A new semi-Heusler ferromagnet NiFeSb: electronic structure, magnetism and transport properties

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Received 9 March 2003; received in revised form 8 May 2003; accepted 9 May 2003 by M. Cardona

Abstract

We report on structural, magnetic, transport, and spin polarization measurements of a new semi-Heusler alloy NiFeSb, which was synthesized successfully by means of electron melt-spinning technique although its ferromagnetic state and crystal structure was predicted to be unstable by the band calculation. The temperature-dependent measurement of magnetizations can both be interpreted well by the spin-wave theory. The electrical resistivity follows a $T^5$ behavior at low temperatures. Point contact Andreev reflection measurements of the spin polarization yield moderate polarization of 52%.

Keywords: D. Magnetism; D. Transport properties; D. Electronic structure; A. Heusler alloy

1. Introduction

Semi-Heusler phases XYZ are ternary compounds involving two different transition metals X and Y and one sp element Z, and crystallized in the C1\textsubscript{1} structure. During the last two decades, both experimental and theoretical investigations of the well-known Heusler systems \cite{1-3} delivered new and exciting results on their physical properties. It was demonstrated, using band structure calculations, the semi-Heusler NiMnSb and PtMnSb compounds belong to a new class of materials, called half-metallic ferromagnets \cite{4-10}. Half-metallic characteristics arise from a strong magnetic band splitting; with the property of an energy gap between valence and conduction bands for the electrons of one spin polarization and the property of continuous bands for the electrons of the other spin polarization. As a consequence we have the remarkable situation here that the conduction electrons at the Fermi level are 100% spin polarized at the Fermi level \cite{11}. Hence, possible applications for such exotica in spin-electronic devices are legion \cite{12}.

In this work, we replaced the Mn atoms of NiMnSb with Fe atoms and synthesized a new semi-Heusler compound NiFeSb by the melt-spinning technique, which is never reported before. The study mainly focused on its electronic structure, magnetic and transport properties. Measurements of the spin polarization by point contact Andreev reflection (PCAR) technique are also presented and discussed.
2. Computational and experimental details

The electronic structure of NiFeSb was calculated by linearized augmented plane-wave (LAPW) calculation within the local spin-density approximation (LSDA). Sixty k points in the irreducible Brillouin zone are used in the calculation. The self-consistency is better than 0.01 me/a.u.³ for charge density and spin density, and the stability is better than 0.1 mRy for the total energy per cell. The lattice constant of 5.72 Å was taken from our experimental data.

The samples studied in this paper were prepared by repeated melting of appropriately composed mixture of high-purity metals (Ni, Fe and Sb with 3N or better) in an arc furnace with argon atmosphere. The weight losses during melting were small. Ingots were homogenized by annealing in a vacuum-sealed quartz tube at 800 °C for 3 days and cooled down to room temperature. Some of the ingots were subsequently broken up and used for melt spinning. The melt-spun ribbons were prepared by a single wheel technique, under an Ar protection. The substrate velocity \( V_s \) of the Cu wheel was about 20 m/s. X-ray diffraction spectrum was taken to identify the crystallographic structure. Differential thermal analysis (DTA) was used to determine the phase transitions. The magnetization curves and the resistivity were measured using a commercial SQUID magnetometer.

3. Results and discussions

3.1. Electronic structure

The spin-projected DOS pictures for NiFeSb are shown in Fig. 1. There exists strong d–d hybridization between metal atoms. The hybridization of Ni d states with Fe d states leads to the formation of the splitting structures and results in the formation of an energy pseudo-gap, at about \(-0.7 \text{ eV}\) below the Fermi level instead of at the Fermi level, thus NiFeSb cannot show the half-metallic character, but the half-metallic property may be obtained with the alloying process just like reported by Ishida [13]. This work is underway. It is worth noting that the Fermi level is situated at a high-density region, according to Refs. [14,15], which makes the ferromagnetic state unstable. Tobola et al. indicated that, as a consequence of such DOS behavior, the total energy of system would be strongly augmented, which leads to the instability of the crystal structure [16]. As seen in Fig. 1, the formation and coupling of the magnetic moments of NiFeSb can also be interpreted schematically based on the Kübler model [17]. Table 1 shows some parameters correlatives with this band calculation.

3.2. Structure

As seen from Fig. 2(a), no sign of C1b structure can be found in precursor ingots, which is possibly composed of the mixture of Ni–Fe and (Ni0.5Fe0.5)Sb2 phases. On the other hand, the C1b structure in ribbons was confirmed evidently. Peaks indexed to the cubic phase included 200, 220, 222, 400, 420, and 422 reflections. The results are consistent with the prediction of the band calculation. The lattice parameter was found to be 5.72 ± 0.02 Å. DTA scans of the melt-spun sample showed a sharp melting peak at 1264 K and no structural phase transition has been found.

3.3. Magnetism

The \( M(T) \) curves were shown in Fig. 3(a). The magnetization \( M_s \) at 5 K is saturated in the magnetic field of about 2400 Oe. The spontaneous magnetization as a function of temperature is shown in Fig. 3(b). At low temperature the curve follows the expression \( M(T) = M(0)(1 - AT^n) \) with \( n = 1.5 \), the value expected from the Bloch’s law. The value of \( A \) is \( 2.508 \times 10^{-5} \text{ K}^{3/2} \). From the spin-wave theory we calculate the spin-wave stiffness.

Table 1

<table>
<thead>
<tr>
<th>Compound</th>
<th>s</th>
<th>p</th>
<th>d</th>
<th>f</th>
<th>( M(\mu_B) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>NiFeSb</td>
<td>Ni</td>
<td>0.195</td>
<td>0.190</td>
<td>4.280</td>
<td>0.007</td>
</tr>
<tr>
<td></td>
<td>Minority</td>
<td>0.202</td>
<td>0.212</td>
<td>4.014</td>
<td>0.008</td>
</tr>
<tr>
<td></td>
<td>Fe</td>
<td>0.182</td>
<td>0.161</td>
<td>4.314</td>
<td>0.008</td>
</tr>
<tr>
<td></td>
<td>Minority</td>
<td>0.155</td>
<td>0.138</td>
<td>1.900</td>
<td>0.011</td>
</tr>
<tr>
<td></td>
<td>Sb</td>
<td>0.616</td>
<td>0.740</td>
<td>0.062</td>
<td>0.020</td>
</tr>
<tr>
<td></td>
<td>Minority</td>
<td>0.602</td>
<td>0.772</td>
<td>0.070</td>
<td>0.021</td>
</tr>
</tbody>
</table>

\( m_{\text{heat}} = 2.670\mu_B \), \( N(E_F) = 4.037 \text{ states/eV cell} \), coefficient of electronic specific heat \( \gamma = 9.52 \text{ mJ/mol deg}^2 \), spin-polarization at Fermi level \( P = 72.3\% \).
coefficient \(D\) to be 144.1 meV Å\(^2\), using the equation
\[ A = \frac{2V}{S} \frac{k_B}{4\pi\lambda D} \frac{3}{2}, \]
where \(V\), the volume per magnetic atom, \(S\), the spin, and \(k_B\), the Boltzmann constant \[18\]. The value of the saturation magnetization at low temperature is 55.81 emu/g, corresponding closely to 2\(3.6\)\(\mu_B\). The small difference between the prediction of the band calculation and the experimental data is probably ascribed to the atom-disorder and the microstructure due to melt-spinning technique \[19,20\].

3.4. Transport properties

Fig. 4 shows the temperature dependence of resistivity. According to the Matthiessen’s rule \[21\], the separation of the total resistivity in its residual and its temperature-dependent part is usually as follows \(\rho = \rho_0 + \rho_L(T)\). By assuming the functional form \(\rho_L(T) = cT^n\) for our data, a plot of \(\ln(\rho - \rho_0)\) as a function of \(\ln(T)\) will yield a curve whose slope corresponds to the local power law parameter \(n\). The low temperature results \((T < 40 \text{ K})\) exhibits a \(T^n\) behavior with \(n \approx 4.91 \) (close to \(n = 5\)) corresponding to the familiar \(T^5\) law typical of simple intraband electron–phonon scattering at low temperatures \[21,22\]. At higher temperatures, the resistivity follows \(T^{2.42}\) and \(T^{1.36}\) behavior, respectively. A combination of electron–electron (proportional to \(T^2\)), electron–phonon (proportional to \(T\) at high temperatures) and electron–magnon scatterings can give a probable interpretation.

The spin polarization was measured at 2.8 K using the PCAR technique. The conversion of normal current to...
supercurrent at the metal/superconductor interface is called Andreev reflection. In the case of a spin-polarized metal, the imbalance between majority spin and minority spin conduction electrons can lead to a suppression of Andreev reflection. Thus, the spin polarization \( P \) of the metal may ideally be determined by analyzing the PCAR conductance curve. With this method the point contact tunneling conductance, \( G = \frac{dI}{dV} \), is interpreted by using the extended Blonde–Tinkham–Klapwijk (BTK) theory \([23, 24]\). Representative PCAR conductance curves for NiFeSb are shown in Fig. 5. Using a Nb point contact, fits to the PCAR conductance curves for NiFeSb yield a value for spin polarization \( P \approx 52\% \), which is smaller than the prediction of the band calculation. The most likely reason for this discrepancy is antisite disorder \([25,26]\), which is ascribed to the semi-Heusler structure. The empty lattice sites of the C1\text{b} structure prefers to be occupied, resulting in their being a dominant source of antisite disorder in the semi-Heuslers \([25]\). A study of the semi-Heusler PtMnSb finds an appreciable disorder of \(-10\% \) \([25]\).

4. Summary

The electronic structure calculation shows that there exists an energy pseudo-gap at about 0.7 eV below the Fermi level due to hybridization of Ni d states with Fe d states, hence NiFeSb is not a half metal. The Fermi level situates in the antibonding region, which would lead to an unstable ferromagnetic state and the structural instability. But it is worth noting from the XRD spectra that the C1\text{b} phase of NiFeSb can be synthesized successfully by the melt-spinning technique. The temperature-dependent measurement of magnetizations can both be interpreted well by the spin-wave theory. The electrical resistivity follows the familiar \( T^3 \) law typical of simple intraband electron–phonon scattering at low temperatures. Our measurements of a spin polarization \(-52\% \) for NiFeSb are lower than the prediction of the band calculation, which is probably due to the atom antisite disorder.

Acknowledgements

This work is supported by National Natural Science Foundation of China Grant No. 50201020.

References


Fig. 5. Conductance versus voltage curves using a Nb point contact at 2.8 K. In each graph, the corresponding spin-polarization \( P \), barrier strength parameter \( Z \), and contact resistance \( R \) is indicated. The solid line is the theoretical fit to the data using a modified Blond-–Tinkham–Klapwijk (BTK) model.