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Magnetotransport and magnetic properties of $\beta$-FeSi$_2$ single crystals

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Abstract. Magnetotransport and magnetic properties have been measured for $\beta$-FeSi$_2$ single crystals from 4.2 to 300 K using a pulsed magnet, a superconducting magnet, and a SQUID magnetometer. Nonlinear Hall effect followed by a normal Hall effect was observed, involving a hysteresis effect at low temperature. The signs of normal Hall coefficients changed from positive to negative ones at around 180 K. The magnetization curves above 180 K are well explained by considering the superparamagnetism. Below 150 K we found that the aggregates of Fe atoms exhibit an inter-aggregate ferromagnetic interaction. At low temperature we also found that the cohesive forces obtained from the $\rho_H$–$B$ curves are larger than those from the $M$–$B$ curves. This discrepancy was discussed qualitatively by a proposed model.

1. Introduction

$\beta$-FeSi$_2$ is one of ecologically friendly semiconductors made from nontoxic and abundant elements. This material has attracted much attention as a promising one for applications in the field of optoelectronics and thermoelectrics, since it has many superior features including a sufficiently high figure-of-merit and absorption coefficient [1]. Therefore, the transport, thermoelectric and optical properties have been extensively studied. However, the studies on the magnetotransport and magnetic properties have been not performed enough yet. In the Hall resistivity on the magnetic field $\rho_H$ of $\beta$-FeSi$_2$, the anomalous contributions to $\rho_H$, that is, nonlinear dependence and hysteresis effect have been reported [2, 3]. These anomalies have been explained by anomalous Hall effect [2] or by the
existence of two types of carriers [3], though their origin remains still unknown. So far, the articles to support the latter are few. On the other hand, there is a report that the anomalies arise from the superparamagnetic clusters [2].

In this study, we investigated the magnetotransport properties of $\beta$-FeSi$_2$ single crystals up to 30 T in the temperature range of 4.2 – 300 K using a combination of pulsed and superconducting magnets. We also investigated their magnetic properties up to 6 T using a SQUID magnetometer. In high field $\rho_H$ measurements, it is found that the sign of normal Hall coefficient changes from positive to negative ones at around 180 K. We will demonstrate that the anomalous Hall effect is due to the superparamagnetism through the analysis of the Hall resistivity and the magnetization under low magnetic field. It will be also demonstrated that the present material exhibits an inter-aggregate ferromagnetic interaction at low temperature.

2. Experiment

The single crystals of $\beta$-FeSi$_2$ were grown by a chemical vapor transport technique using iodine as a transport agent. The grown crystals have a needle-like shape elongated along the [010] axis.

Magnetotransport experiments were performed by a six-probe method in the temperature range of 4.2 – 300 K. High field and low field Hall effects were measured for several single crystals up to 30 T using a 40 T class pulsed magnet and a superconducting magnet. Magnetization experiments were also performed with single crystals up to 6 T using a SQUID magnetometer.

3. Results and discussions

Figure 1 shows the magnetic field dependences of Hall resistivity $\rho_H$ up to (a) 1 T and (b) 30 T for various temperatures. All experimental data obtained under the pulsed magnet were plotted only for the down-sweep process to avoid a reduction of signal to noise ratio caused by some induction noise. As shown in Fig. 1, at 4.2 K, $\rho_H$ increases rapidly with increasing magnetic field $B$, tends to saturate around 5 T, and then increases gradually with a positive slope. Such a nonlinear behaviour is also observed at the whole temperature range, though the slope of $\rho_H$ in the higher field region changes from negative to positive ones at around 180 K. As seen from $\rho_H$–$B$ curve under the low magnetic field in Fig. 1(a), on the other hand, $\rho_H$ exhibits a hysteresis with the cohesive force of 600 mT at 4.2 K, in addition to the nonlinear dependence. But this hysteresis disappeared above 180 K. The above results are in qualitatively agreement with the previous data reported [2]. In such a case, the nonlinear Hall

![Figure 1](image_url)
effect may be interpreted by the anomalous Hall effect. That is, as for the ferromagnetic materials, the Hall resistivity would be expressed by the form $\rho_H = R_H B + R_A M$, where $R_H$ and $R_A$ denote the normal Hall and anomalous Hall coefficients, respectively, and $M$ the magnetization.

Figure 2 shows the magnetic field dependences of $M$ for various temperatures. As shown in Fig. 2(a), the $M$–$B$ curve at 4.2 K exhibits a nonlinear field dependence with a narrow hysteresis. It should be noted that the cohesive force is rather smaller (200 mT) than that in the $\rho_H$–$B$ curve. This fact suggests that the nonlinear Hall dependence cannot be explained by the simple anomalous Hall effect above-mentioned. As can be seen from Fig. 2(a), the hysteresis of the $M$–$B$ curve disappeared at 180 K, similar to the case of $\rho_H$–$B$ curve.

In connection with the nonlinear dependence of $M$–$B$ curves, we measured the temperature dependences of zero-field cooled and field-cooled (ZFC and FC) magnetizations. The results revealed that the present samples behave as a superparamagnetism (SP) whose blocking temperature is 30 K at 30 mT, though the stoichiometric $\beta$-FeSi$_2$ shows a paramagnetism. From this, therefore, it is suggested that the nonlinear dependence of $M$–$B$ curves is related to the SP. Probably, the present system must contain a small amount of excess Fe atoms and consist of some aggregates (AG) of ferromagnetic Fe atoms, with the result that they are responsible for the SP.

Figure 2(b) illustrates the $M$–$B$ curves up to 2 T for various temperatures. The magnetization curves almost saturate above 1 T at all the temperatures. In case of the SP, the magnetization can be expressed by the form $M = M_{\text{sat}} L(\mu B/k_BT)$, in which $M_{\text{sat}}$ denotes the saturated value of $M$, $L$ the Langevin function as a function of $B/T$, $\mu$ the magnetic moment, and $k_B$ the Boltzmann constant [4].

The experimental data in the higher temperature range above 180 K where the hysteresis was not observed can be well fitted by this formula, while those below 180 K could not be fitted. Therefore, we extended $L$ as $M = M_{\text{sat}} L(\lambda B(T + T_{\text{c}}^{\text{IAG}}))$, in which $T_{\text{c}}^{\text{IAG}}$ denotes the temperature characterising an inter-aggregate (IAG) ferromagnetic interaction. The plausibility for such an extension in the Langevin function has been already demonstrated in Fe cluster systems embedded in Ag films [4]. The result of Langevin fittings for the present system is shown in Fig. 3. From the fitting analysis, the temperature-independent value of $\mu$ was estimated to be $-5,600 \mu_B$ ($\mu_B$: the Bohr magneton). The value is 2,000 times as larger as the magnetic moment of the isolated Fe atom. Below 180 K, $T_{\text{c}}^{\text{IAG}}$ increases up to 80 K with decreasing temperature.

The above results give us the following important insights. (1) The present system can be understood entirely by the SP feature. (2) Since all the magnetization data can be explained

![Figure 2](image-url)

**Figure 2.** Magnetic field dependences of (a) the magnetization $M$ below $|B| \leq 0.4$ T at 4.2 and 180 K and of (b) the magnetization $M_{\text{up}}$ measured in the up-sweep processes below $|B| \leq 2$ T.
quantitatively by the modified Langevin function with single temperature-independent value of $\mu$, the size distribution of the ferromagnetic AG’s is nearly mono-dispersion. (3) Below 180 K, where the hysteresis effect appears, the IAG interaction increases with decreasing temperature.

Finally, there remains a problem on the discrepancy between the cohesive forces obtained from the $\rho_{H-B}$ and $M-B$ curves. In order to explain this discrepancy, we propose a SP model in which AG’s with short IAG distance show strong IAG interaction but others with long IAG distance show weak interaction: the former gives rise to $M_1$ with a relatively large $R_{A1}$ and cohesive force, and the latter gives rise to $M_2$ with a relatively small $R_{A2}$ and cohesive force. As a result the above-mentioned formula for the anomalous Hall effect is modifies as follows: $\rho_{H} = R_{H}B + R_{A1}M_1 + R_{A2}M_2$.

According to our proposal, the discrepancy between the cohesive forces can be interpreted qualitatively.

4. Summary
Magnetotransport properties have been measured for $\beta$-FeSi$_2$ single crystals from 4.2 to 300 K up to 30 T using the pulsed magnet. The magnetic field dependences of the Hall resistivity and the magnetization have been also measured under weak field conditions using the superconducting magnet and the SQUID magnetometer. Nonlinear Hall effect followed by linear Hall component was observed, involving the hysteresis effect at low temperature. It was found that the sign of the ordinal Hall coefficients changes from positive to negative ones at around 180 K. The magnetization curves above 180 K were well interpreted by the formula for the SP. Below 180 K, where the hysteresis effect exists, we found that the aggregated Fe ions exhibit the inter-aggregate ferromagnetic interaction. This means that the present system consists of some aggregates of ferromagnetic Fe atoms. At low temperature we found that the cohesive forces in the $\rho_{H-B}$ curves are larger than those in the $M-B$ curves. This discrepancy was discussed qualitatively on the basis of the proposed SP model.

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