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Low temperature magnetic H-T phase diagram of FeSi

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Abstract

The low-temperature magnetic H-T phase diagram of iron monosilicide is reconstructed from the analysis of magnetotransport, magnetooptical and magnetic characteristics measured for FeSi single crystals in a wide range of temperatures (1.8-40K) and magnetic fields (up to 120 kOe). The obtained results have been also used to estimate the effects of exchange interaction and to determine the microscopic parameters of correlated electrons in low temperature paramagnetic ($T>T_C$=15K), mictomagnetic ($T<T_m$=7K) and ferromagnetic ($T_m<T<T_C$) phases of FeSi. A set of transport and magnetic features as well as anomalous magnetooptical absorption observed in the vicinity of H^*~35 kOe allowed us to conclude in favor of a field induced magnetic transition to collinear phase in magnetic fields $H>H^*$ at $T<T_m$.

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Anomalous magnetic and transport properties of the almost magnetic narrow gap (ε_g~60 meV) semiconductor FeSi stay the subject of debates during last five decades [1-9]. The most known anomaly which was firstly noted by Benoit [1] is the non-monotonous high temperature behavior of magnetic susceptibility observed in the absence of long range magnetic ordering [2,3] (Fig.1a). The commonly used classification treats iron monosilicide as the only transition metal based Kondo insulator [4]. However, this approach which ascribes the unusual properties to a spin gap formation from Kondo compensation of magnetic moments provides no consistent picture for low temperature anomalies observed in transport and optical properties. For another hand, this compound is one of the most striking examples of the successful application of spin fluctuation theory [5], which explains the enhancement in terms of temperature induced localized magnetic moments at Fe centers in the FeSi matrix at $T>$ 100K. At the same time this model gives no microscopic explanation for the low temperature anomalies of FeSi (Fig.1) which could have intrinsic origin rather than extrinsic one [6].

It has been recently shown [7-9] that a consistent interpretation of low temperature anomalies in FeSi can be obtained in framework of Mott-Hubbard model for critical region of Coulomb interaction $2<U\leq 3D$ (D - band half-width) [10]. In this approach lowering the temperature below T^*~80K induces the transition from high-temperature semiconducting to

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low-temperature correlated metallic state. This transition results from the dramatic renormalization of density of states (DOS) with formation of the narrow (Δ_p~6 meV) many-body resonance in the middle of gap [7-9]. The states within the resonance can be associated with short range (<10Å) spin polarons with effective mass m^*~50-100m_0 which are formed on the charge carriers in the upper Hubbard band of FeSi [9]. So the low temperature anomalies of physical properties of this compound (see, *e.g.,* the saturation of resistivity below 3K shown in Fig.1b) should be mainly determined by transformations in the system of the heavy fermions arisen from fast electron density fluctuations on Fe-sites.

To obtain an adequate information about low temperature state of strongly correlated electron system, the study of transport, magnetic and magnetooptical characteristics has been carried out on FeSi single crystals in a wide range of temperatures (1.8-300K) and magnetic fields (up to 120 kOe). For reconstructing of the low temperature magnetic phase diagram, the present data have been analyzed together with previous results [7-9].

Before discussing of the low temperature magnetic phase diagram of FeSi note that the transition between high temperature insulating (**PI**, $T>T^*$ Fig.2) and low temperature correlated metallic (**PM**, $T<T^*$ Fig.2) states seems to correspond to a wide temperature interval shown by shaded area in the Fig.2. This statement can be proved by the gradual decrease of effective magnetic moment from ~2μ_B at T~250K to ~μ_B at T~100K observed in [11]. The change of effective moment seems to reflect the crossover from temperature induced localized magnetic moments (S=1) to delocalized charge carriers (S=1/2) thus supporting the consistency of spin fluctuation approach [5] at T>200K.

The temperature range 15K<T<T^* corresponds to the paramagnetic correlated metallic state of FeSi (**PM**, Fig.2), in which strong Hubbard correlations induce a substantial DOS renormalization with the formation of many-body resonance in the vicinity of Fermi level. Analysis of transport characteristics [7-8] shows that the many-body states within the resonance can be effectively described as short range (r_{sp}=5Å) spin polarons of low concentration n~10^{17}-10^{18} cm^{-3}. These spin polaronic states characterize by binding energy Δ_p=6 meV and strongly enhanced effective mass m^*~100m_0. In this model, the dramatic elevation of susceptibility observed at lowering temperature below T^* (Fig.1a) allows us to estimate the effects of the DOS renormalization giving the amplitude of the many-body resonance N~20 (eV·cell)$^{-1}$ [9].

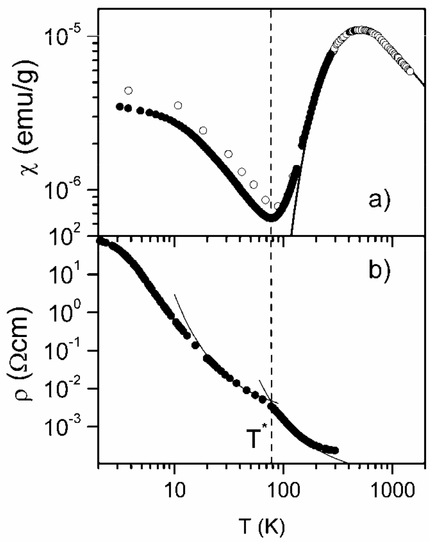

Fig.1. The temperature dependencies of a) magnetic susceptibility χ and b) resistivity ρ measured on FeSi single crystals. The open circles show the data taken from [3]. Solid lines show an activation type behavior (χ~T^{-1}exp(-E_a/k_BT), ρ~exp(-E_a/k_BT)) with parameter E_a≈ε_g at T>100K and E_a≈Δ_p at T<T^*.

Decreasing the temperature below T_C=15K in FeSi is accompanied by the appearance of anomalous Hall contribution $R_H^A(T)$<0. Taking into account the increase in the local magnetic properties observed near T_C [12], the anomalous transport characteristics of FeSi at 7K<T<15K should be attributed to the onset of the coherent regime of spin fluctuations. This transition is accompanied by the transformation of spin polarons into nanosize ferromagnetic regions (ferrons) (ferromagnetic phase **F**, Fig.2). This conclusion is proved by the hysteresis observed in the angular dependences of Hall resistance in magnetic fields H<3.6 kOe at T<T_C [8] (data 1 in Fig.2). Our analysis showed that this effect could be attributed to the magnetic anisotropy of ferrons with an anisotropy field of H_{an}=3±1 kOe (data 2, phase **F** in Fig.2). It should be emphasized that the onset of spontaneous magnetization in FeSi at $T<T_C$ is consistent to ferromagnetism criterion similar to the Stoner one $(UN>1)$, which has the form UN~5 for Hubbard parameter U=0.27 eV [8]. However, the low concentration of ferrons ($<10^{18}$ cm^{-3}) results only to weak singularities of integral magnetic characteristics (data 3 in Fig.2) since the volume magnetization and susceptibility of FeSi are mainly determined by the paramagnetic contribution of the FeSi matrix in this temperature range [8-9].

To estimate the charge carrier parameters in the ferromagnetic (**F**) phase of FeSi the temperature dependencies of effective concentration of charge carrier [12] have been used. Assuming for strong electron-phonon interaction, the relaxation time for charge carriers in FeSi $<\tau_{e\text{-}ph}>=1/(3\gamma_i^{ph})\approx1.7\cdot10^{13}$ s can be estimated from the linewidth of optical phonons in the frequency range 180-400 cm^{-1} γ_i^{ph}~10 cm^{-1} [13]. The same values of $<\tau_{e\text{-}ph}>$ are obtained from the reciprocal linewidth of magnetic scattering of polarized neutrons Γ=3÷4 meV [14]. Using the expression for the Hall mobility $\mu_H=R_H/\rho=e\tau/m^*$, the effective mass of charge carriers in FeSi have been also calculated. The estimation results to the value of $m^*/m_0(T>10K)=80\pm20$ which is consistent to the value $m^*/m_0=50$ obtained from the results of measurements of optical conductivity [15]. The correlation of the effective masses of charge carriers (spin polarons in **PM** phase and ferrons in **F** phase) can be treated as an additional argument in favor of the proposed description of the low-temperature transport and magnetic anomalies in FeSi.

When analyzing the parameters of the **F** phase at $T<T_C$=15K, the results of low temperature heat capacity study [16] should be taken into account. At particular, this study discovered a broad peak on the $C(T)$ curves around T~8.5K. This Schottky anomaly allowed to estimate the characteristic energy of splitting of the narrow band of many-body states Δ=2 meV [16]. Under the assumption that the onset of ferromagnetic state at temperatures $T<T_C$ is accompanied by the exchange-induced splitting of the many-body resonance having width $\Delta_p\approx6$ meV,
the exchange field can be estimated from the splitting energy: $H_{ex}=\Delta/\mu_B$=350 kOe. Such a good correlation between this result and the value of $H_{ex}=350\pm100$ kOe obtained from transport and magnetic parameters of FeSi [8-9] seems to strongly support the spin polaronic interpretation of the low-temperature properties of FeSi.

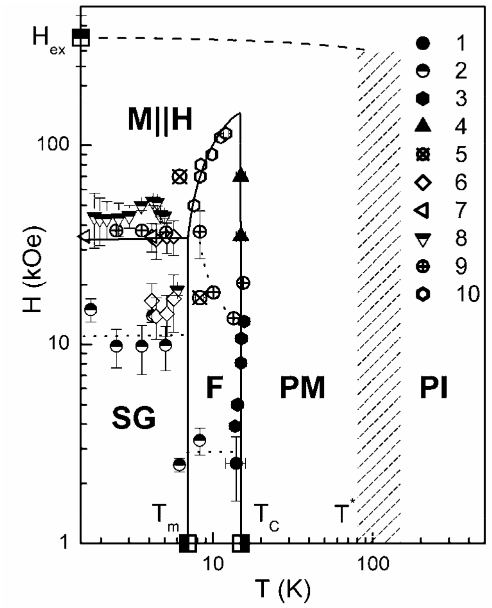

Fig.2. Low temperature magnetic phase diagram of FeSi. PI – paramagnetic semiconducting state, PM – paramagnetic strongly correlated metallic state, F and SG – ferromagnetic and mictomagnetic phases, respectively, M||H – phase of collinearly oriented ferrons (see text). The notations of presented data are given in the text.

Considering the region of $T<T_m$=7K, $H<H_m\approx35$ k0e on the H-T phase diagram of FeSi, we note that anomaly of the normal (spin-polaronic) component of the Hall effect $R_H^{SP}(T)$ is observed at T_m=7K (data 5 in Fig.2). Additionally, the magnetoresistance $\Delta\rho/\rho$ reveals a negative contribution at $T<T_m$ [12]. The above effects seem to reflect the change of ferrons' parameters. The analysis of galvanomagnetic data showed that the transition into magnetic state at $T<T_m$ is accompanied by a considerable decrease in Hall concentration and charge carriers effective mass to the values of $n=(R_H^{SP}e)^{-1}$~10^{16}-10^{17} cm^{-3} and $m^*/m_0=20\pm5$, respectively. This temperature range is also characterized by the substantial decrease in the effective magnetic moment μ_{eff} of ferromagnetic regions from (10-20)μ_B at $T<T_m$ to (4-6)μ_B at T=1.8K [12]. Allowing for the considerable increase in the anisotropy field to the values of H_{an}=12±2 k0e (data 2

in **SG** phase, Fig.2), the above features can be well understood within the formation of ferromagnetic microregions from interacting ferrons. So the anomalies of transport parameters observed at the transition to the mictomagnetic (**SG)** phase seems to be attributed to the strong scattering by inhomogenities and, as a consequence, to a sharp decrease in the mobility of charge carriers in the vicinity of T_m=7K.

When comparing FeSi with classical band ferromagnet, it should be emphasized that the behavior of physical parameters of FeSi is determined by the direct participation of nanosize magnetic regions in charge transport at low temperatures [8, 12]. This situation doesn't rule out a possible formation of new spatial magnetic structures as well as an occurrence of metamagnetic transitions. In our opinion, such a magnetic transition in the system of nanosize ferromagnetic microregions is responsible for the features of the physical parameters of FeSi observed in magnetic field H_m=35 k0e at $T<T_m$. The most pronounced anomalies (hysteresis) are observed in the magnetooptical absorption (data 6 Fig.2). Additionally, an anisotropic contribution to the magnetoresistance at $H\approx 35$ k0e [8] (data 7 in Fig.2) is appeared together with the inflection point on the $\Delta\rho/\rho(H)$ curves (data 8 in Fig.2). For $H\leq 35$ k0e the kink in the asymptotic behavior of the second Hall harmonic [12] is also observed (data 9 in Fig.2). In our opinion, these features reveal the change in the magnetic state of FeSi induced by the destruction of intracluster bonds in the mictomagnetic **SG** phase with the following reorientation of ferrons parallel to the applied field (**M||H** phase in Fig. 2). This suggestion is proved by the significant increase of the charge carriers' effective mass to the value of m^*/m_0=70±20 in magnetic field H=70.3 k0e [12] which is consistent to the values of the effective mass of spin polarons in **P** phase and ferrons in the **F** phase. Additionally, the peak observed on the temperature dependencies of the differential susceptibility during the measurement of magnetic characteristics (data 10 in Fig.2) can be also interpreted in terms of the new magnetic **M||H** phase on the low temperature phase diagram of FeSi. However, additional measurements of transport and magnetic characteristics of FeSi in the range of magnetic fields comparable to the exchange one $H\sim H_{ex}$ are necessary to determine the details of the low temperature phase diagram between the collinear magnetic (**M||H)** and paramagnetic (**PM**) phases in FeSi.

Acknowledgements

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