↓ ↓ ↑	-16520.1737	0	0
↑ ↓ ↑ ↓	-16520.1048	156	6.22
↑ ↑ ↓ ↓	-16519.4409	1660	66.16
↑ ↑ ↑ ↑	-16519.4192	1711	68.12
No order	-16518.9687	2732	109.30

3.5 Angular dependence of magnetostatic energy

The investigation of magnetostatic energy in magnetic materials, particularly in the context of temperature-dependent phase transitions, reveals complex interactions between anisotropy, dipole-dipole coupling, and thermal effects. When evaluating the total magnetostatic energy for the 10 K structure (as illustrated in Fig. 5b of main text), a critical relationship emerges as the tilt angle θ varies from 0° (in-plane antiferromagnetic order, IP-AFM) to $\pm 90^\circ$ (out-of-plane antiferromagnetic order, OP-AFM). At this low temperature, the system exhibits a pronounced preference for OP-AFM, primarily governed by the dominance of the single-ion anisotropy energy or zero field splitting energy (E_{MM} as shown equation 1.30). When θ shifts from in-plane to out-of-plane alignment, E_{MM} decreases by approximately 1.5 meV per iron atom. This represents a substantial reduction on the characteristic energy scale of magnetic systems, where anisotropy energies are typically on the order of 0.1–10 meV per atom. The physical origin of this decrease lies in the reorientation of magnetic moments relative to the crystal lattice. In uniaxial antiferromagnets like α -Fe₂O₃ (hematite), E_{MM} arises from spin-orbit coupling, which

locks the electron spins to the crystallographic axes. At $\theta = 0^\circ$ (IP-AFM), spins align perpendicular to the easy axis, maximizing anisotropy energy; at $\theta = 90^\circ$ (OP-AFM), spins parallel the easy axis, minimizing E_{MM} . This energy reduction is the principal driver of the low-temperature OP-AFM stability.

Concurrently, the magnetic dipole-dipole interaction energy E_{MH} increases by about 30 $\mu\text{eV}/\text{Fe}$ during this reorientation. This rise occurs because OP-AFM configurations enhance long-range dipole couplings. In IP-AFM, magnetic moments lie within the basal plane, leading to partial cancellation of dipole fields due to symmetry. In OP-AFM, moments align perpendicularly, amplifying dipole interactions as parallel spin components generate stronger stray fields. Despite this increase, E_{MH} remains negligible in the total energy balance at 10 K, constituting merely $\sim 2\%$ of E_{MM} 's variation (30 μeV vs. 1500 μeV). This insignificance stems from the inherently short-range, weak nature of dipole interactions compared to the robust single-ion anisotropy. Consequently, E_{MM} overwhelmingly dictates the energy landscape, favoring OP-AFM as the ground state.

As temperature elevates, thermal fluctuations progressively destabilize the OP-AFM phase. The single-ion anisotropy, being a local, atomic-scale property, is highly sensitive to phonon-induced lattice vibrations and spin disorder. These fluctuations diminish the effective anisotropy barrier. Around the Morin transition temperature T_M , E_{MM} 's thermal suppression erodes its dominance, allowing previously subordinate energies to dictate spin reorientation.

This reorientation is vividly demonstrated at 300 K, where IP-AFM prevails. Here,

the anisotropy energy—now thermally softened—no longer suffices to maintain out-of-plane order. Instead, the smaller but temperature-resilient dipole energy, combined with entropy gains, tips the balance. Crucially, the transition is first-order, exhibiting hysteresis and latent heat, as confirmed by calorimetry^[31]. External factors like pressure or doping can tune T_M by altering E_{MM} via lattice compression or spin-orbit modulation.

At low temperatures, the spin configuration of the antiferromagnetic ground state is readily determined, and the presence or absence of the Morin transition is not difficult to establish. However, beyond the contributions of single-ion anisotropy energy and dipolar interaction energy, the lattice constants and exchange interaction energy also vary with temperature, together with magnon excitation. Critically, the exchange interaction energy can exceed 100 meV/Fe (Table 3.2). The driving factors of the Morin transition are thus so complex that its in-plane angle becomes difficult to predict accurately. Nevertheless, it is noteworthy that the in-plane anisotropic distribution of dipolar interaction energy at 10 K (shown in Fig.3.5) exhibits no trigonal symmetry. This indicates that it is primarily governed by the non-trigonal symmetry of the local spin density distribution.

Therefore, the 10 K energy landscape is dominated by single-ion anisotropy, favoring OP-AFM despite a minor dipole-energy penalty. With heating, thermal fluctuations degrade anisotropy, empowering dipole interactions and entropy to drive the Morin transition. At 300 K, IP-AFM emerges, illustrating how temperature reconfigured the hierarchy of microscopic energies—a delicate balance pivotal for

designing next-generation magnetic materials.

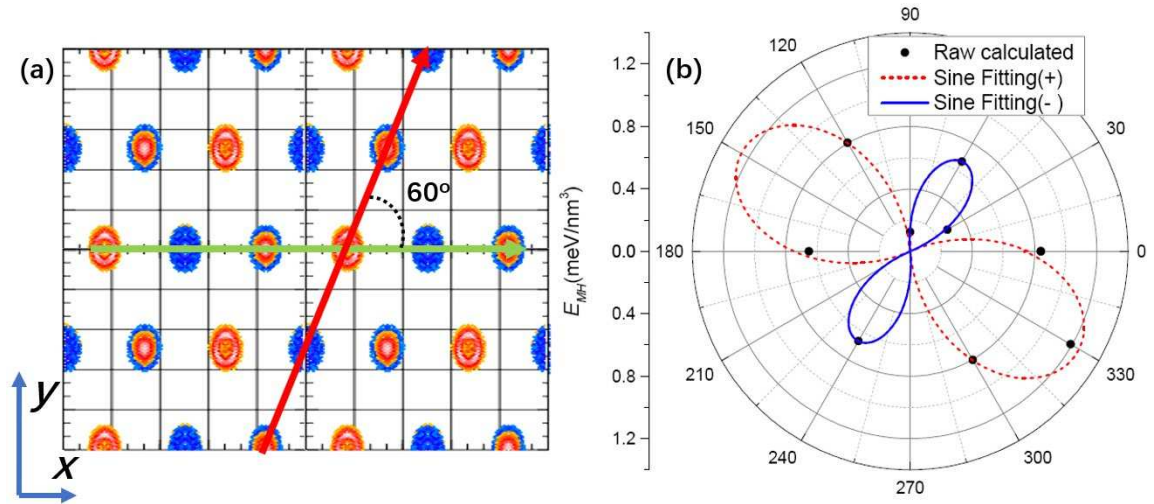


Fig.3.5 Schematics of magnetostatic anisotropy in hematite at 10 K: (a) a top-view representation of the spin density in the $2 \times 2 \times 1$ orthogonal supercell along the z -direction; (b) the in-plane anisotropy of magnetic dipole interaction energy.

3.6 Surface relaxation on field attenuation

To probe the attenuation of the magnetic field at the surface, the current author computed from first principles the changes in the vector magnetic field arising from spin density relaxation during vacuum cleavage along the XY plane. All simulations were performed for hematite at 300 K, employing its experimental unit cell parameters and an in-plane (IP) spin configuration. The surface contribution was isolated by calculating the spin densities for two distinct systems: a conventional bulk structure with full three-dimensional periodicity and a freestanding slab model, created by truncating the bulk to three unit cells in thickness along Z , with periodicity maintained only along the X and Y directions.

As shown in Fig.3.6, the magnetic field decay below the hematite surface exhibits

two distinct regimes. Within the first 0.5 nm, the field strength drops precipitously by over two orders of magnitude. Beyond 1 nm, it decays more gradually at a rate of approximately 60% per nanometer, yielding magnetic flux densities of 0.067 mT (B_z) and 0.16 mT (B_x) at a height of 4 nm. Furthermore, surface relaxation enhances the magnetic field above the 1 nm height, amplifying the B_x and B_z components by factors of 2 and 5, respectively, despite inapparent variations on relaxation process within the first 0.5 nm.

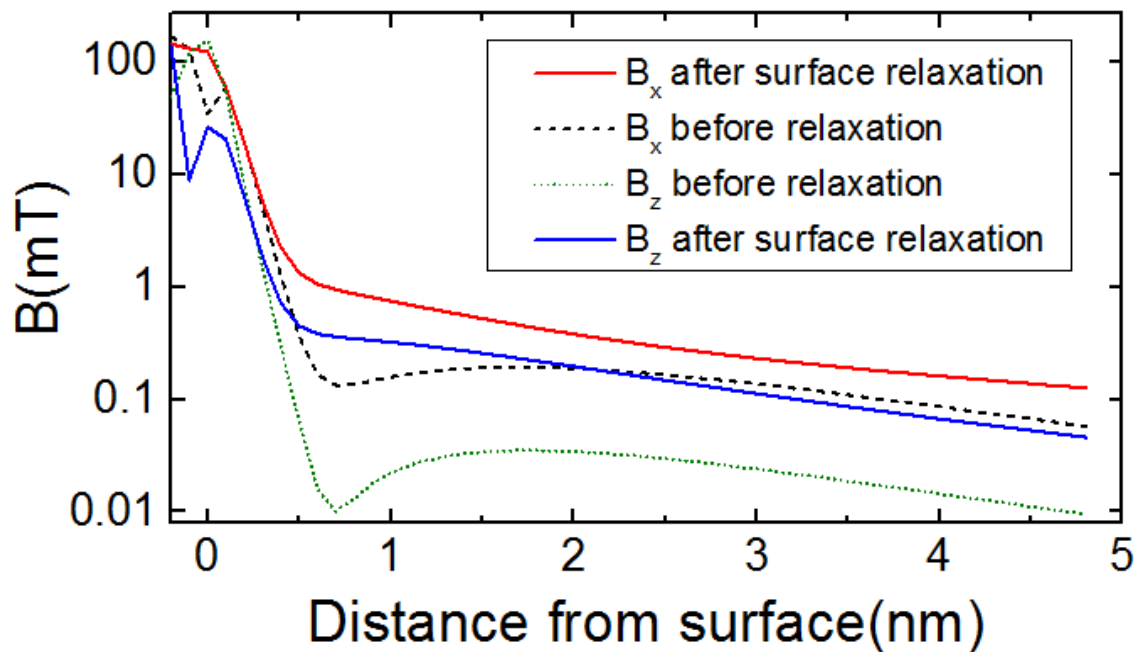


Fig.3.6 Magnetic field decay profiles from the hematite (001) surface. The red and blue solid lines correspond to the B_x and B_z components after surface relaxation, respectively; the black and green dashed lines represent the components before relaxation.

3.7 Noncollinear calculation from 3D vector spin density

3.7.1 Preparing of 3D vector spin density

A DFT calculation of the vector spin density was conducted to examine how the non-collinear spin structure above the Morin transition influences the magnetic field distribution in hematite, starting from the unit cell parameters at 300 K.

First-principles calculations based on non-collinear spin density functional theory (ncDFT) are carried out using QUANTUM ESPRESSO to directly compute the 3D vector spin density^[32]. The calculation is a self-consistent field (scf) run on a hexagonal unit cell (ibrav = 4), incorporating two critical relativistic effects: full non-collinear magnetism (noncolin = .true.), allowing the spin moment vector to point in any direction in 3D space, and spin-orbit coupling (lspinorb = .true.).

To better describe the strongly correlated 3d electrons of iron, the DFT+U method (lda_plus_u = .true.) is applied with a Hubbard U parameter of 5 eV for both Fe species. A key feature is the definition of an initial collinear antiferromagnetic configuration in the crystallographic a-b plane. This is achieved by setting starting_magnetization to +0.3 and -0.3 for the two Fe sites (Fe and FeX), with their spin moments oriented at specific polar angles (angle1, angle2) to be anti-parallel along the x-axis. The atomic structure is defined using crystal coordinates, and the Brillouin zone is sampled with a 10×10×4 k-point mesh. Fully relativistic, spin-polarized projector augmented-wave (PAW) pseudopotentials are specified for Fe and O. Convergence is controlled by setting an electronic energy threshold of 1.0d-

8 Ry and allowing up to 500 diagonalization steps, using the David algorithm.

To ensure reproducibility, the current author provides the QUANTUM ESPRESSO input file used to calculate the non-collinear 3D vector spin density, starting from an in-plane spin configuration.

```
!*****Head of Input*****
&control
  calculation = 'scf'
  prefix = 'fe2o3'
  pseudo_dir = './pseudo'
  outdir = './out_ncol'
  restart_mode = 'restart'
&system
 ibrav = 4
  celldm(1) = 9.515
  celldm(3) = 2.732
  nat = 30
  ntyp = 3
  ecutwfc = 60.0
  ecutrho = 480.0
  noncolin = .true.
  lspinorb = .true.
  lda_plus_u = .true.
  lda_plus_u_kind = 1
  Hubbard_U(1) = 5
  Hubbard_U(2) = 5
  Hubbard_U(3) = 0
  starting_magnetization(1) = 0.3
  angle1(1) = 90
  angle2(1) = 0
  starting_magnetization(2) = 0.3
  angle1(2) = 90
  angle2(2) = 180
  occupations = 'tetrahedra'
  nbnd = 400
&electrons
  conv_thr = 1.0d-8
  electron_maxstep = 500
  diagonalization = 'david'
  mixing_beta = 0.1
  mixing_mode='plain'
```

```

mixing_ndim=4
! CELL_PARAMETERS angstrom
!   5.0351280000  0.0000000000  0.0000000000
!   -2.5175640000  4.3605487593  0.0000000000
!   0.0000000000  0.0000000000  13.7581000000
ATOMIC_SPECIES
Fe      55.845  Fe.rel-pbesol-spn-kjpaw_psl.1.0.0.UPF
FeX     55.845  Fe.rel-pbesol-spn-kjpaw_psl.1.0.0.UPF
O       15.999  O.pbesol-n-kjpaw_psl.1.0.0.UPF
ATOMIC_POSITIONS crystal
Fe      0.00000000  0.00000000  0.14406059
O       1.00856237  0.31145987  0.75500525
Fe      0.00000000  0.00000000  0.86261308
O      -0.00000016  0.69710210  0.25166842
O       0.69710226  0.69710210  0.75500525
O       0.31145995  0.31145987  0.25166842
O       0.31145978  1.00856197  0.75500525
O       0.69710243  0.00000000  0.25166842
FeX     1.00856221  1.00856197  0.35927625
FeX     1.00856221  1.00856197  0.64739742
Fe      0.67237488  0.33618732  0.47961848
O       0.67237481  0.64764720  0.08388947
Fe      0.67237488  0.33618732  0.19149730
O       0.67237496  0.02472745  0.58722631
O       0.36091494  0.02472745  0.08388947
O       0.98383483  0.64764720  0.58722631
O       0.98383490  0.33618732  0.08388947
O       0.36091487  0.33618732  0.58722631
FeX     0.67237488  0.33618732  0.69483413
FeX     0.67237488  0.33618732  0.98295531
Fe      0.33618732  0.67237465  0.81517637
O       0.33618725  0.98383452  0.41944736
Fe      0.33618732  0.67237465  0.52705519
O       0.33618740  0.36091478  0.92278420
O       0.02472738  0.36091478  0.41944736
O       0.64764727  0.98383452  0.92278420
O       0.64764734  0.67237465  0.41944736
O       0.02472731  0.67237465  0.92278420
FeX     0.33618732  0.67237465  0.02371836
FeX     0.33618732  0.67237465  0.31183953
K_POINTS automatic
10 10 4 0 0 0
!*****End of Input*****

```

3.7.2 First-principle determined 3D vector spin density

The study finds that the non-collinear calculated energy is lower by 706 meV/Fe and 752 meV/Fe compared to collinear and non-magnetic calculations, respectively. The detailed energy comparison is presented in Table 3.3. The three components of the spin density are presented in Fig.3.7, with a summary analysis of the corresponding spin magnetic moment per unit cell provided in Table 3.4. It is found that the spin density is dominated by the x-component (the total moment contributions from the y- and z-directions are each less than 1%). However, the antiferromagnetic axis slightly deviates from the x-direction, exhibiting a small tilt with polar and azimuthal angles of 1.1° and 0.7° , respectively. Furthermore, although a weak ferromagnetic canting is present, its angle is only 0.02° and lies within the xy-plane, which is consistent with previous calculations^[33].

Table 3.3 Comparison of DFT calculation energy for 3 spin configurations

QE-Spins	E (Hartree/cell)	ΔE_1 (meV/Fe)	ΔE_2 (eV/nm ³)
No order	-16609.714	752	30
Collinear	-16609.734	706	28
Non-col	-16610.046	0	0

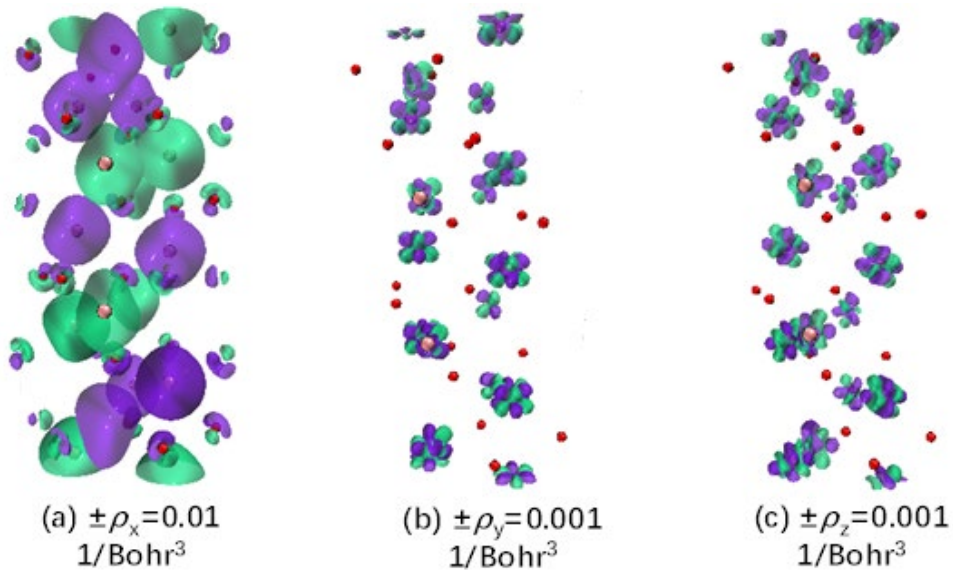


Fig.3.7 Isosurface plots of the three components of the vector spin density: (a) ρ_x , (b) ρ_y , (c) ρ_z , where brown and red spheres indicate Fe and O atoms respectively.

Table 3.4 Vector spin moment analysis of non-collinear calculated results.

m	m_x	m_y	m_z
$\Sigma M , \mu_B$	50.9	0.32	0.53
$ \Sigma M , \mu_B$	0.0008	0.01	0.0001
$\Sigma M/m_0 $	0.999	0.0063	0.01
$ \Sigma M /m_0$	0	0.0002	0
$(\theta, \varphi)_{\text{AFM}}$	$\varphi = 0.7^\circ$		$\theta = 1.1^\circ$
$(\theta', \varphi')_{\text{canting}}$	$\varphi' = 0.02^\circ$		$\theta' = 0$

3.7.3 Magnitude Distribution of the Vector Magnetic Field from Non-Collinear Vector Spin Density

Under the Hubbard U parameter set to 5 eV, the non-collinear calculation (QE) yields a band gap of 2.0 eV. This value is highly consistent with the 2.1 eV band

gap derived from the collinear in-plane antiferromagnetic configuration (CP2K) discussed in the main text. Despite this minimal difference in the electronic band gaps, their reconstructed magnetic fields exhibit a quantitative scaling difference. As shown in Fig.3.8, the magnetic field derived directly from the non-collinear vector spin density shows an overall intensity 1.3 times higher than the field derived from the scalar spin density. Nevertheless, both approaches yield a virtually identical spatial distribution of the magnetic field. Its most probable field strength, however, remains lower than that from the out-of-plane configuration. The origin of the intensity inconsistency lies in the different computational setups: the non-collinear calculation utilized the GGA functional with a pseudopotential basis, in contrast to the meta-GGA functional and all-electron basis employed for the scalar spin-density calculation in the main text, which provides superior accuracy.

Upon comparing the magnetic fields produced by individual spin-density components, as shown in Fig.3.9, it is found that the x-component yields a field nearly identical to the total field. In contrast, the contributions from the y- and z-components are significantly weaker, being lower by about two and three orders of magnitude, respectively.

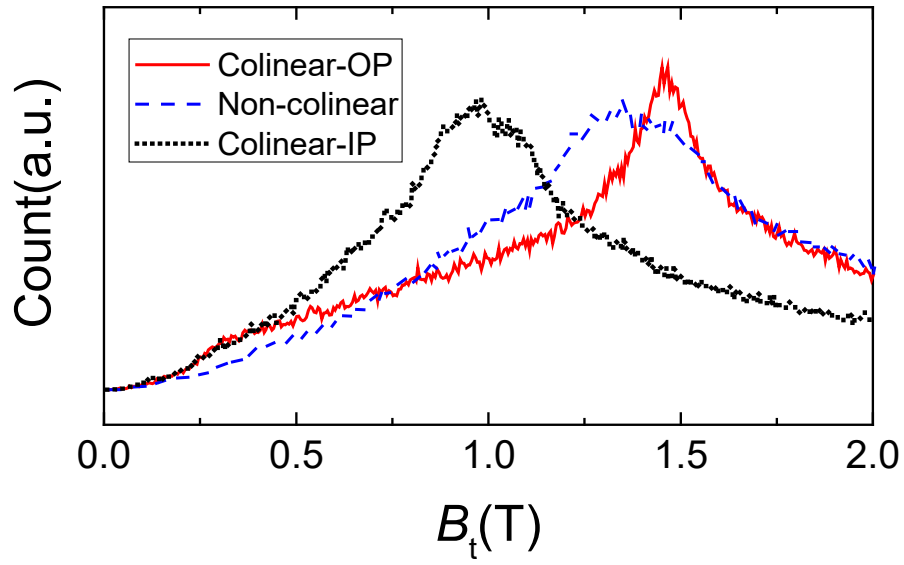


Fig.3.8 Field-dependent volume fraction density of total field for non-colinear spin calculation and colinear scalar calculation with in-plane and out-of-plane configuration.

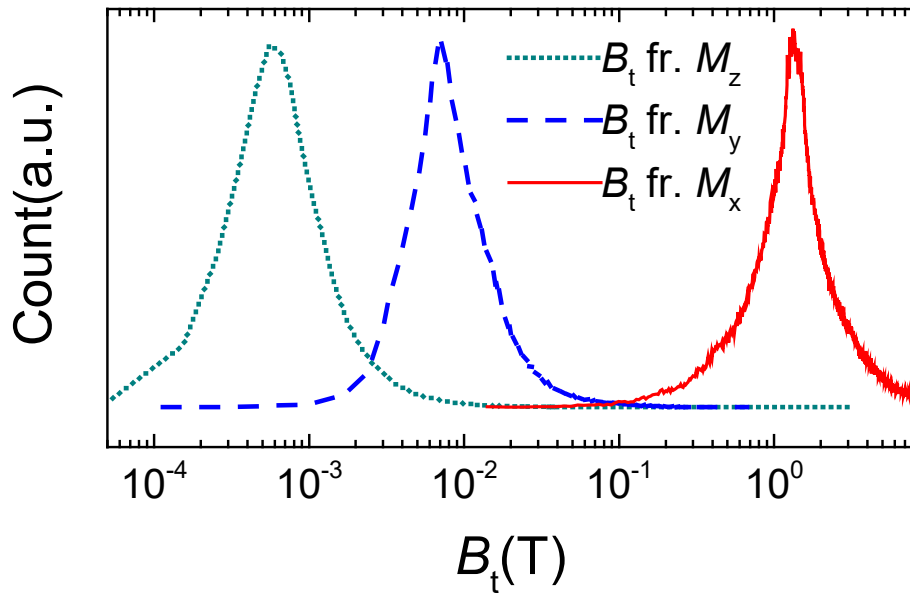


Fig.3.9 Field-dependent volume fraction density of total field for three component of non-colinear calculated vector spin density.

3.7.4 Spatial Mapping of the Vector Magnetic Field Obtained from Non-collinear Vector Spin Density Calculations

A comparative analysis was conducted on the three-dimensional spatial distribution of magnetic fields generated by the full vector and individual component spin densities. The magnetic field intensity maps for the out-of-plane and in-plane configurations derived from scalar spin density are presented in Fig.3.10- Fig.3.11 and Fig.3.12-Fig.3.13, respectively. The results from the total vector spin density and its total, x-, y-, and z-components are shown in Fig.3.14- Fig.3.15, Fig.3.16-Fig.3.17, Fig.3.18-Fig.3.19, and Fig.3.20, respectively. The analysis reveals two key results: First, the 1.3 T isosurface of the 3D vector magnetic field obtained directly from first-principles non-collinear spin density shows no significant morphological difference from the 1.0 T isosurface generated by the in-plane antiferromagnetic configuration based on scalar spin density. Second, the overall field distribution is predominantly determined by the x-component of the spin density. In contrast, the magnetic contributions from the y- and z-components are substantially weaker, lower by approximately two and three orders of magnitude, respectively—a finding consistent with the intensity analysis in the preceding section.

Therefore, it can be concluded that the extremely small antiferromagnetic canting angle has a negligible impact on the 3D magnetic field distribution. Consequently, both the spatial pattern and intensity of the magnetic field produced by setting an in-plane antiferromagnetic spin configuration for hematite using scalar spin moments are reliable.

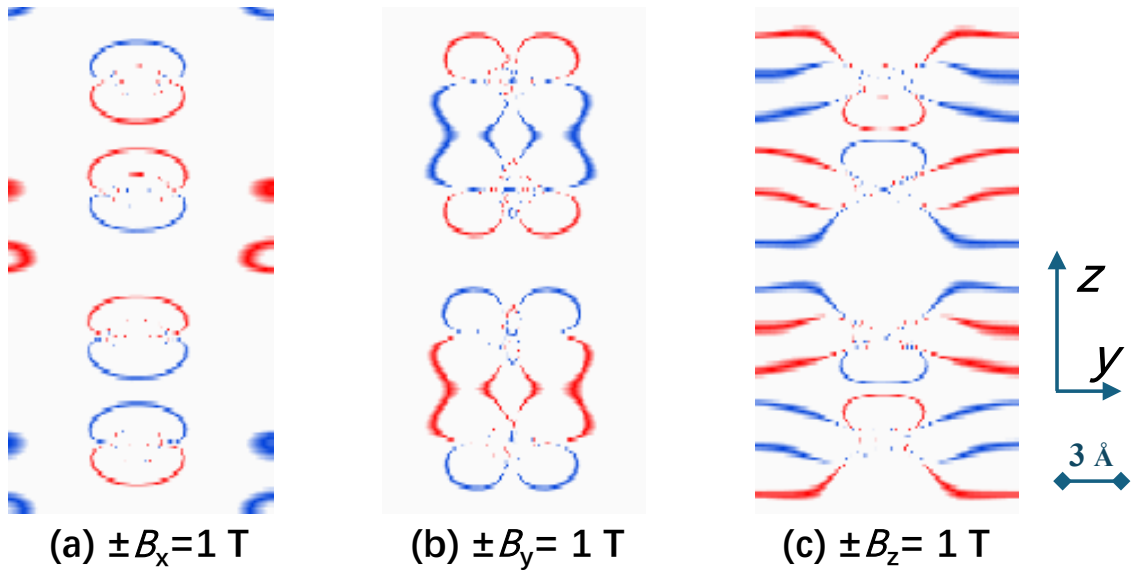


Fig.3.10 Magnetic field map corresponding to $1 \text{ T} \pm 25\%$ on the $x=0$ plane of the orthogonal unit cell: (a) B_x , (b) B_y , and (c) B_z . These fields originate from out-of-plane configuration of the scalar spin density and are depicted with red/blue coloring for positive/negative values.

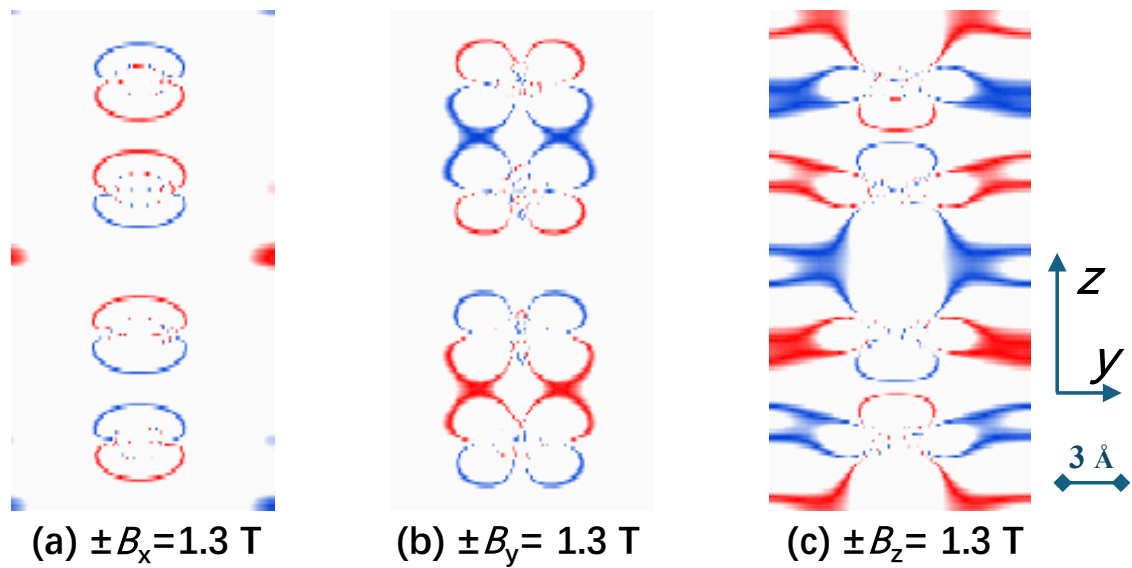


Fig.3.11 Magnetic field map corresponding to $1.3 \text{ T} \pm 25\%$ on the $x=0$ plane of the orthogonal unit cell: (a) B_x , (b) B_y , and (c) B_z . These fields originate from out-of-plane configuration of the scalar spin density and are depicted with red/blue coloring for positive/negative values.

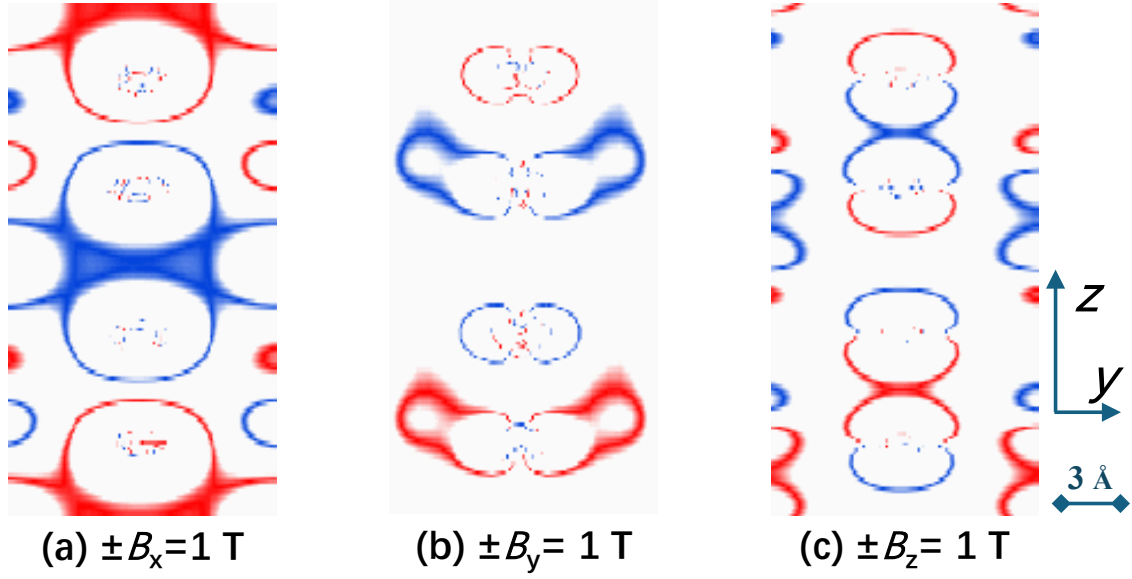


Fig.3.12 Magnetic field map corresponding to $1 \text{ T} \pm 25\%$ on the $x=0$ plane of the orthogonal unit cell: (a) B_x , (b) B_y , and (c) B_z . These fields originate from in-plane configuration of the scalar spin density and are depicted with red/blue coloring for positive/negative values.

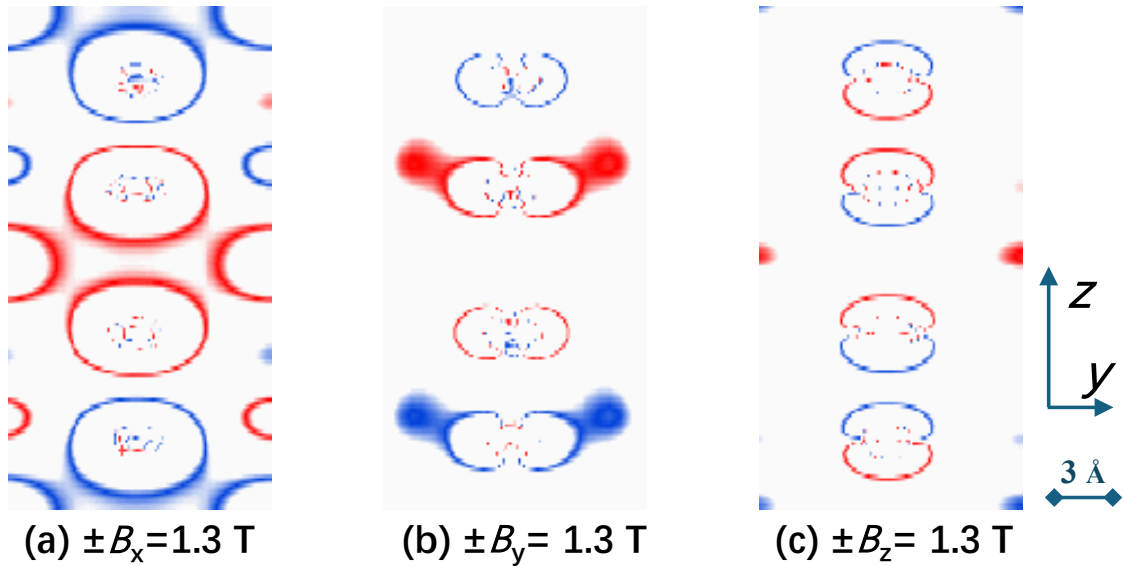


Fig.3.13 Magnetic field map corresponding to $1.3 \text{ T} \pm 25\%$ on the $x=0$ plane of the orthogonal unit cell: (a) B_x , (b) B_y , and (c) B_z . These fields originate from in-plane configuration of the scalar spin density and are depicted with red/blue coloring for positive/negative values.

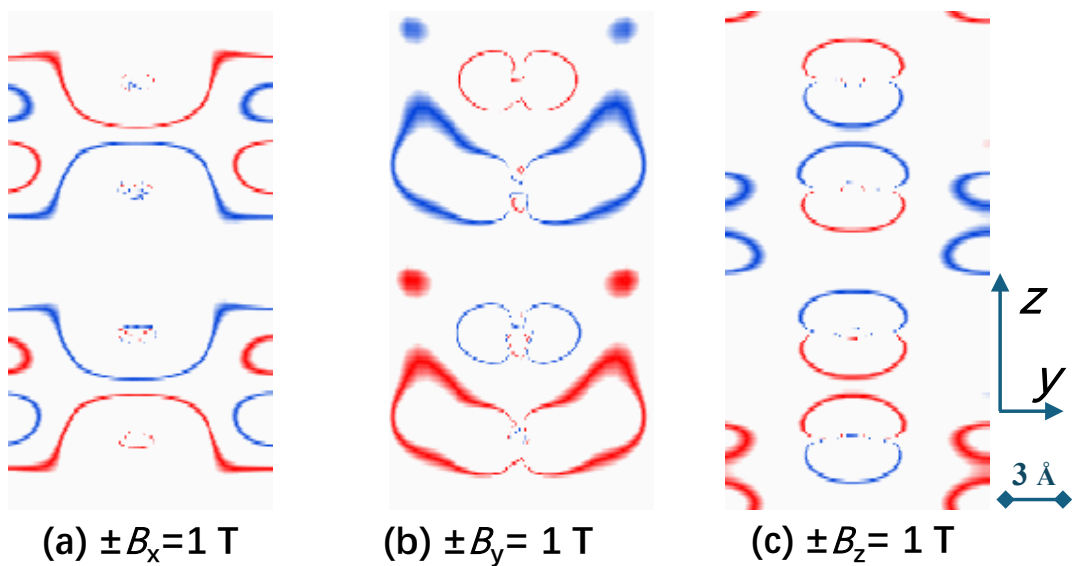


Fig.3.14 Magnetic field map corresponding to $1 \text{ T} \pm 25\%$ on the $x=0$ plane of the orthogonal unit cell: (a) B_x , (b) B_y , and (c) B_z . These fields originate from total three components of the vector spin density and are depicted with red/blue coloring for positive/negative values.

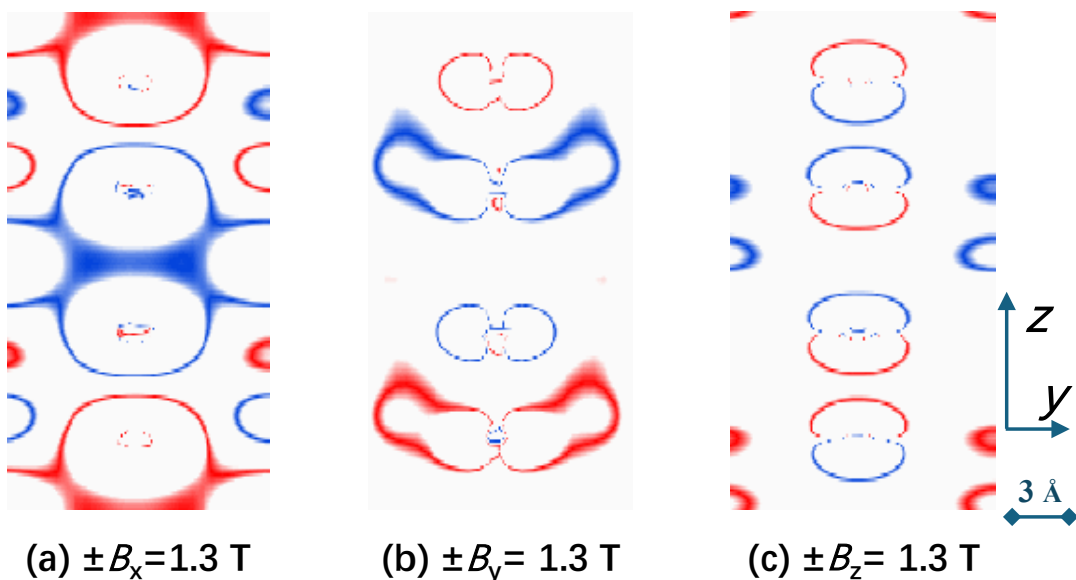


Fig.3.15 Magnetic field map corresponding to $1.3 \text{ T} \pm 25\%$ on the $x=0$ plane of the orthogonal unit cell: (a) B_x , (b) B_y , and (c) B_z . These fields originate from total three components of the vector spin density and are depicted with red/blue coloring for positive/negative values.

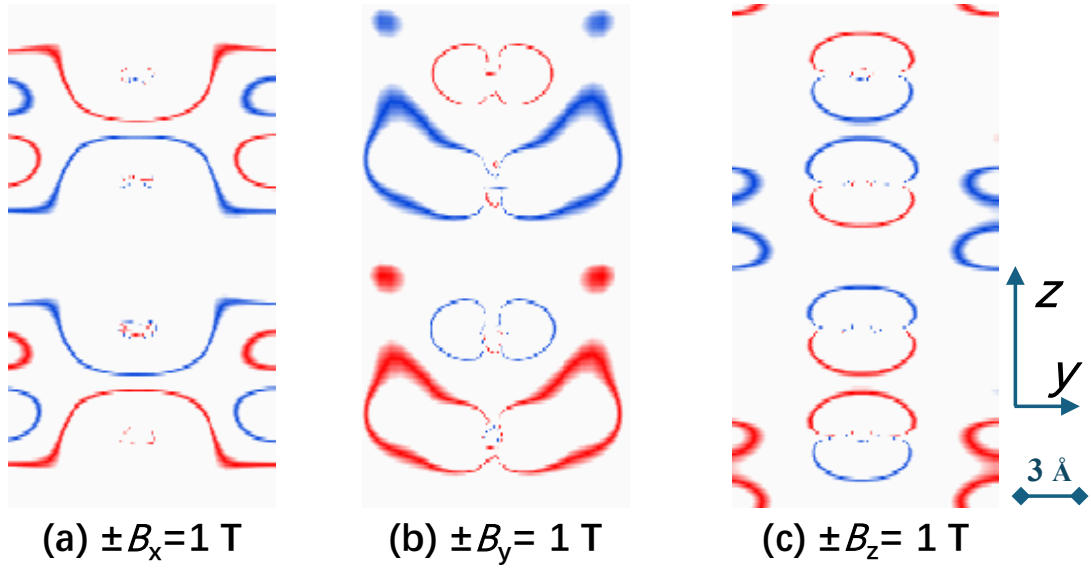


Fig.3.16 Magnetic field map corresponding to $1 \text{ T} \pm 25\%$ on the $x=0$ plane of the orthogonal unit cell: (a) B_x , (b) B_y , and (c) B_z . These fields originate from the x -component of the vector spin density and are depicted with red/blue coloring for positive/negative values.

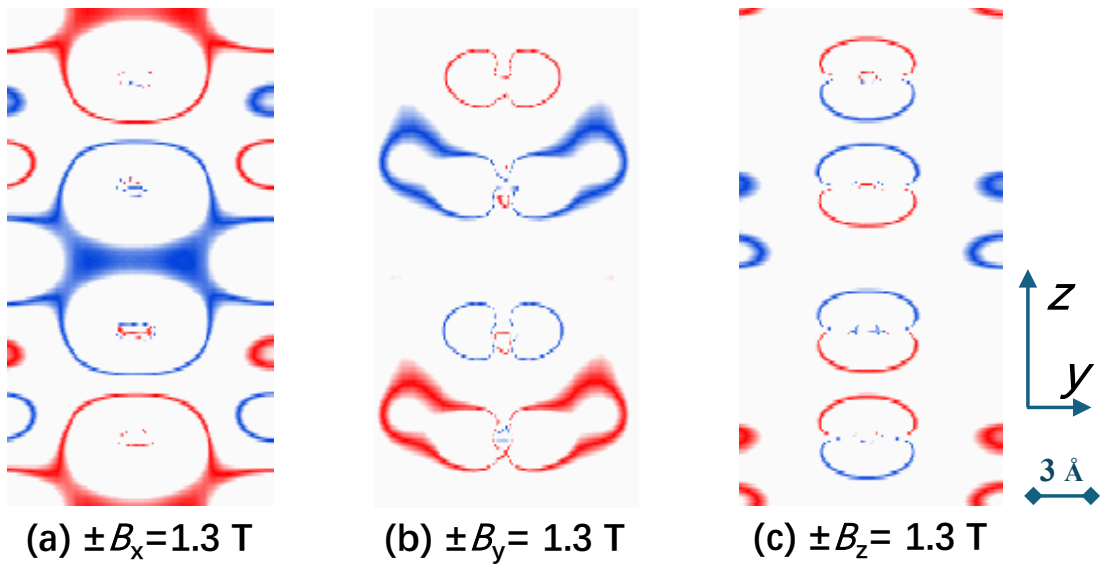


Fig.3.17 Magnetic field map corresponding to $1.3 \text{ T} \pm 25\%$ on the $x=0$ plane of the orthogonal unit cell: (a) B_x , (b) B_y , and (c) B_z . These fields originate from the x -component of the vector spin density and are depicted with red/blue coloring for positive/negative values.

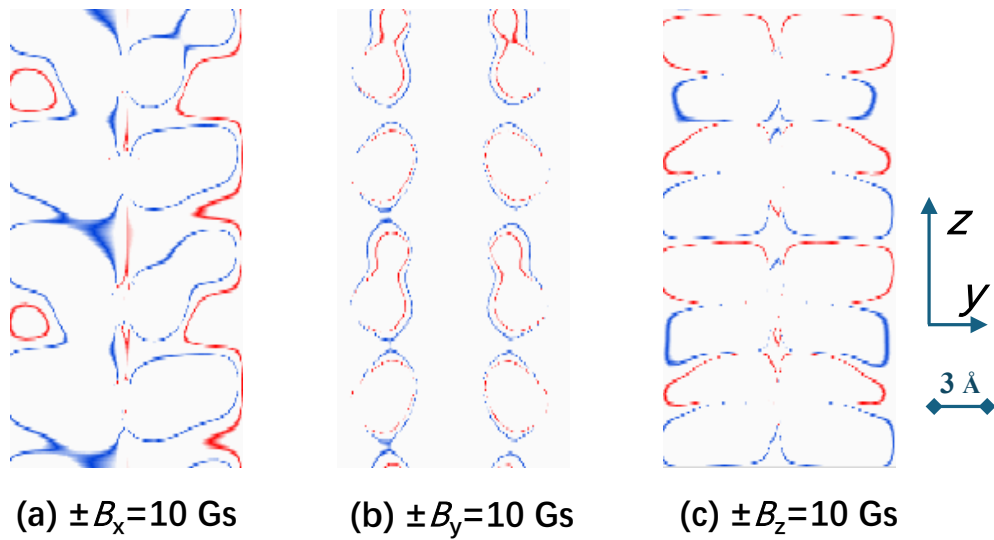


Fig.3.18 Magnetic field map corresponding to 10 Gs $\pm$ 25% on the x=0 plane of the orthogonal unit cell: (a) B_x , (b) B_y , and (c) B_z . These fields originate from the y-component of the vector spin density and are depicted with red/blue coloring for positive/negative values.

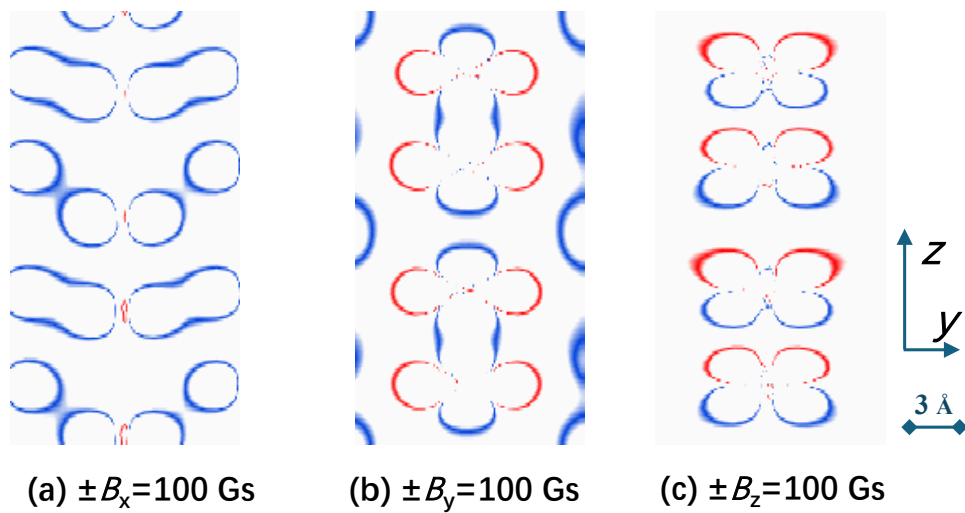


Fig.3.19 Magnetic field map corresponding to 100 Gs $\pm$ 25% on the x=0 plane of the orthogonal unit cell: (a) B_x , (b) B_y , and (c) B_z . These fields originate from the y-component of the vector spin density and are depicted with red/blue coloring for positive/negative values.

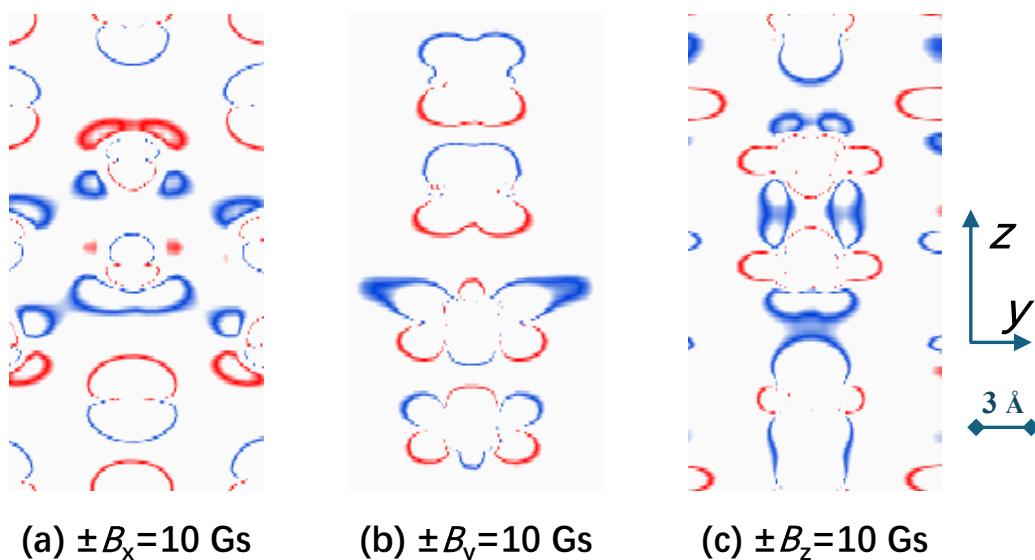


Fig.3.20 Magnetic field map corresponding to $10 \text{ Gs} \pm 25\%$ on the $x=0$ plane of the orthogonal unit cell: (a) B_x , (b) B_y , and (c) B_z . These fields originate from the z -component of the vector spin density and are depicted with red/blue coloring for positive/negative values.

4. Summary

To help readers clearly understand and reproduce results in the main text, this supplementary document provides a comprehensive guide to a computational workflow for processing and analyzing 3D vector fields, particularly in the context of magnetic field calculations. Theoretical discussions are given as guidance of DFT and molecular magnetism with practical steps for data preparation, field processing, and visualization. The workflow is supported by a suite of Labview packages, input files, and multimedia resources, making it a supporting resource of main text for researchers and practitioners in the field of computational magnetism in microscopic materials science.

X. Description of supplementary multimedia files

In addition to the current supplementary document, all other supplementary multimedia files have been packaged in a SOP-MM.zip file. Following is introduction of contained files:

./_sop-Manual_with_video-chain.pptx, main entrance of the operation. As demonstrated in section 2, this presentation includes step-by-step operation procedures for obtaining sub-lattice 3D vector magnetic fields. Key slices have been embedded a video for better reproducing.

./0-cif_BS-preparing, directory for step 0. For the reader's convenience, the *Fe2O3.cif* file and the *BASIS_BSE_pcseg-2_FeO* file are demonstrated for the crystallography and the pcseg-2 basis set of hematite. Readers can refer section 2.0, Fig.2.1, and slice 2~5 with a video in the .pptx file, if necessary.

./1-cp2k_inp/Fe2O3.inp, input file for demonstration of DFT calculation in CP2K. Readers can refer section 2.1, Fig.2.2 and slice 6 with a video in the .pptx file, if necessary.

./2.1-spin-density2Moment, directory for step 2.2, referring section 2.2.1, Fig.2.3, and slice 7 with a video in the .pptx file, if necessary.

alpha-Fe2O3-SPIN_DENSITY-1_0.cube, input file for transformation from spin

density to spin moment distribution (output file from CP2K and input for field calculation).

cubeReading.llb, Labview source file package for transformation from spin density to moments (Entrance from the vi program with the same name as the llb file).

MyDll.dll, a dll file of transformation program to read spin density file quickly.

./2.2-H-CalcScript-Generation, directory for step 2.2, referring section 2.2.2, Fig.2.4, and slice 8 with a video in the .pptx file, if necessary.

_xyzM.bin, input file for preparation of field calculation script (output file from step 2.1).

xyzM2H-CalcScript.llb, Labview source file package for transformation from moments to field calculation script (Entrance from the vi program with the same name as the llb file).

./3-H_Calc, directory for step 3, referring section 2.3, Fig.2.5, and slice 9 with a video in the .pptx file, if necessary.

xyzM.inp, input file for 3D vector field calculation (output file from step 2.2).

setting-Fe2O3.ini, should be copied together with *xyzM.inp*, with calculation settings of cut-off radius, assigned CPU threads, and calculation field range.

multiThreadYslices-FileInput.llb, Labview source file package for field calculation from 3D vector moment distribution (Entrance from the vi program with the same name as the llb file).

./ 4.1-Preparing-for-energy, directory for step 4.1, referring section 2.4.1, Fig.2.6, and slice 10 in the .pptx file, if necessary.

_M-TransCube.bin, input lattice information for preparing energy calculation (output file from step 2.1).

_xyzM.bin, input moment distribution file for preparing energy calculation (output file from step 2.2)

FileDataSliceVisualizing-preparing.llb, Labview source file package for preparing energy calculation (Entrance from the vi program with the same name as the llb file).

./ 4.2-FieldProcessing, directory for step 4.2, referring section 2.4.2, Fig.2.7, and slice 11 with a video in the .pptx file, if necessary.

dlt10k-cut16p-op.zip, input field distribution file package for 3D vector field processing including energy calculation (output file from step 2.2, need unzipping into a directory for usage).

_M-trueCube_9Lines.bin, input file of sorted moment for 3D vector field processing including energy calculation (output file from step 4.1).

BHMERging.llb, Labview source file package for 3D vector field processing including energy calculation (Entrance from the vi program with the same name as the llb file).

./ 5.1-StatisticalVision, directory for step 5.1, referring section 2.5.1, Fig.2.8, and slice 12 with a video in the .pptx file, if necessary.

_Ht_9-LineHeader.bin, input file of statistical analysis for 3D vector field processing results (output file from step 4.2, the given example indicates total magnetic field $\mathbf{H}_t$).

BinaCube-histoDemo.llb, Labview source file package of statistical analysis for 3D vector field processing results (Entrance from the vi program with the same name as the llb file).

./ 5.2-slice_by_sliceVision, directory for step 5.2, referring section 2.5.2, Fig.2.9, and slice 13 with a video in the .pptx file, if necessary.

_Bz_9-LineHeader.bin, input file of magnetic field component tomography for 3D vector field processing results (output file from step 4.2).

FileDataSliceVisualizing-1slicePresent.vi, Labview source file package of 3D vector tomography for magnetic field processing results (Entrance from the vi program with the same name as the llb file).

./ 5.3-isosurface3dVision, directory for step 5.3, referring section 2.5.3, Fig.2.10, and slice 14 with a video in the .pptx file, if necessary.

_Bx_9-LineHeader.bin, example file of $\mathbf{B}_x$ data for 3D isosurface visualization.

bin2cub.llb, Labview source file package of transformation binary field data into proper cube data for 3D isosurface visualization of magnetic field processing results (Entrance from the vi program with the same name as the llb file).

vmd.rc and *showcub.vmd*, controlling script of the 3D visualization tool for .cub file

reading, color and other settings (need to be copied into VMD main directory).

A flow chart of these procedures for generating microscopic magnetic fields is shown in Fig.X. These multimedia supplementary materials may not always be visible. To facilitate readers, related supplementary files can be accessed through the following link: <https://maglab.iphy.ac.cn/M00/cn/tools/pm3circuit.html> .

Just enjoy your trip in the sub-lattice magnetic world.

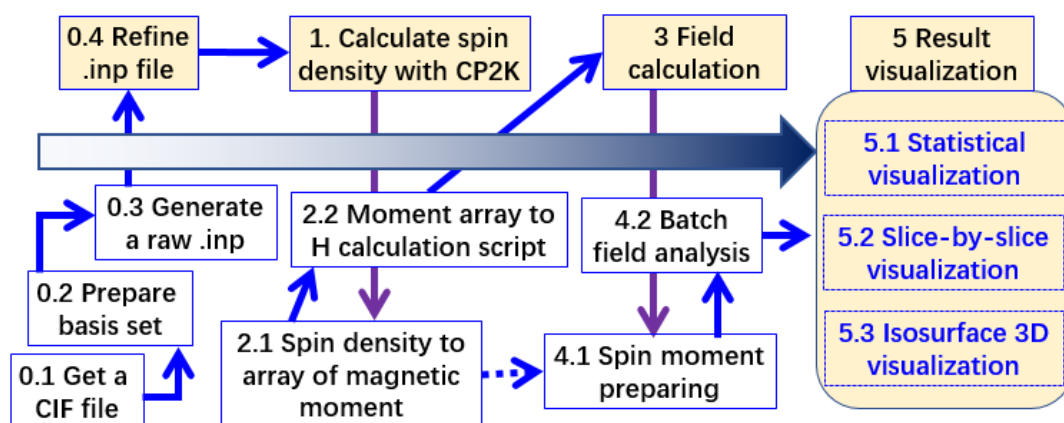


Fig.X Overview of main operational procedures in a flow chart

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