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Analysis of the approximation to saturation magnetization and dynamics of demagnetization of PrDyCoFeB amorphous spin glass

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An analysis of the field and time dependencies of the magnetization of PrDyFeCoB amorphous crystalline microwires is presented. It is found that the magnetization curve near the saturation field has a smoother approximation to saturation than in a ferromagnet, according to theoretical predictions for the spin-glass state of the alloy in the amorphous state within the framework of the random magnetic anisotropy model. Deviations of the dynamics of relaxation of the magnetization of microwires from the logarithmic time dependence and the disappearance of this difference when observing magnetic relaxation in a magnetic field are found. This indicates the typical dynamics of the magnetization reversal of a spin glass in a zero field and the ferromagnetic character of demagnetization in a nonzero field. The results indicate the presence in the PrDyFeCoB microwires of an exotic magnetic state of the material with stochastic local magnetic anisotropy.

Keywords: spin glass, ferrimagnet, rare earth alloys, random magnetic anisotropy.

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1. Introduction

Spin glasses with high local magnetic anisotropy have long attracted attention as new media with unusual magnetic properties. Amorphous alloys of DyFeB intermetallides, which have been intensively studied some time ago, can be considered as an example of such materials [1–5]. A special feature of this group of alloys is the significant energy of single-ion magnetic anisotropy D, which greatly exceeds the energy of exchange interaction of ions J. The fulfillment of the condition $D \gg J$ results in such a situation in an amorphous material, when magnetic anisotropy variations and anisotropy axes chaotization in individual ions lead to magnetization disorder, rather than to variation of interatomic distances r, modulating the value of exchange interaction $J(r)$ [1]. The significant asymmetry of single-ion anisotropy in rare earth alloys results in the appearance in them of specific states of spin glass, such as the Ising spin glass, as well as in the appearance of spin-reorientation transitions caused by a relatively weak magnetic field. Note that the physical nature of the spin glass with rare earth ions differs from the spin-glass states in amorphous alloys based on transition metals (FeSi, FeCoSi, etc.), in which atomic disorder creates a chaotic distribution of the exchange interaction value $J(r)$, rather than magnetic anisotropy.

In the studies [6,7], a spin-reorientation transition in PrDy-FeCo-B microwires was discovered, which manifests itself as a fast growth of microwire magnetic moment by 3–10 times when 100–1000 Oe magnetic field is applied along the microwire at 250 K. At other temperatures, a different critical field was required, so that the dependence of the critical temperature of this transition on the critical field of switching states obeyed the well-known Almeida–Thouless law, obtained theoretically for spin glasses with high magnetic anisotropy [8]. PrDy-FeCo-B alloy in crystalline phases 2-14-1, 1-4-1 and etc. is isostructural to the previously studied alloy DyFeB, in which the theoretically predicted Almeida–Thouless transition was discovered. The only difference is that in transition metal subarray a part of the iron ions is substituted by Co^{2+} ions with greater anisotropy, and in the rare earth subarray a part of Dy^{3+} ions is substituted by close to them Pr^{3+} ions with slightly lower values of single-ion anisotropy. In all other respects, as well as in the studies [1–5], in amorphous microwires a spin-reorientation transition between the spin-glass (SSG) and ferrimagnetic (FM) states is manifested. The physical processes underlying such a transition were considered using the cluster theory of percolations (or percolation theory) [9], which is applied to amorphous material islands arising randomly as a result of correlation of single-ion anisotropy axes in local material regions [10]. The growth of magnetic field results in the growth of such islands and the gradual distribution of ferrimagnetic regions displacing the spin-glass state. With decreasing temperature, overcoming the magnetic anisotropy barrier becomes more

difficult, and an ever larger field is required to switch to the ferrimagnetic state. The feasibility of the Almeida–Thouless predictions for the boundary of such a transition in the field-temperature space $(H-T)$ was checked by us in the previous studies, that confirmed the transition SSG $\rightarrow$ FM [6,7].

Another sign of the spin-glass state, even more general, suitable for spin glasses of various types, is a smoother approximation to saturation magnetization M_S with increasing magnetic field [11], as well as specific time dependences of magnetization with an external field change, different from the logarithmic dependence [7] observed in structurally imperfect ferromagnets. The approximation of magnetization to saturation in amorphous ferromagnets was studied in Kronmüller's studies [10,12,13], and also analyzed in the studies by Chudnovsky and his colleagues [14–17], who proposed an explanation of this phenomenon based on the idea of the chaotic exchange interaction distribution. In the study [18] similar theoretical explanations of the magnetization curves for the situation when magnetic anisotropy distribution by size and sign is chaotic were obtained.

The purpose of this study was to check the shapes of magnetization field dependences when approaching to saturation (in strong fields) in amorphous microwires, as well as to establish time dependences of spontaneous demagnetization of amorphous PrDyFeCoB microwires and their comparison with magnetic relaxation of ferromagnet state induced by the field in the same PrDyFeCoB microwire.

2. Procedure and samples

Amorphous microwires with a volume fraction of amorphous material up to 86% were obtained by ultrafast cooling of a PrDyFeCoB liquid-alloy drop on a rotating copper cylinder cooled with water. The cooling rate of the liquid-alloy under similar conditions is usually 10^6 K/s [7]. A liquid-alloy drop was obtained on a sintered magnet tip when it was heated with an electron beam. The fast cylinder rotation with a linear surface velocity of 50 m/s drew out the liquid-alloy solidifying in the microwires form, close to cylindrical, with the length of 10–20 mm and diameter 50–100 μm (Fig. 1, *a*). We have earlier advised about the study of the chemical and phase composition of microwires using transmission electron microscopy (TEM), X-ray diffraction analysis (XRD), phase contrast of backward scattered electrons (BSE), X-ray photoelectron spectroscopy (XPS) and energy-dispersion spectra (EDX) obtained in a scanning electron microscope (SEM) [6,7].

We used a high-resolution transmission electron microscope HR TEM JEOL to obtain electron diffraction images, which assured that the bulk of the microwires is an amorphous phase demonstrating a halo (Fig. 1, *b*). The integral magnetic moment of the microwire and its dependence on the field and time were recorded in MPMS XL Quantum design SQUID magnetometer at 300 K.

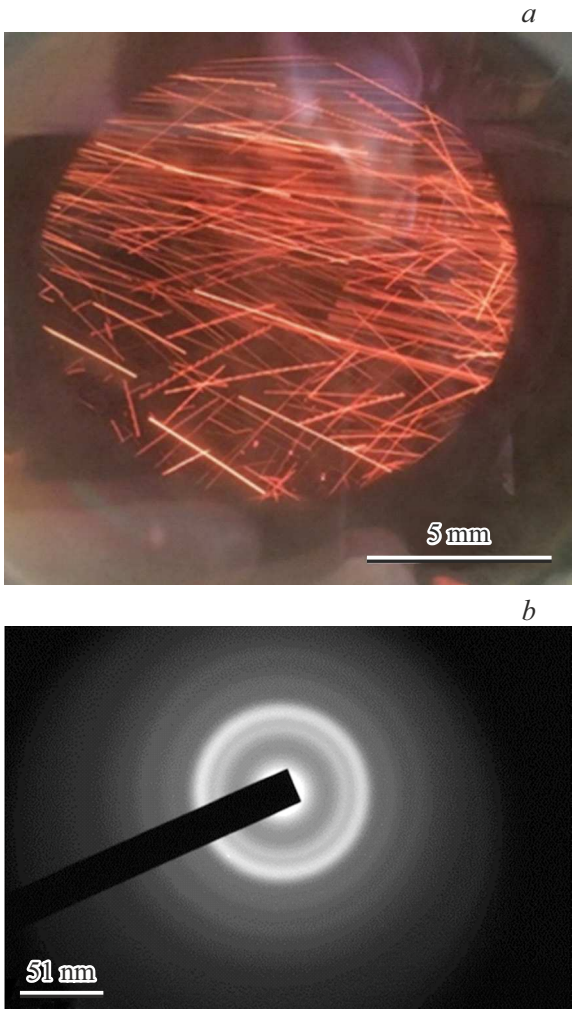

Figure 1. (*a*) Formation of microwires in the process of drawing out liquid-alloy on the cooling cylinder. (*b*) Electron diffraction image obtained for a lamella cut from a microwire (slit width is 51 nm).

3. Experimental findings

The microwire magnetic moment dependence on the magnetic field is shown in Fig. 2, *a*. The figure shows that the approximation to saturation is characterized by a long, slightly sloping section of the $M(H)$ curve, that is not typical of the ferromagnet state. To test the feasibility of the theory predictions, the section where $M/M_S > 0.85$ (as it is usually done in the studies devoted to the analysis of the field dependence of magnetization) was built in the coordinates $M(1/H^2)$, where a rectification of the field dependence of the magnetic moment was observed (Fig. 2, *b*).

Further we have carried out the experiments relating to the study of magnetic moment relaxation under different

conditions. In the first series of experiments the field was abruptly switched over from the value +1 kOe to zero value, and after the establishment of the zero value (in about 40 s), the recording of the dependence of the magnetic moment on time began. The results are shown in Fig. 3, *a*. The figure shows that in semilogarithmic coordinates this dependence does not rectify, although when we studied polycrystalline magnets of similar chemical composition, this rectification took place [19]. Thus, in a zero magnetic field, when it is expected that an amorphous microwire is in a spin-glass state, the magnetic relaxation of its magnetic moment differs from the ferrimagnet logarithmic relaxation.

In the second series of experiments aimed at studying the magnetic relaxation dynamics, the magnetic field was abruptly switched over from the value +1 kOe to the negative value −1 kOe (i.e., the direction of the external magnetic field was changed). Since it was previously discovered that at high temperatures in a magnetic field 1 kOe the SSG → FM transition occurs, it could be expected that the microwire in a magnetic field greater than the threshold value is in a ferrimagnetic state. Fig. 3, *b* shows the microwire magnetic moment relaxation recorded after switching over the field from +1 kOe to −1 kOe. We can see that in the same time range as within the first series of experiments, there is a logarithmic dependence of the magnetic moment on time, that was previously observed in polycrystalline materials of close chemical composition [20].

Thus, in amorphous PrDyFeCoB microwires we can see a slow approximation of magnetization to saturation

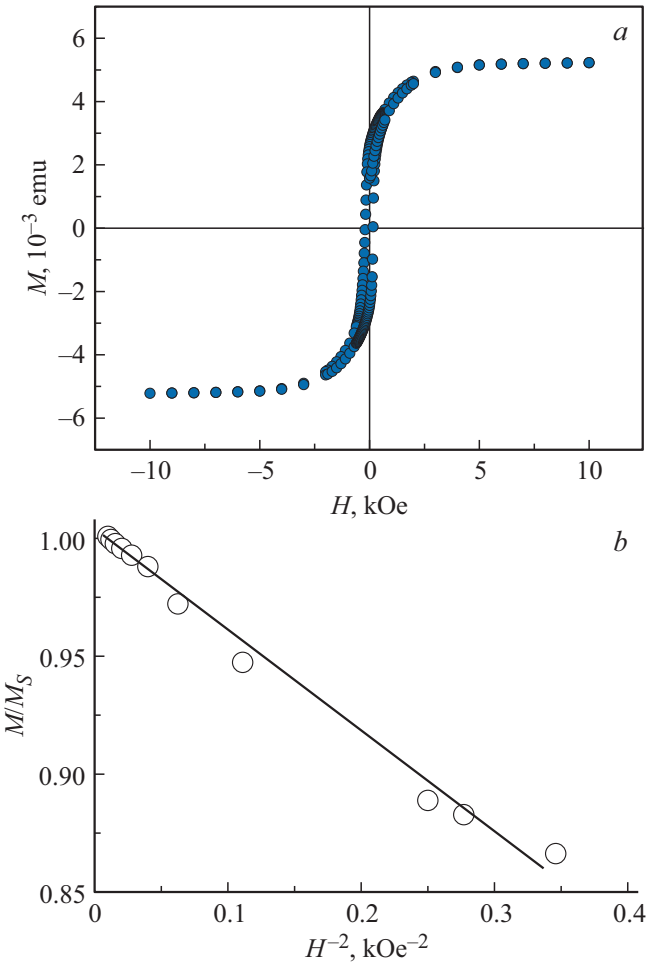

Figure 2. Dependence of the microwire magnetic moment on the magnetic field (*a*), and the obtained from it the fragment of dependence of the brought to saturation magnetization magnetic moment M/M_S on $1/H^2$ (*b*) value. Solid line show the approximation discussed in the text.

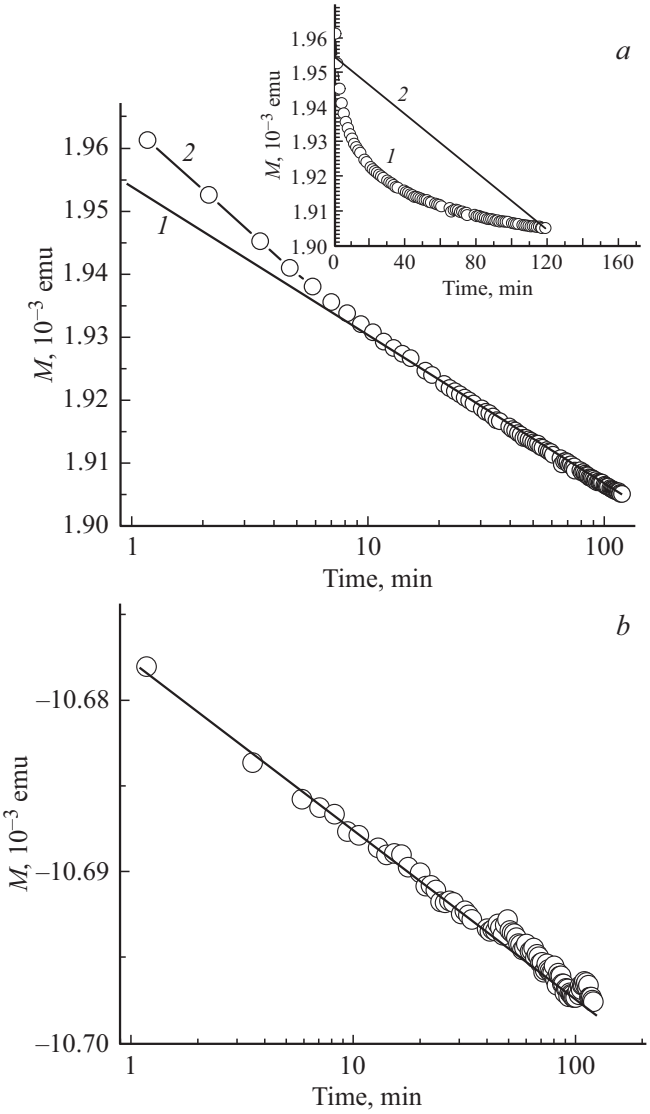

Figure 3. (*a*) Dependence of the microwire magnetic moment on time after switching off the magnetic field 1 kOe to zero. Solid line *1* shows logarithmic dependence $M = S_0 \ln(t/\tau)$, and solid line *2* — approximation by formula (5) described in the text. The same dependence with a logarithmic scale on the vertical axis and a linear scale on the horizontal axis is shown on the inset. (*b*) Dependence of the microwire magnetic moment on time after switching off the field from +1 kOe to a negative value −1 kOe. The solid line shows logarithmic dependence $M = S_0 \ln(t/\tau)$.

as far as magnetic field grows, as well as a deviation of the magnetization reversal time dependence from the logarithmic dependence in zero magnetic field.

4. Discussion of experimental findings

According to the theory by E.M. Chudnovsky and his colleagues, the field dependence of the change of the magnetic moment of ΔM sample should correspond to the following predictions. In „weak“ fields, which are less than the exchange interaction value, expressed in field units, i.e. at $H \ll H_{ex}$:

$$\Delta M/M_S(H) = v/30(H_r/H_{ex})^2(H_{ex}/H)^{1/2}, \qquad (1)$$

in „strong“ fields, which are greater than the exchange interaction value, i.e. at $H \gg H_{ex}$:

$$\Delta M/M_S(H) = 1/15(H_r/H)^2. \qquad (2)$$

We have used equation (2) to plot $M(1/H^2)$ dependence, and we assured of its correctness (Fig. 2, *b*).

Going to a discussion of the dependence of magnetization on time, we should note that the classical function describing the ferromagnet relaxation with a wide range of domain walls obstacles (with an equally distributed probability density of the obstacles amplitude) usually obeys a logarithmic dependence:

$$M(t) = S_0 \ln(t/\tau), \qquad (3)$$

where S — is a constant value called magnetic viscosity, and a τ — is a constant characterizing the process duration. Such a dependence would have to be straightened in semilogarithmic coordinates $M \sim \lg(t/\tau)$, however, from Fig. 3, *a* it follows that there is no rectification (solid line *1* in Fig. 3, *b* corresponds to function (3)). In a perfect ferromagnet, this might indicate that the energy spectrum of obstacles differs from equidistribution. However, it is obvious that sintered magnets of this type, a rapidly cooled liquid-alloy, are objects with a wide range of obstacles and it is difficult to expect a different distribution in them. Besides, the probability density of obstacles energy in the form of a narrow function close to the δ-function should result in an exponential dependence $M(t) \sim \exp(-t/t_0)$. The time dependences of the magnetic moment in a spin glass can also obey an exponential function with an exponent $0 < n < 1$ [4]. At temperatures below „freezing“ temperature of chaotic spin orientations, the time dependence of the magnetic moment is as follows:

$$M(t) \sim \exp(-t/t_0)^n. \qquad (4)$$

However, neither a purely exponential function nor an exponential function with degree (4) describe the obtained time dependence, as the deviation from the line in the coordinates $\lg(M(t))$ becomes even greater (see the inset in Fig. 3, *a*). Another way to describe the dynamics of magnetization reversal of spin-glass materials is the function obtained within fractal-cluster model described in the introduction:

$$\partial M/\partial t = A(t/\tau)^{-k} \exp(-t/\tau)^{-kf}. \qquad (5)$$

In this function k and f — are static and dynamic exponents, and $\tau \sim 10^{-13}$ s — is the time constant characterizing the duration of the spin transition under spin relaxation. The approximation of the dependence $M(t)$ by function (5) is shown by the line *2* in Fig. 3, *a*. As you can see, not only the field dependence (Fig. 2, *b*), but also the time dependence (Fig. 3, *a*) are well described within the same fractal-cluster model.

Finally, the possibility to initiate SSG → FM magnetic transition with a magnetic field, that was discovered in [6,7,17], shows that magnetic relaxation in the same microwire can be typical of a ferromagnet, if a spin-glass state is suppressed by the field. To check this assumption, the dynamics of magnetization reversal in the negative (opposite in direction) magnetic field was constructed, where the logarithmic dependence $M = S_0 \ln(t/\tau)$ (Fig. 3, *b*) was observed. This fact means that 1 kOe magnetic field is sufficient for magnetization reversal dynamics to correspond to a ferrimagnetic with a wide range of domain walls movement obstacles.

In Fig. 4 the border between SSG and FM states, determined by a microwire magnetic moment surge measured at different temperatures and magnetic fields, is shown. This border represents the dependence of transition critical field H_{cr} on transition critical temperature T_{cr}. In many articles devoted to Almeida–Thouless transition, the dependence is

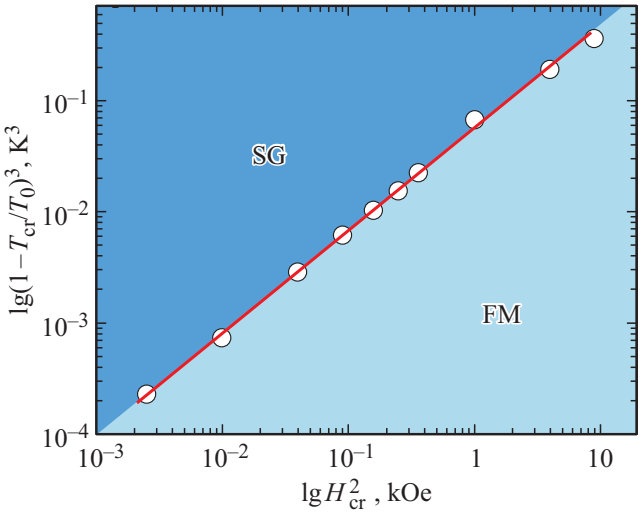

Figure 4. The border between spin-glass (SSG) and ferrimagnetic (FM) states of a microwire in the field–temperature space $(H_{cr}^2 - T_{cr}^3)$. The solid line shows the approximation by Almeida–Thouless formula.

described by the formula [21]:

$$H_{cr} = H_0\left(1 - T_{cr}(H)/T_{SG}(0)\right)^{\alpha}, \qquad (6)$$

where exponent α plays a key role. It defines the type of spin glass. There are two threshold cases: Heisenberg spin glass, characterized by $\alpha = 3/2$ value and Ising spin glass, for which α may differ from 3/2. We have plotted the dependence of the critical field square H_{cr}^2 on the critical temperature cube T_{cr}^3, in order to check, whether phase boundary is a straight line in these coordinates. Fig. 4 shows that the transition condition SSG → FM is fulfilled in the investigated microwires.

5. Conclusions

1) PrDyFeCoB microwires with a high proportion of the amorphous phase up to 86% obtained, where approximation of magnetization to saturation obeys the well-known regularity $M \sim 1/H^2$, obtained within cluster-percolation approach for spin glass.

2) The dynamics of magnetization reversal in a zero field, where the spin-glass state is not suppressed, obeys the predictions of cluster-percolation theory in spin glasses.

3) The dynamics of magnetization reversal in a nonzero field 1 kOe, where the spin-glass state is suppressed and a transition into a ferrimagnetic state occurred, obeys a logarithmic dependence, widely known for ferromagnets with an equiprobable energy distribution of domain walls movement obstacles.

4) The data obtained in combination with the previously discovered SSG → FM transition border indicate that amorphous PrDyFeCoB microwires are spin glass with an unusual spin disorder supported not by variations of exchange interaction (as it takes place in transitional amorphous alloys), but by variation of random directions of local axes of single-ion anisotropy of Pr^{3+} and Dy^{3+} ions.

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Conflict of interest

The authors declare that they have no conflict of interest.

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