ISSN 1063-7761, Journal of Experimental and Theoretical Physics, 2007, Vol. 105, No. 1, pp. 1–3. © Pleiades Publishing, Inc., 2007.
Original Russian Text © G.S. Patrin, V.V. Beletskiĭ, N.V. Volkov, D.A. Velikanov, O.V. Zakieva, 2007, published in Zhurnal Éksperimental'noĭ i Teoreticheskoĭ Fiziki, 2007, Vol. 132, No. 1, pp. 7–10.

LOW-TEMPERATURE SOLID-STATE PHYSICS

Physical Properties of $Fe_{1-x}Dy_xSi$ Crystals

G. S. Patrin[a, b], V. V. Beletskiĭ[b], N. V. Volkov[a], D. A. Velikanov[a], and O. V. Zakieva[a]

[a] *Kirenskii Institute of Physics, Siberian Division, Russian Academy of Sciences, Akademgorodok, Krasnoyarsk, 660036 Russia*

[b] *Siberian Federal University, Krasnoyarsk, 660041 Russia*

e-mail: patrin@iph.krasn.ru

Abstract—The results of experimental investigation of magnetic and electric properties of $Fe_{1-x}Dy_xSi$ crystals are reported. It is shown that the magnitude and position of the anomaly observed in the temperature dependences of magnetization are controlled to a considerable extent by the external magnetic field. It is found that the introduction of Dy ions leads to a weak magnetoresistive effect.

PACS numbers: 75.20.Hr, 75.47.Pq

DOI: 10.1134/S1063776107070011

In view of their unique physical properties, FeSi crystals have been attracting the attention of researchers for many years. The behavior of magnetic susceptibility, as well as thermal and electrical parameters in this crystal is found to be unusual. For example, resistivity ρ first monotonically decreases upon heating by four orders of magnitude, attaining its lowest value near $T \approx 300$ K (the $\rho(T)$ dependence in this case cannot be described by a simple thermal-activation-controlled dependence), and then slowly increases with temperature. Due to this feature, the FeSi crystal is attributed to the class of Kondo compounds [1] or classified as a material exhibiting the semiconductor–metal transition [2]. From the magnetic standpoint, this material does not match the traditional behavior of known magnets. The magnetic susceptibility ($\chi \sim 10^{-4}$–10^{-6}) first decreases upon an increase in temperature to $T \approx 90$ K, and then substantially increases, attaining its peak value at $T \approx 500$ K [3]. The methods of Mössbauer spectroscopy [4] and neutron diffraction [5] revealed no magnetic order in this compound; at $T > 550$ K, the temperature dependence of the magnetic susceptibility obeys the Curie–Weiss law. However, more elaborate experiments with polarized neutrons [6] reveal magnetic scattering in such crystals, which is interpreted as the existence of microscopic regions with ferromagnetic correlations of spin fluctuations.

A complex of detailed studies of magnetic, thermoelectric, and galvanomagnetic properties [7–9] of the FeSi crystal led to the conclusion that the results can be interpreted using the Hubbard model with strong electron correlations. The authors of these works attribute the low-temperature peculiarities in the physical properties to the formation of spin polarons. It should be noted that some authors attribute the low-temperature peculiarities to the presence of iron impurity ions due to nonstoichiometric composition of the crystal [3, 8, 10], while the high-temperature dependences are explained by the transition of electrons in the d^4 configuration from the e_{2g}^4 state ($S = 0$) to the $e_{2g}^3 t_{2g}^1$ state ($S = 1$) [3]. However, no generally accepted pattern has been adopted for describing the entire set of physical properties of FeSi.

In some publications, an attempt has been made at modifying the properties of the FeSi crystal by doping it with various impurities. For example, the substitution of 4*d* iridium ions for Fe ions [11] increases the height of the magnetic susceptibility peak and completely changes the resistivity, leading to the metal-type conduction for an Ir concentration of $x > 10$ at %. The addition of 3*d* cobalt ions [12] leads to the formation of magnetic order and hole conductivity mechanism, preserving the temperature variation of ρ on the whole. The substitution of aluminum for silicon [13] results in metallization of the state of the sample and in an overall increase in the magnetic susceptibility.

In the series of magnetic elements, 4*f* ions are characterized by the fact that many of these ions readily form chemical compounds with silicon [14] and have localized magnetic moments. At low temperatures, individual properties of rare-earth ions are manifested the most strongly, which makes it possible to use them as probes in the task-oriented variation of physical properties of crystals.

In this communication, we report on the results of experiments on the effect of 4*f* dysprosium impurity ions on the electric and magnetic properties of the iron monosilicide crystal.

Polycrystalline $Fe_{0.99}Dy_{0.01}Si$ samples were obtained by the method described in [10]. X-ray measurements were performed on the DRON-4 diffractometer on polycrystalline samples taken from different parts of the crucible. All diffraction patterns were iden-

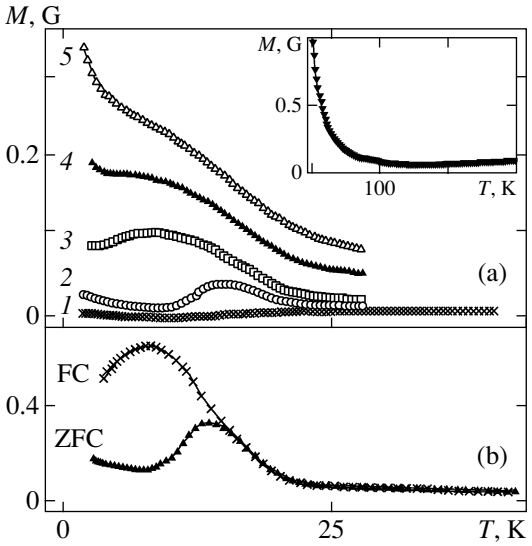

Fig. 1. Temperature dependences of magnetization of $Fe_{1-x}Dy_xSi$: (a) in the ZFC regime in different magnetic fields $H = 10$ (*1*), 100 (*2*), 500 (*3*), 1500 (*4*), and 2500 (*5*) Oe; (b) in the FC and ZFC regimes for $H = 400$ Oe. The inset shows the dependence for $H = 10$ kOe.

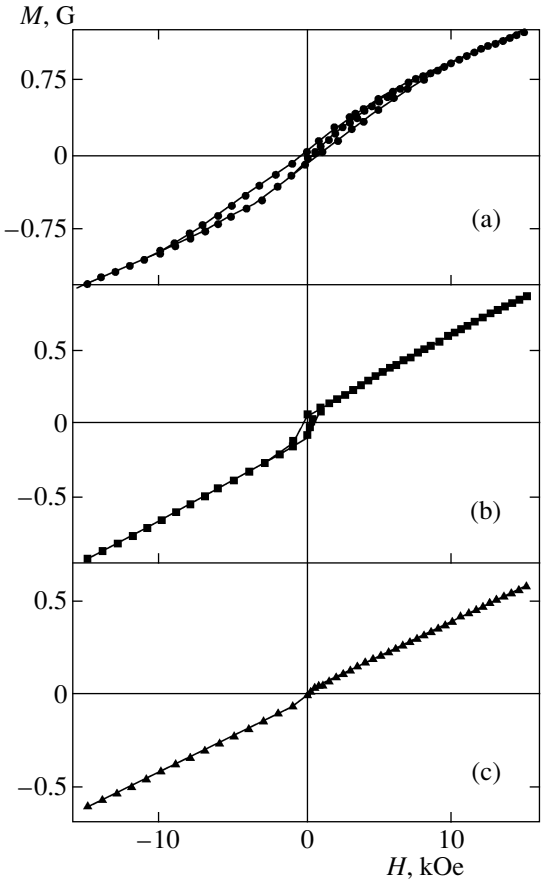

Fig. 2. Field dependences of magnetization at $T = 2$ (a), 10 (b), and 20 (c) K.

tical. The magnetization was measured on a SQUID magnetometer in weak magnetic fields and on a PPMS device in strong fields. Before each measurement, the sample was placed in a demagnetizer. The temperature and field dependences of resistivity were obtained by the four-probe technique.

A comparison of diffraction patterns for the doped and nominally pure crystals revealed that doping of the FeSi crystal with Dy ions with concentrations ≤1 at % does not change the structure. No traces of compounds in the Dy–Fe and Dy–Si systems were revealed either.

It is well known [10] that a nominally pure crystal exhibits no magnetic susceptibility anomalies in the low-temperature region. The implantation of Dy ions strongly affects the magnetic properties at low temperatures in weak magnetic fields (Fig. 1). Measurements of magnetization M in the zero-field cooling (ZFC) mode revealed an anomaly in the form of a peak on the temperature dependence $M(T)$. Upon an increase in the magnetic field, the position (blocking temperature T_c) of this peak is shifted towards low temperatures, its height increases, and the peak becomes much broader (Fig. 1a). In the field cooling (FC) mode in a magnetic field equal to the measuring field, the anomaly is preserved, but the peak is displaced towards lower temperatures, its height being much larger than in the ZFC regime (Fig. 1b). In this case, the $M(T)$ curve has the same shape for heating and for cooling. In magnetic fields $H \geq 5$ kOe, the $M(T)$ dependences for a nominally pure crystal and for a Dy-doped crystal behave analogously (see inset to Fig. 1). Figure 2 shows the field dependences of magnetization at various temperatures. Hysteresis phenomena are observed; however, it can be seen from the figure that such a behavior takes place at temperatures $T < 25$ K, while no anomaly is observed at higher temperatures.

Measurements of resistivity of a Dy-doped crystal reveal that the shape of the temperature dependence $\rho(T)$ is qualitatively the same as for a nominally pure crystal. The only difference is that the resistivity ρ_{Dy} of the doped crystal is lower than that (ρ_{NP}) for the nominally pure crystal under identical experimental conditions. Figure 3 shows the temperature dependences of the difference $\Delta\rho = \rho_{Dy} - \rho_{NP}$ obtained in different magnetic fields. It can be seen that the magnetic field dependence of $\Delta\rho$ is observed only at temperatures $T < 40$ K, while a constant but magnetic-field-independent variation of resistivity is observed at higher temperatures. At a fixed temperature, the resistivity of the $Fe_{1-x}Dy_xSi$ crystal increases with the magnetic field, and the experimental curve can be approximated by a quadratic dependence (see inset to Fig. 3) of the form

$$\rho_j(H) = A_j + B_jH + C_jH^2,$$

where j = Dy or nominally pure crystal (NP). The table contains the values of the coefficients in the fitting dependence for $\rho(H)$.

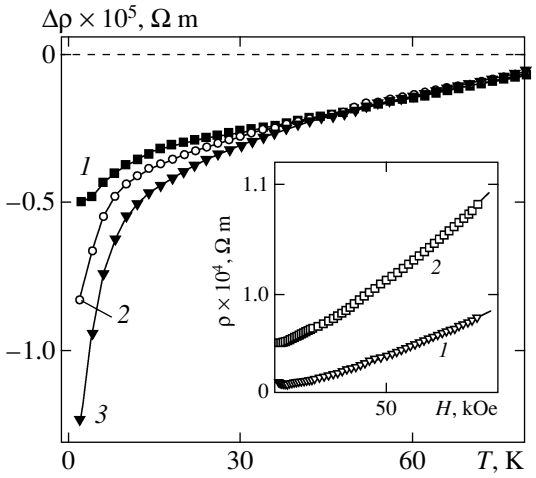

Fig. 3. Temperature dependences of resistivity variation in various magnetic fields $H = 10$ (*1*), 50 (*2*), and 90 (*3*) kOe. The inset shows the field dependence of resistivity at $T = 2$ K for a $Fe_{1-x}Dy_xSi$ crystal (*1*) and FeSi crystal (*2*).

Thus, our experiments show that doping of the FeSi crystal with Dy ions noticeably changes its magnetic and transport properties. In the nominally pure crystal, a magnetic hysteresis exists in fields $H < 100$ Oe, but it has a different shape and is due to the interaction between iron clusters [10]. In this case, we can assume that impurity clusters containing Dy ions are additionally formed along with "purely iron" magnetically isotropic clusters on account of departures from stoichiometry. It is well known [15] that iron and dysprosium in Fe–Dy alloys participate in the antiferromagnetic exchange. If dysprosium appears in the tetravalent state, it is in the $^5I_8$ state and its energy spectrum consists of a series of strongly anisotropic quasi-doublets separated by long distances. Splitting of these quasi-doublets is caused by the exchange field produced by iron environment. The magnetization peak can be associated with the population of low-lying levels of dysprosium ions, while the field and temperature hysteretic loops are due to magnetization reversal of precisely these clusters. Such a scenario explains the presence of the energy barrier separating various metastable states of a cluster. The height of this barrier determines the blocking temperature T_c, while the behavior of the entire subsystem resembles the spin-glass behavior [16].

As regards the electric properties, the situation is not so clear. On the one hand, the introduction of dysprosium ions must change the energy spectrum and the number of charge carriers; on the other hand, it must lead to the emergence of one more magnetic scattering channel. However, we observe an overall decrease in the resistivity of the crystal upon doping and its further decrease as a result of application of a magnetic field.

Table

Sample	$A \times 10^5$	$B \times 10^{11}$	$C \times 10^{16}$
FeSi	9.5	8.8	6.4
$Fe_{1-x}Dy_xSi$	9.2	8.3	4.0

ACKNOWLEDGMENTS

This study was supported by the program "Russian Foundation for Basic Research—Enisey" (project no. 05-02-97708-a).

REFERENCES

1. G. Aeppli and Z. Fisk, Comments Condens. Matter Phys. **16**, 155 (1992).
2. M. Imada, A. Fujimori, and Y. Tokura, Rev. Mod. Phys. **70**, 1039 (1998).
3. V. Jaccarino, G. K. Wertheim, J. H. Wernick, et al., Phys. Rev. **160**, 476 (1967).
4. G. K. Wertheim, V. Jaccarino, J. H. Wernick, et al., Phys. Lett. **18**, 89 (1965).
5. M. Kohgi and Y. Ishikawa, Solid State Commun. **1**, 37 833 (1998).
6. K. Tajima, Y. Endoh, J. E. Fischer, and G. Shirane, Phys. Rev. B **38**, 6954 (1998).
7. N. E. Sluchanko, V. V. Glushkov, S. V. Demishev, et al., Europhys. Lett. **51**, 557 (2000).
8. N. E. Sluchanko, V. V. Glushkov, S. V. Demishev, et al., Zh. Éksp. Teor. Fiz. **119**, 359 (2001) [JETP **92**, 312 (2001)].
9. N. E. Sluchanko, V. V. Glushkov, S. V. Demishev, et al., Phys. Rev. B **65**, 064404 (2002).
10. G. S. Patrin, V. V. Beletskiĭ, D. A. Velikanov, et al., Fiz. Tverd. Tela (St. Petersburg) **48**, 638 (2006) [Phys. Solid State **48**, 700 (2006)].
11. B. C. Sales, E. C. Jones, B. C. Chakoumakos, et al., Phys. Rev. B **50**, 8207 (1994).
12. S. Asanare, D. Shimoda, and Y. Sasaki, Phys. Rev. [Sect. A] **134**, A774 (1964).
13. J. F. Di Tusa, K. Frielmelt, E. Ducher, et al., Phys. Rev. Lett. **78**, 2831 (1997).
14. V. M. Denisov, S. A. Istomin, O. V. Podkopaev, L. I. Serebryakova, L. T. Antonova, É. A. Pastukhov, and V. V. Beletskiĭ, *Silicon and Its Alloys* (Ural. Otd. Ross. Akad. Nauk, Yekaterinburg, 2005) [in Russian].
15. M. I. Darby and K. N. R. Taylor, *Physics of Rare Earth Solids* (Chapman and Hall, London, 1972; Mir, Moscow, 1974).
16. C. Y. Huang, J. Magn. Magn. Mater. **51**, 1 (1985).

Translated by N. Wadhwa