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LETTER TO THE EDITOR

Electronic structure and the metamagnetic transition of FeSi at extremely high magnetic fields

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Abstract. A field-induced moment for the narrow-band-gap semiconductor FeSi with a B20-type cubic structure is calculated by a self-consistent linear muffin-tin orbital method within the atomic sphere approximation, by introducing empty spheres in a unit cell. The fixed-spin-moment calculations give two successive metamagnetic transitions. It is found that the first metamagnetic transition will occur at a magnetic field of about 350 T from the nonmagnetic semiconductor to a metallic ferromagnet with a small moment of about 0.1 μ_B/Fe. This is consistent with the observed anomaly in the electrical conductivity at about 390 T. The second metamagnetic transition takes place from the small-moment state to a large-moment one with about 1 μ_B/Fe at about 700 T. The critical field of the first metamagnetic transition is shown to depend strongly on the value of the band-gap where the Fermi level lies.

1. Introduction

FeSi with a B20-type cubic structure is a semiconductor with a narrow band-gap of about 50 meV [1–4]. The magnetic susceptibility of this compound shows an unusual temperature dependence with a broad maximum at around 500 K [2, 5, 6]. Various models have been proposed to explain the unusual behaviours of the temperature dependencies of the susceptibility and specific heat. Jaccarino *et al* [6] gave a very-narrow-band description for these phenomena. However, the density-of-states curve assumed by them was considered to be unrealistic. A spin-fluctuation model [7–10] and a Fermi liquid model [11] were also proposed to explain these unusual behaviours of FeSi. Moreover, this compound was suggested to be an insulator with strongly correlated electrons, where the band-gap is destroyed by the electron correlation [12–16]. However, Evangelou and Edwards [8] have shown in the spin-fluctuation theory that the fluctuating exchange fields destroy the band-gap of FeSi at finite temperature. There still exist some controversies as regards how to explain the anomalous temperature dependence of the resistivity of FeSi.

The early band calculations gave a wide band-gap of about 0.1–0.2 eV [17, 18]. However, Kulatov *et al* [19, 20] have recently obtained a narrow band-gap of about 30–40 meV by using the full-potential linear muffin-tin orbital method. They have shown that the optical conductivity calculated with the wavefunctions obtained in the band calculation is in good agreement with the one observed by them [3]. It has also been shown that the results of the first-principles pseudopotential plane-wave calculations of the lattice constant and bulk

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modulus of FeSi are in good agreement with the observed values [21]. As far as the ground-state properties of FeSi are concerned, the band calculations are found to be available, in this way.

Another important point is the field-induced metamagnetic transition (MT) of FeSi. As sharp peaks of the density of states (DOS) exist on both sides of the Fermi level, the MT from the nonmagnetic state to the ferromagnetic one may occur at a high magnetic field [22]. When the ferromagnetic state is stabilized, the Fermi level lies in the conduction band with the majority spin and in the valence band with the minority spin. In this case, the system becomes metallic. This is just the field-induced semiconductor–metal transition. To prove this point, Anisimov *et al* [23] have performed a band calculation for FeSi in the local density approximation with an additional Coulomb interaction parameter U, and found a first-order transition from the nonmagnetic semiconductor to the ferromagnetic metal. Their result without the additional interaction U shows that the MT from the nonmagnetic state to the ferromagnetic (metallic) state with about 1 μ_B/Fe occurs at an extremely high magnetic field. The critical field is about 3500 T.

On the other hand, Kulatov *et al* [19,20] have calculated the moment of FeSi induced by the magnetic field. They have found that the MT from the nonmagnetic state to the ferromagnetic state with a small moment of about 0.2 μ_B/Fe occurs at a magnetic field of about 170 T. Subsequently, Kulatov *et al* [20] found that, as the field increases, the magnetic moment grows rapidly up to about 1 μ_B/Fe, even after the MT occurs. These results differ very considerably from the one obtained by Anisimov *et al* [23]. Recently, by means of microwave measurements, an anomaly in the electrical conductivity was found at an extremely high magnetic field of 390 T [24, 25], which will mean the field-induced semiconductor–metal transition. It is noted that no sign of the MT has been observed at magnetic fields up to 120 T [26]. Quite recently, Kudasov *et al* [27] observed a MT at about 350 T.

The critical fields of the MT depend strongly on the value of the band-gap. The calculated results for the MT [19, 20, 23] differ greatly from each other. The band-gap calculated by Anisimov *et al* [23] is, in fact, rather large, about 0.2 eV. On the other hand, Kulatov *et al* [19,20] obtained a band-gap of 30–40 meV. In this letter, we show the magnetization curve for FeSi with the observed value of the band-gap, by performing fixed-spin-moment (FSM) calculations by the self-consistent linear muffin-tin orbital (LMTO) method and using the atomic sphere approximation (ASA).

2. Details of the calculation

In our LMTO-ASA calculation of the electronic structure for FeSi, the basis set of wave-functions with angular momenta up to $l = 3$ has been adopted for the Fe and Si atoms. The spin–orbit interaction was not included but the calculation was scalar relativistic, including mass–velocity and Darwin terms. Self-consistent calculations are carried out at more than 176 $\boldsymbol{k}$-points in the irreducible 1/24 Brillouin zone. The convergence in charge density was achieved in such a way that the root mean square of the moments of the occupied partial density of states became smaller than 10^{-6}. The exchange–correlation potential given by von Barth and Hedin [28] was made use of.

The observed value of the band-gap for FeSi can be obtained by introducing four empty spheres with the atomic number $Z = 0$ in the unit cell. Without these empty spheres, the packing fraction is 56% if the space is filled up by touching rigid spheres of Fe and Si. Here, the radii of the atomic spheres of Fe and Si are assumed to be equal. Four Fe atoms are located at the positions (u, u, u), $(-u, 1/2+u, 1/2-u)$, $(1/2-u, -u, 1/2+u)$ and $(1/2+u, 1/2-u, -u)$ and four Si atoms are at $(-u, -u, -u)$, $(u, 1/2-u, 1/2+u)$, $(1/2+u, u, 1/2-u)$ and

$(1/2 - u, 1/2 + u, u)$ in units of the lattice constant, where u is the internal lattice parameter 0.3525. When four empty spheres are put at the positions (0, 0, 0), (0, 1/2, 1/2), (1/2, 0, 1/2) and (1/2, 1/2, 0) in the unit cell, the packing fraction becomes 62%. In this case, an empty sphere touches three Fe and three Si atoms.

For the ASA, the radii of the atomic spheres should be expanded so that the total volume of the atoms coincides with the unit-cell volume. However, there are ambiguities as regards how to expand each 'atomic' radius including those of the empty spheres. Figures 1(a), 1(b) and 1(c) show the local DOSs of Fe (solid curve) and Si (broken curve) calculated with the ratios $r_{ES}/r_{Fe} = 0.6035$, 0.55 and 0.5077, respectively, where r_{ES} and r_{Fe} are the radii of the empty atomic spheres and of the atomic spheres of the Fe atoms. Here, we assumed that the atomic radii of Fe and Si are equal. The observed value of the lattice constant is used in the present calculations. The ratio between r_{ES} and r_{Fe} in figure 1(a) is obtained such that the radii of all 'atoms' including empty spheres are expanded in the same ratio. On the other hand, the ratio r_{ES}/r_{Fe} in figure 1(c) is determined such that the radii of Fe and Si are expanded in the same ratio, but the radius of the empty sphere is kept as that of the touching sphere. The observed band-gap is obtained for the ratio $r_{ES}/r_{Fe} = 0.55$, between those used for figures 1(a) and 1(c). The local DOS for this ratio is shown in figure 1(b). For the sake of comparison, the local DOS without empty spheres is shown in figure 1(d). Figure 2 shows the local DOS calculated for Fe, Si and the empty sphere ES with the ratio $r_{ES}/r_{Fe} = 0.55$ over a wider energy region.

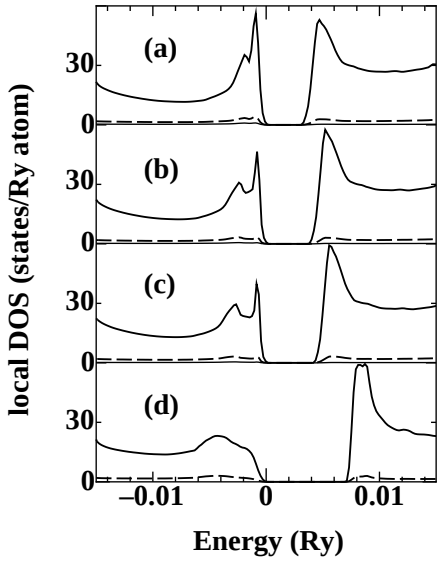

Figure 1. The calculated local DOS for Fe and Si shown by solid and broken curves. The curves in (a), (b), (c) and (d) are those calculated with the ratios $r_{ES}/r_{Fe} = 0.6035$, 0.55, 0.5077 and 0, respectively.

3. Results and discussion

The calculated values of the band-gap shown in figures 1(a), 1(b), 1(c) and 1(d) are 3.17, 3.87, 4.29 and 7.0 mRyd, respectively. It is found that the band-gap decreases as the ratio r_{ES}/r_{Fe} increases. The observed band-gap of about 50 meV (3.7 mRyd) is obtained at the ratio $r_{ES}/r_{Fe} = 0.55$, as shown in the previous section. The FSM calculations are carried out

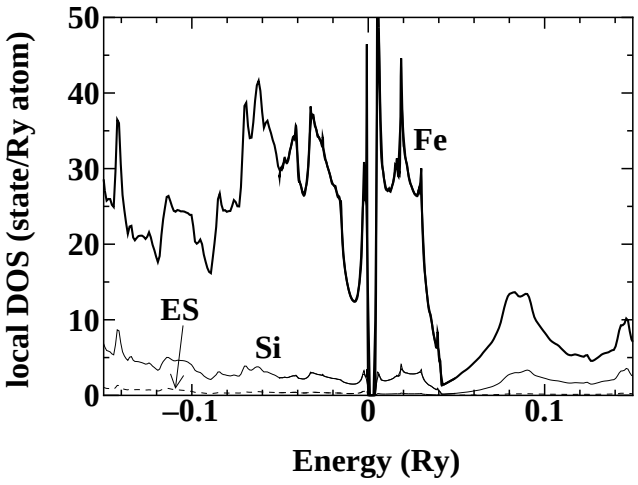

Figure 2. The calculated local DOS for Fe, Si and the empty sphere ES shown by the solid, thin and broken curves, respectively. The position of the Fermi level E_F is in the narrow band-gap.

for this ratio r_{ES}/r_{Fe}. The numbers of electrons with majority and minority spins, $N_\uparrow$ and $N_\downarrow$, are given by $(N + M)/2$ and $(N - M)/2$ where N and M are the number of electrons and the spin moment divided by μ_B. Under the constraint of fixed values of $N_\uparrow$ and $N_\downarrow$, the electron densities $n_\uparrow(r)$ and $n_\downarrow(r)$ and the potentials for electrons may be calculated by using the usual Kohn–Sham self-consistent equations, which give two Fermi levels, $\mu_\uparrow$ and $\mu_\downarrow$, in the majority- and minority-spin bands, respectively. For an arbitrary M, the system is not in the equilibrium state, so $\mu_\uparrow \neq \mu_\downarrow$. However, the constrained state becomes an equilibrium at the magnetic field $H = (\mu_\uparrow - \mu_\downarrow)/2\mu_B$. This means that the state with the given M is stabilized at this magnitude of H. That is, M is obtained as a function of H [29, 30].

The closed circles in figure 3 show the calculated values of $\mu_\uparrow$ and $\mu_\downarrow$ as functions of the magnetic moment M. At small values of M, it takes a long CPU time to get the self-consistent

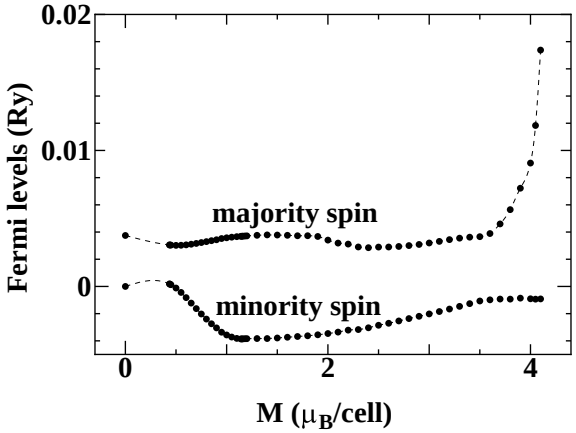

Figure 3. The Fermi levels in the majority- and minority-spin bands, $\mu_\uparrow$ and $\mu_\downarrow$, calculated in the FSM as a function of the magnetic moment M.

solutions. However, $\mu_\uparrow$ and $\mu_\downarrow$ at the limit of $M = 0$ are the bottom of the upper conduction band and the top of the lower valence band, respectively. By using the method of least mean squares, $\mu_\uparrow$ and $\mu_\downarrow$ are obtained at very small values of M as shown by the broken curve in figure 3.

The open circles in figure 4 show the magnetization curve obtained from figure 3 and the relation $\mu_\uparrow - \mu_\downarrow = 2\mu_B H$. In the regions of H between about 300 and 450 T and between 550 and 900 T, the calculated value of M is a triple-valued function of H and the state with an intermediate value of M is unstable. The states with larger and smaller values of M are stable or metastable. The critical field H_C of the MT is estimated from the Maxwell relation:

$$\int_{M_1}^{M_2} H(M)\,\mathrm{d}M - (M_2 - M_1)H_C = 0$$

where M_1 and M_2 are smaller and larger moments for the stable or metastable states [22]. The curve in the region of very small M in figure 4 is estimated from the broken curve in figure 3. The first MT may occur at 354 T from the nonmagnetic state to a small-moment state, although this is not an actually calculated value. The present result is very close to the observed value for the MT at extremely high magnetic field [27]. The second successive MT occurs at 718 T from the small-moment state to a large-moment one. The first MT is similar to that obtained by Kulatov *et al* [19, 20]. However, the hysteresis of the MT is rather small, compared with that obtained by Kulatov *et al*. This is because our calculated band-gap is a little wider than that obtained by them and the critical field depends strongly on the band-gap. On the other hand, Anisimov *et al* [23] have obtained a MT from the nonmagnetic state to a large-moment state, not to a small-moment one. This is probably because their calculated band-gap is much wider than the band-gap being discussed.

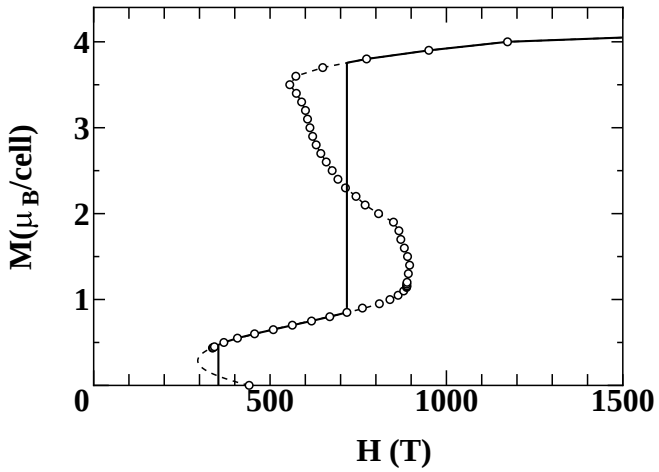

Figure 4. The calculated magnetization curve. The critical fields of the MTs are estimated to be about 300 and 750 T.

In order to see the dependence of the critical fields of the MTs on the value of the band-gap, we have carried out the same calculation for the case of $r_{ES}/r_{Fe} = 0.6035$ (figure 1(a)) where the band-gap is 3.17 mRyd, a little smaller than the observed one. The calculated values of the critical fields are 257 and 703 T, respectively. The critical field of the first MT is thus found to be very sensitive to the value of the band-gap, while that of the second MT does not depend so much on the value of the band-gap. This is because the band-gap is caused by the

hybridization [17, 20], and the shape of the DOS just above and below the band-gap is very sensitive to the value of the band-gap, as shown in figure 1. The shape of the DOS except near the band-gap is not sensitive to the value of the band-gap, so the critical field for the second MT from the small-moment state to the large-moment one does not depend so much on the band-gap.

In this letter, we have shown by performing FSM calculations in the LMTO-ASA that successive MTs may occur from the nonmagnetic state to a small-moment one and from the small-moment state to a large-moment one, by introducing empty spheres to get the observed band-gap. The critical field of the first MT was obtained as about 350 T. At magnetic fields higher than that, the narrow-gap semiconductor FeSi will show a metallic behaviour. The magnetic field 390 T, at which the anomaly in the electrical conductivity was observed for FeSi [24, 25], is very close to the critical field of the first MT described in the present letter. Therefore, the system becomes metallic at this field. In order to show this explicitly, the DOS calculated at 342 T is shown in figure 5, as a representative of the small-moment state obtained after the first MT takes place. It is seen that the Fermi level E_F lies in the conduction band with the majority spin and in the valence band with the minority spin. In this way, FeSi becomes metallic at the critical field of the first MT.

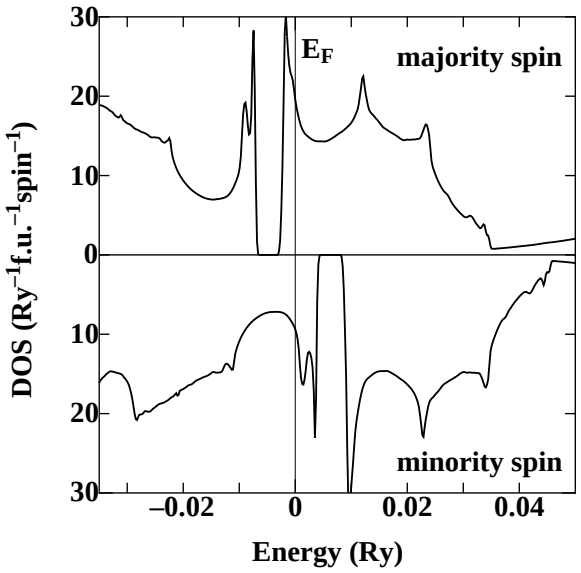

Figure 5. The calculated DOS at the magnetic field 342 T. E_F denotes the Fermi level.

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