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Magnetic Properties of $Fe_{1-x}Co_xSi$ Single Crystals at Low Co Impurity Concentrations

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Magnetostatic properties of FeSi and $Fe_{0,98}Co_{0,02}Si$ single crystals have been studied. It has been found that the temperature and field dependences of the magnetization of monocrystal FeSi are strongly affected by introduction of a small amount of Co (2 %). A description of the results were provided by a model accounting for the formation of superparamagnetic iron clusters, as well as Fe-Co complexes. It is assumed that Fe-Co complexes form a ferromagnetic phase, which is approximately 0.6% of the $Fe_{0,98}Co_{0,02}Si$ sample weight

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$Fe_{1-x}Co_xSi$ compounds has attracted extensive interest because of unusual magnetic structures in them. The pure FeSi is nonmagnetic [1], but the introduction of Co impurities leads to appearance of magnetic ordering in the form of a helimagnetic or conical phase, depending on external factors [2]. In heavily doped samples a skyrmion lattice forms [3], which can be observed using various methods [4, 5]. It is also assumed that there are "chiral bubbles", new stable states on the surface of $Fe_{1-x}Co_xSi$ [6].

The magnetic and magnetotransport properties of the $Fe_{1-x}Co_xSi$ compounds depend on the concentration of Co. The change in concentration causes an increase in the critical temperature and a change in chirality to the opposite [7]. Bulk and film $Fe_{1-x}Co_xSi$ samples have a positive magnetoresistance, the value of which also depends on x [8, 9]. Due to the combination of all these factors, FeSi is considered as a promising material for spintronics and magnetic storage [6, 9, 10].

There are articles that reflect the properties of heavily doped $Fe_{1-x}Co_xSi$ systems. But there is interest in research in the range of low impurity concentration. Even at $x = 0.02$ a dielectric-metal transition is observed [11]. This is about the same critical concentration as was found in the $Fe_{1-x}Cr_xSi$ compound, where the deviation of the thermopower and the Sommerfeld coefficient is observed [12]. Weak doping with heavy impurities such as Ir and Os affects the scattering of phonons, decreasing the phonon thermal conductivity [13]. The $Fe_{1-x}Co_xSi$ compound with a concentration of $x \leqslant 0.3$ has the same characteristic[14].

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1. Experiment

The $Fe_{x-1}Co_xSi$ compounds were obtained by gas-transport reaction method. The size of crystallites in such samples are of the order of $\leqslant 10^{-2}$ mm with an average transverse size of the single crystal of about 1.5 mm [15]. A monocrystal FeSi (containing no cobalt) and a $Fe_{0,98}Co_{0,02}Si$ (with a content of 2% Co) samples were prepared for the present study.

The structure and composition of the samples were studied using semiquantitative X-ray phase analysis. X-ray patterns of FeSi and $Fe_{0,98}Co_{0,02}Si$ samples are identical (Fig. 1, top). Also, these patterns agree with the reference FeSi spectrum (Fig. 1, bottom). Peaks that correspond to metallic Co are absent. This indicates that the cobalt atoms are dissolved in FeSi. In addition to the main phase, only the Co_7Fe_3 phase can be present in the samples, the lines of which can be overlapped by FeSi lines. The composition of the samples is confirmed and corresponds to the nominal value.

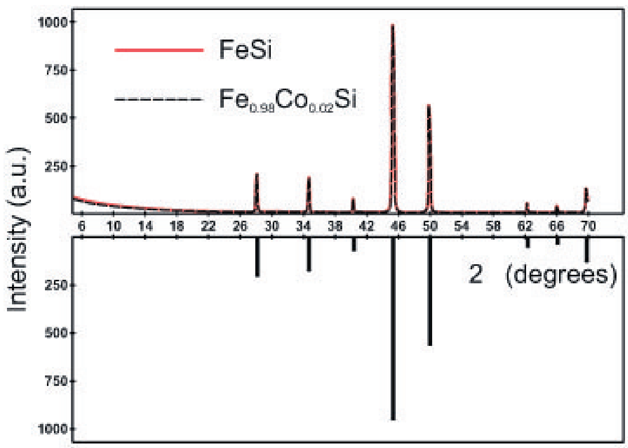

Fig. 1. Top — X-ray diffraction patterns of FeSi (red) end $Fe_{0,98}Co_{0,02}Si$ (black dotted) samples. Bottom — the reference FeSi spectrum (black strokes)

2. Magnetic measurements

Studies of magnetostatic characteristics were performed on a MPMS-XL SQUID magnetometer. Samples do not have pronounced ferromagnetic properties. Magnetization curves have a paramagnetic form at a temperature of 2 K (Fig. 2a). A sample of pure FeSi reaches saturation at about 1.5 emu/g, while in $Fe_{0,98}Co_{0,02}Si$ this one is about 2.5 emu/g. The magnetization curve of the FeSi sample becomes almost linear at a temperature of 300 K (Fig. 2b). In addition, the difference between magnetic moments of FeSi and $Fe_{0,98}Co_{0,02}Si$ samples becomes larger (0.45 and 1.6 emu/g, respectively).

The temperature dependence of the magnetic moment was measured in a field of 10 kOe for the $Fe_{0,98}Co_{0,02}Si$ sample and in a field of 5 kOe for the FeSi sample. It is known that iron atoms in FeSi form superparamagnetic clusters, and their behavior can be described by the Langevin function [16]. If impurity Co atoms are surrounded by iron atoms (Fe-Co complexes), they become magnetic and create an additional magnetic moment. Therefore, the theoretical fit is accomplished using the following expression:

$$M(H,T) = mN\{\coth(mH/k_BT) - k_BT/mH\} - M_{Fe-Co}, \quad (1)$$

where the first term describes the moment associated with superparamagnetic Fe clusters, and M_{Fe-Co} is the magnetic moment from the Fe-Co complexes. The temperature dependence of

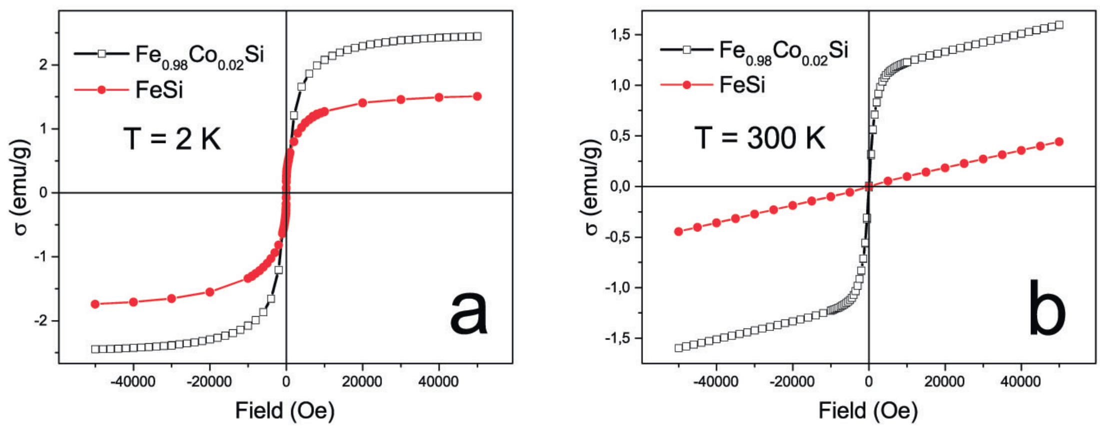

Fig. 2. The magnetization curves of FeSi and $Fe_{0,98}Co_{0,02}Si$ samples: a) at T = 2 K, b) at T = = 300 K

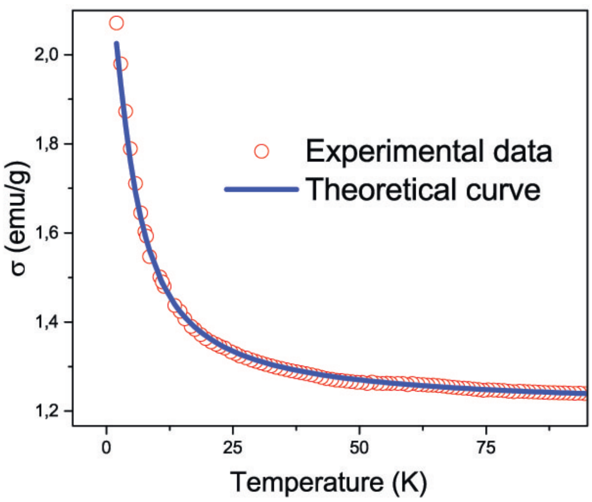

Fig. 3. The temperature dependence of the magnetic moment of the $Fe_{0,98}Co_{0,02}Si$ sample. The theoretical curve is plotted using the formula (1)

the magnetic moment of the $Fe_{0,98}Co_{0,02}Si$ sample is well described by this formula when using the parameters $m = 1,3 \cdot 10^{-19}$ emu, $N = 6 \cdot 10^{16}$, and $M_{Fe-Co} = 1,2$ emu/g. The theoretical curve calculated in this way coincides with the experimental dependence (Fig. 3). Such a fitting for FeSi single crystal does not give any satisfactory result.

The dependence of the susceptibility $\chi_{Fe-Co}(H)$ of Fe-Co complexes was found using the field dependences of the magnetic moment of the $Fe_{0,98}Co_{0,02}Si$ sample, as well as the parameters m and N from the fit. To do this, the magnetic moment, calculated through the Langevin function

$M_L = \coth(mH/k_BT) - k_BT/mH$, was deducted from the experimental values of the magnetic moment M_{exp}. Obtained field dependences of the susceptibility of the sample $Fe_{0,98}Co_{0,02}Si$ at different temperatures almost coincide (Fig. 4a). This suggests that the magnetism of Fe-Co complexes is the same at any temperature. Thus, one can assume the presence of a ferromagnetic phase in the composition of the $Fe_{0,98}Co_{0,02}Si$ sample.

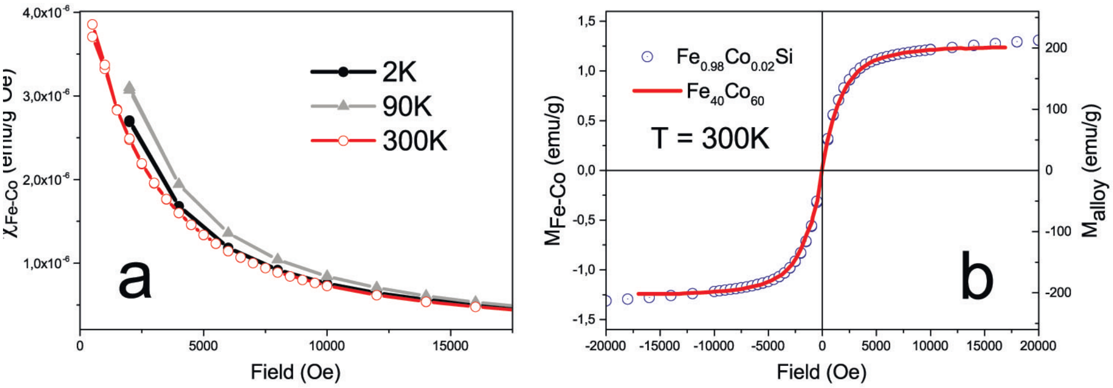

Fig. 4. a — field dependence of the susceptibility of Fe-Co complexes at different temperatures. b — the magnetization curves of Fe-Co complexes (left scale) and $Fe_{40}Co_{60}$ alloy [17] (right scale)

Various magnetization curves were compared with the field dependence of the magnetization associated only with Fe-Co complexes ($M_{Fe-Co}(H) = M_{exp} - M_L$). It was ascertained that $M_{Fe-Co}(H)$ is most similar to the magnetization curve of the $Fe_{40}Co_{60}$ alloy [17]. The Curie temperature of such an alloy is much higher than room temperature. From the ratio of division values of the left and right scales (Fig. 4b), one can estimate that the Fe-Co complexes constitute approximately 0.6 % of the entire $Fe_{0,98}Co_{0,02}Si$ sample weight.

The typical temperature dependence of the magnetic susceptibility for FeSi has a minimum of about 100 K, after which it increases with increasing temperature (in the range under study up to 300 K). This is exactly what was obtained (curve 1 in Fig. 5). However, the susceptibility

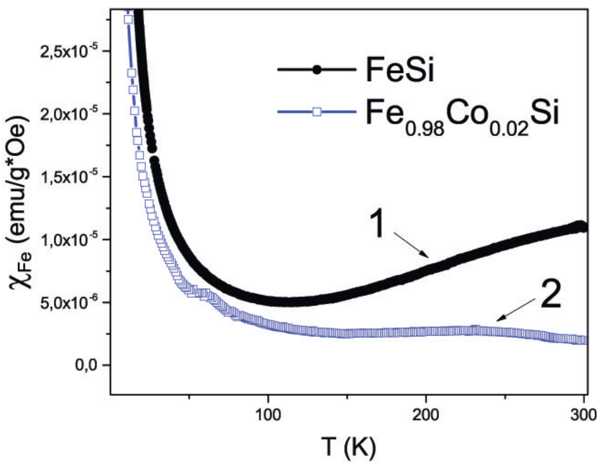

Fig. 5. The temperature dependences of the magnetic susceptibility, associated only with Fe clusters, of FeSi (curve 1) and $Fe_{0,98}Co_{0,02}Si$ (curve 2) single crystals

of the sample $Fe_{0,98}Co_{0,02}Si$ decreases monotonically with increasing temperature. The assumed contribution from Fe-Co complexes was subtracted from the experimental data and the susceptibility $\chi_{Fe} = (M(H,T) - M_{Fe-Co})/H$, associated only with Fe clusters, was calculated (curve 2 in Fig. 5). However, the temperature dependence of the susceptibility of the FeSi sample, which does not have Fe-Co complexes, is significantly different from that of the $Fe_{0,98}Co_{0,02}Si$ sample, where the contribution of Fe-Co complexes is excluded.

Conclusion

The low concentration of cobalt in the $Fe_{x-1}CoxSi$ single crystal causes significant changes in the magnetic properties. Lightly doped $Fe_{0,98}Co_{0,02}Si$ sample demonstrates the temperature dependence of the magnetic susceptibility different from that of pure FeSi. This is attributed to the presence in the crystal of an additional ferromagnetic Fe-Co phase. These results make it promising to study the structure of such samples, as well as samples with different concentrations of Co.

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Магнитные свойства монокристаллов $Fe_{1-x}Co_xSi$ при малой концентрации примеси Co

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В работе представлено исследование магнитостатических характеристик образцов FeSi и $Fe_{0,98}Co_{0,02}Si$. Обнаружено, что внесение небольшого количества примести Co (2 %) значительно влияет на температурные и полевые зависимости намагниченности монокристалла FeSi. Результаты обработаны в рамках модели, учетывающей образование суперпарамагнитных кластеров железа, а также комплексов Fe-Co. Предполагается, что комплексы Fe-Co образуют ферромагнитную фазу, которая составляет примерно 0,6 % от массы образца $Fe_{0,98}Co_{0,02}Si$.

Ключевые слова: силицид железа, примесь Co, суперпарамагнетизм.