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Thermopower in the regime of strong Hubbard correlations in FeSi

N. E. SLUCHANKO¹, V. V. GLUSHKOV¹, S. V. DEMISHEV¹, M. V. KONDRIN¹, K. M. PETUKHOV¹(*), N. A. SAMARIN¹, V. V. MOSHCHALKOV² and A. A. MENOVSKY³

 Low Temperatures Laboratory, General Physics Institute RAS Vavilov str., 38, 117942 Moscow, Russia
 Laboratorium voor Vaste-Stoffysica en Magnetisme, Katholieke Universiteit Leuven Celestijnenlaan 200 D, B-3001 Leuven, Belgium
 Van der Waals-Zeeman Laboratory, University of Amsterdam Valckenierstraat 67, 1018 XE Amsterdam, The Netherlands

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Abstract. – The Seebeck coefficient anomalies in the so-called Kondo-insulator compound FeSi have been studied at liquid helium and intermediate temperatures. It is shown that the most complete and consistent interpretation of the iron monosilicide magnetic, transport and optical properties can be achieved in the framework of the Hubbard model. The estimation of microscopic parameters provides additional arguments in favour of the spin polarons formation and dramatic renormalization of the intragap electronic density in FeSi at low temperatures.

In spite of the growing interest in the cubic transition metal compound FeSi, the unusual physical properties of this narrow-gap semiconductor have not yet received an adequate explanation [1–9]. Different models have been used to interpret the low-temperature anomalies of the electronic and magnetic properties of iron monosilicide (Kondo-insulator model [1–7], d-p valence fluctuations [9] etc., see, e.g., [8]). However, most of them are inconsistent with the results of band structure calculations [10–14] where a high electronic density of states is found in the vicinity of an indirect gap ($E_g \approx 60 \text{ meV}$) in this almost magnetic narrow-gap semiconductor. According to [10–14] the FeSi Fermi level lies in the Fe 3d bands and the density of states (DOS) on both sides of the gap is dominated by the Fe 3d states which are only slightly (about 10%) hybridized with the Si 3p electrons.

In this letter we present experimental arguments and estimations in favour of the formation of the spin-polaron Mott metallic state with strong Hubbard correlations in iron monosilicide at low temperatures ($T < 100 \,\mathrm{K}$). In our opinion, this approach is very useful for understanding the nature of the low-temperature anomalies [1–9] and a coherent ground state formation in this unusual narrow-gap semiconductor.

^(*) Present address: 1 Physikalisches Institut, Universität Stuttgart, D-70550 Stuttgart, Germany.

To test the effects of Hubbard correlations and spin-polaron states formation the "key experiment" is the study of the Seebeck coefficient S(T) [15, 16]. Thermopower of FeSi was investigated earlier in [6,7], where the S(T) features have been recorded in the temperature interval T < 200 K and were attributed to the phonon drag effects. However, very low values of charge carrier mobility (μ_n , $\mu_p < 10 \text{ cm}^2/\text{V} \cdot \text{s}$) deduced from transport measurements for high-quality single crystals of FeSi [8] are in contradiction with the phonon drag interpretation of the low-temperature S(T) anomalies. Indeed, a strong electron-phonon scattering and spin fluctuations should suppress the non-equilibrium effects in the phonon distribution function of FeSi resulting in a significant slacking of phonon drag thermopower (see, *e.g.*, [17]). A flat and extended top of the low-temperature S(T) maximum in FeSi, which is not typical for phonon drag thermopower in metals and semiconductors, provides additional arguments against the interpretation of the Seebeck coefficient behaviour proposed in [6,7].

To clarify this point, the detailed measurements of Seebeck coefficient of FeSi single crystals are presented in this letter together with the quantitative analysis of the experimental data in combination with the previously reported results on magnetic, transport and optical properties and within the framework of the Hubbard model. The original technique of precision thermopower measurements was described elsewhere [18]. The samples of iron monosilicide are high-quality single crystals with a high resistivity ratio $\rho(T \rightarrow 0)/\rho(300 \text{ K}) \approx 5 \cdot 10^5 - 10^6$ and a small amount of magnetic impurities [19]. The cubic crystal symmetry of FeSi, a distorted rocksalt structure, was confirmed by X-ray diffraction. Electron-probe microanalysis showed a stoichiometric and homogeneous single phase.

A typical experimental S(T) curve obtained for one of the FeSi samples is given in fig. 1a. The temperature range $T > 100 \,\mathrm{K}$ corresponds to the intrinsic conduction region of this narrow-gap semiconductor [8], so the temperature dependence of thermopower can be described by the equation

$$S_1(T) = \frac{k_{\rm B}}{e} \left\{ \frac{b-1}{b+1} \frac{E_{\rm g}}{2k_{\rm B}T} + A \right\} \,, \tag{1}$$

where $b = \mu_{\rm n}/\mu_{\rm p}$ is the ratio of electron to hole mobility, $k_{\rm B}$ is Boltzmann's constant, e the the charge of electron and A the kinetic coefficient [15–17]. The parameter b of eq. (1) $b = \mu_{\rm n}/\mu_{\rm p} \approx 1.5$ was evaluated from the experimental curve of fig. 1 (100 K < T < 300 K), we also used the S(T) results [6] for the interval 300–700 K and the value $E_{\rm g} \approx 60 \,\mathrm{meV}$ [8].

On the other hand, the low-temperature contribution S_2 in S(T) (see fig. 1a) is determined by intra-gap states and does not depend on the temperature $S(T) = S_2 \approx \text{const} \approx 900 \,\mu\text{V/K}$ in the range 15 K < T < 50 K. At intermediate temperatures 60-120 K (fig. 1a) conductivity $\sigma(T)$ and thermopower S(T) can be described with the help of a simple model which takes into account the sum of the contributions from both the charge carriers excited through the indirect gap (σ_1 , S_1) and from the intra-gap states (σ_2 , S_2):

$$\sigma(T) = \sigma_1(T) + \sigma_2(T) = \sigma_{01} \exp[-E_{\rm g}/2k_{\rm B}T] + \sigma_{02} \exp[-E_{\rm ex}/k_{\rm B}T], \qquad (2)$$

$$S(T) = \frac{\sigma_1(T)S_1(T) + \sigma_2(T)S_2(T)}{\sigma_1(T) + \sigma_2(T)}.$$
(3)

When eqs. (1)-(3) are applied to analyse the Seebeck coefficient behaviour, an exponential asymptotic curve is expected for the difference $S_d(T) = S(T) - S_1(T)$ at intermediate temperatures 60–120 K:

$$S_{\rm d}(T) = S(T) - S_1(T) \approx \frac{\sigma_{01}}{\sigma_{02}} \exp\left[\frac{E_{\rm g}/2 - E_{\rm ex}}{k_{\rm B}T}\right] S_2(T) \,.$$
 (4)



Fig. 1 – Temperature dependence of a) Seebeck coefficient S(T), b) mobility difference $R_{\rm H}\sigma = \mu_{\rm n} - \mu_{\rm p}$ and c) magnetic susceptibility $\chi(T)$ in FeSi.

Fig. 2 – Activation behaviour of a) parameter $S_d(T) = S(T) - S_1(T)$ (see text) and b) conductivity $\sigma = \sigma_1 + \sigma_2$. The temperature intervals I and II correspond to intrinsic conduction ($T \ge T^* \approx 70 \text{ K}$) and intra-gap states transport in FeSi.

The $S_d(T)$ parameter, as obtained from experimental results of fig. 1a, is shown in fig. 2 together with different conductivity contributions (σ_1, σ_2). An activation behaviour of the $S_d(T)$ with characteristic energy $E_{Sd} \approx 407 \text{ K}$ is clearly seen which is closely related to the indirect gap value $E_g/2 \approx 30 \text{ meV}$. This implies that the drastic changes of S(T) in FeSi at intermediate temperatures are mainly determined by the temperature dependence of conductivity in the intrinsic conduction range and are not connected with the phonon drag effects.

It is interesting to note the presence of the saturation of $S(T) \approx S_2(T) \approx \text{const}$ in the temperature interval 15–50 K, where $S_2(T)$ changes only slightly within the range 880– 920 μ V/K (fig. 1a). In such a situation, when the activation behaviour of the Hall coefficient and conductivity in FeSi at low temperatures T < 70 K is accompanied by thermopower saturation $S(T) \approx \text{const}$, an adequate description of the Seebeck coefficient can be obtained by using Heikes's formula

$$S_2(T) = -\frac{k_{\rm B}}{|e|} \ln\left(\frac{1-\nu}{\nu}\right) \,,\tag{5}$$

where $\nu = N/N_{\rm Fe}$ is the reduced carrier concentration in Hubbard bands. Equation (5) corresponds to the thermopower behaviour in the regime of the strong on-site Coulomb (Hubbard) correlations [15, 16] and can be applied to estimate the charge carrier (hole) concentration in the lower Hubbard band. Taking the value of FeSi volume density $6100 \, \rm kg/m^3$ and $S_2(T) = \rm const \approx +920 \, \mu V/K$ we have $N_{\rm Fe} \approx 2.2 \cdot 10^{22} \, \rm cm^{-3}$, $\nu \approx 1.3 \cdot 10^{-5}$ and



Fig. 3 – a) Schematic DOS for the one-band Hubbard model with $2 \le U/D \le 3$ at different temperatures (taken from refs. [19–21]) and its corresponding optical spectra $\sigma(\omega)$ b) as calculated in [19] for a metallic side of the Mott-Habbard transition $U/D \le 3$ and c) as deduced in [22] from reflectivity measurements of FeSi at different temperatures.

 $p \approx 5.8 \cdot 10^{17} \,\mathrm{cm}^{-3}$. It is worth mentioning that these low ν values allow us to reduce eq. (5) to a very simple relation $S_2(T) \approx \frac{k_{\mathrm{B}}}{|e|} \ln \nu$ which is consistent with the thermopower expression for the spin polaron transport [15, 16]. According to [15] in the regime of strong Hubbard correlations the charge carriers are spin polarons. For the case of almost magnetic semiconductor FeSi, these heavy quasiparticles may be supposed to appear as a result of a strong polarization of the Fe magnetic moments located in the nearest environment of the charge carrier in the Hubbard band.

To clarify the origin of the spin polaron states, we shall rely upon the recent results of DOS and optical conductivity $\sigma(\omega)$ calculations in the framework of the one-band Hubbard model [20] (see also [21,22]). As was shown in [20] by the local impurity self-consistent approximation in the limit of a large number of spatial dimensions, the metal-insulator transition (MIT, Mott-Hubbard type) takes place at an intermediate value of the on-site Coulomb interaction $U_c \approx 3D$ (D is the band half-width). Moreover, the metallic ($U < U_c$) side is characterised by a DOS with a three-peak structure: the central feature at the Fermi energy that narrows as one moves toward U_c from below, and two broader incoherent features that develop at $\pm U/2$, namely the lower and upper Hubbard bands (see fig. 3a). In fig. 3a the evolution of the central peak $\rho(\omega)$ is sketched also as a function of T (see [22]): the width of this resonance does not vary much, but $\rho(0)$ decreases dramatically. When $T = T^* \approx \delta$ (δ is the width of the resonance) the peak has a vanishing weight instead of being broadened [21, 22]. Therefore, at temperatures around T^* , the transition to the narrow-band transport representing a manybody quasi-particle resonance can be expected for $U \leq U_c$ instead of a Mott transition to the dielectric state [20].

In the framework of the approach developed in [20–22] it is natural to interpret the activation behaviour of the Hall coefficient [8] and conductivity (fig. 2b) in FeSi at $T < T^*$ in combination with S(T) = const (fig. 1a) in terms of the transport of charge carriers through the polaronic states at $E_{\rm F}$ [15, 16]. In these terms the potential of the spin-polaron states $E_{\rm ex}$ can be applied here to estimate the central resonance width $\delta \approx E_{\rm ex} \approx 6$ meV. The renormalized density of states $\rho(\omega)$ (fig. 3) and optical conductivity $\sigma(\omega)$ (fig. 3b) also calculated in [20] for the one-band Hubbard model allow us to analyse the low-temperature optical spectra $\sigma(\omega)$ measured for FeSi in a wide IR-range (fig. 3c) [23]. From the comparison of the position and width of the $\sigma(\omega)$ maximum at $\omega = U/2$ (see fig. 3b and c) it is easy to extract the following parameters of the Hubbard model in FeSi: $U \approx 0.27 \,\text{eV}$, $2D \approx 0.21 \,\text{eV}$, $E_{\rm g} = U - 2D \approx 60 \,\text{meV}$ and $U/D \approx 2.6$. It should be noted that the gap value $E_{\rm g} \approx 60 \,\text{meV}$ and the magnitude of parameter $U/D \approx 2.6$ are in a good agreement with both experimental data for $E_{\rm g}$ and the theoretical values of parameters which determine the instability region of this dielectric state in the Hubbard model [20–22].

The above-mentioned renormalization of the DOS and the transition to the narrow-band transport can be formulated also in terms of a substantial increase of the effective mass of charge carriers. The increase should be seen in galvanomagnetic and magnetic properties of FeSi at low temperatures. We think that the drastic changes of the Hall mobility difference $\mu_{\rm p} - \mu_{\rm n} = R_{\rm H}\sigma$ ($R_{\rm H}$ is the Hall coefficient) (fig. 1b, [8]) can be attributed to the formation of these "heavy fermions" in the temperature range $T \leq T^* \approx 70 \,\mathrm{K}$ in FeSi. Moreover, the magnetic susceptibility increase at the same temperatures $T \leq T^*$ (see fig. 1c) reflects apparently the effects of the DOS renormalization at $E_{\rm F}$. Indeed, when analysing the low-temperature magnetic susceptibility in FeSi as Pauli paramagnetic contribution from the charge carriers in the narrow rectangular (for the approximation) conduction band of width δ at $E_{\rm F}$,

$$\chi_{\rm P}(T) = 1/2g^2 \mu_{\rm B}^2 N^*(E_{\rm F}) \int_{-\delta/2}^{\delta/2} \left(-\frac{\partial f}{\partial E}\right) \mathrm{d}E \tag{6}$$

(g is the g-factor and $\mu_{\rm B}$ is the Bohr magnetic moment), we have the following evaluation for the amplification factor: $\chi_{\rm p}(T = 4.2 \,\mathrm{K})/\chi_{\rm p}(T = 100 \,\mathrm{K}) \geq 100$ (see fig. 1c). According to the arguments of [24] the integral in eq. (6) gives an increase factor of about 4–5, so the real DOS renormalization with temperature in FeSi can be estimated as $N^*(E_{\rm F}, T = 4.2 \,\mathrm{K})/N^*(E_{\rm F}, T =$ $100 \,\mathrm{K}) \geq 20$. This interpretation is also suitable to explain the so-called Curie-Weiss–like rise at low temperatures in FeSi samples mentioned previously (see, *e.g.*, [25]). It should be noted that the independent conclusion about an essential DOS renormalization in FeSi in the vicinity of indirect gap in the 3*d*-band was also deduced from tunnelling spectra [19] and high-resolution angle-resolved photoemission data [26]. The resonance picture is in agreement with the general classification schema of Kondo systems [27].

Within the framework of our approach it is possible to evaluate an effective mass $m^*(T \approx 200 \text{ K})$ of carriers in the FeSi Hubbard bands at temperatures above the MIT with the help of relation $m^* = e\langle \tau_{e-ph} \rangle / \mu_{n,p} (\langle \tau_{e-ph} \rangle$ is the relaxation time). For the estimation of $\langle \tau_{e-ph} \rangle$ the value of the polarised neutrons scattering linewidth $\Gamma \approx \hbar / \langle \tau_{e-ph} \rangle \approx 3-4 \text{ meV}$ [28] can be used giving $\langle \tau_{e-ph} \rangle \approx 1.7 \cdot 10^{-13} \text{ s}$. The similar magnitude of $\langle \tau_{e-ph} \rangle$ can be obtained from the linewidth of optical phonons [23]. As a result, taking $\mu_{n,p} \approx 4-6 \text{ cm}^2/\text{V} \cdot \text{s}$ [8], one can find the effective mass of charge carriers in the intrinsic conductivity region $m^*(T \approx 200 \text{ K}) \approx 50m_0$.

For quantitative estimations of the radius of the short-distance spin polaronic states in the FeSi cubic lattice, the effective mass and the activation energy of conductivity and the Hall coefficient $E_{\rm ex} \approx \delta \approx 6 \,\mathrm{meV}$ can be inserted into the expression $a_{\rm p} = \hbar/\sqrt{2E_{\rm ex}m^*(200\,\mathrm{K})}$ that gives $a_{\rm p} \approx 3.5\,\mathrm{\AA}$. On the other hand, the $a_{\rm p}$ value in the vicinity of the Mott-Hubbard metal-

insulator transition can be calculated by using the formula $a_{\rm p} \approx (2\pi U/2D)^{1/5} \approx 1.75a \approx 4.8 \text{ Å}$ [15] (the value of $a = r_{\rm Fe-Fe} \approx 2.75 \text{ Å}$ is equal to the shortest Fe-Fe distance in a cubic crystalline structure of FeSi). Both independent evaluations of the localisation radius $a_{\rm p}$ are in good agreement, thus giving additional arguments in favour of the proposed interpretation. To develop further the present approach, the magnetic phase transition in FeSi near $T_{\rm c} \approx 8 \text{ K}$, which was detected earlier in [8], can be attributed to a magnetic ordering of the short-distance spin polarons in the regime of a low charge carriers concentration ($5.8 \cdot 10^{17} \text{ cm}^{-3}$). However, full description of the nature of the FeSi ground state requires additional measurements of quasioptical spectra in the far infra-red region ($< 50 \text{ cm}^{-1}$), in combination with precision measurements of the magnetic properties of iron monosilicide single crystals at liquid-helium temperatures.

In summary, we have found that the Seebeck coefficient anomalies in the so-called Kondoinsulator compound FeSi at liquid-helium and intermediate temperatures can be attributed to the transition from an intrinsic conduction behaviour to the regime of strong Hubbard correlations in this unusual narrow-gap semiconductor. It has been shown that the most complete and consistent interpretation of the iron monosilicide magnetic, transport and optical properties can be given in the framework of the one-band Hubbard model.

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