Magnetic properties and phase transitions in amorphous Er–Fe alloys

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An investigation was made of magnetic properties of amorphous Er–Fe alloys. A new model was developed for the description of the magnetic state of amorphous magnetic materials based on a hypothesis of the existence of a stochastic domain structure (of the Imry–Ma type) in these alloys and on the concept of a random crystal field acting on the f ions and considered in the Harris–Plischke–Zuckermann approximation. A microscopic description of the $4f$ subsystem made it possible to determine the temperature dependences of the anisotropy constant of domains, of the total magnetization, and of the coercive force, as well as the field dependences of the magnetization. Good agreement between the experimental results and those predicted by theoretical modeling was achieved.

1. INTRODUCTION

In spite of the considerable progress made in the study of magnetic properties of amorphous rare earth–transition metal (R–TM) alloys, some important problems in the physics of magnetic phenomena of these materials are far from the final solution. This applies, for example, to the investigations[1–3] of the behavior of these alloys in an external magnetic field below the Curie point and in the region of the compensation temperature: the influence of the local magnetic anisotropy on the magnetization processes had not been investigated sufficiently thoroughly. The difficulty encountered in the description of magnetic properties of an amorphous alloy is that both chemical and structural atomic disorder results in fluctuations of all the parameters of the magnetic system: the exchange interactions, the magnetic moment, and the magnetic anisotropy.

Earlier theoretical models[1–3] treated alloys containing one magnetic moment, whereas the magnetic properties of R–TM alloys are governed largely by the specific properties of the $4f$ and $3d$ ions, and by their interaction. The present paper reports investigations of certain magnetic properties of amorphous Er_xFe_{100-x} alloys and proposes a theoretical model for the description of the temperature and field dependence of the magnetization of these alloys.

2. SAMPLES AND EXPERIMENTAL METHODS

The Er_xFe_{100-x} system ($x = 26, 32, 54$) of amorphous alloys was prepared in the laboratory of I. V. Zolotukhin by the method of ion-plasma sputtering from a mosaic target in an atmosphere of spectroscopically pure argon on a substrate cooled with liquid nitrogen. The substrate was chemically pure Al and its thickness was 70 μm. The thickness of an amorphous film on the substrate was 20–30 μm. X-ray structure analysis showed that at room temperature all the alloy samples were amorphous.

The magnetization was investigated in the temperature range 4.2–300 K in fields up to 15 kOe using a vibrating-sample magnetometer. Investigations of the susceptibility were carried out by the standard method in the temperature range 4.2–300 K using fields up to 10 Oe at frequencies 70–1000 Hz.

3. EXPERIMENTAL RESULTS

All the amorphous Er_xFe_{100-x} alloys we investigated were isotropic, i.e., their remanent magnetization was zero along any direction of measurement. At room temperature none of the alloys exhibited magnetic ordering: they were all paramagnetic. The temperature dependence of the magnetization was determined as follows. An alloy was cooled to $T = 4.2$ K in the absence of a magnetic field and then the temperature dependence of the magnetization was deter-

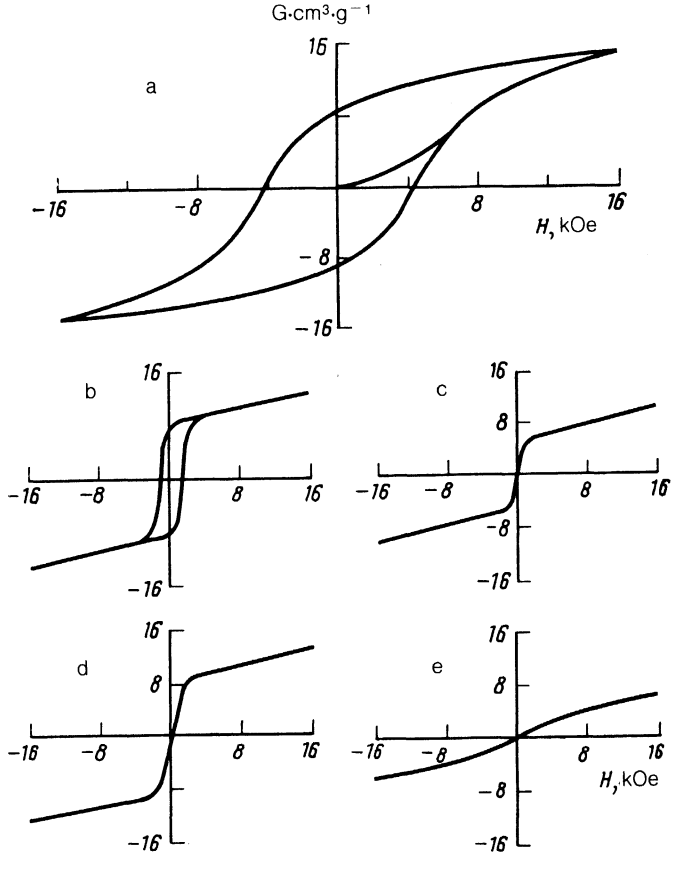

FIG. 1. Hysteresis loops and initial magnetization curves of an amorphous sample of $Er_{26}Fe_{74}$ recorded at various temperatures: a) 28 K; b) 65 K; c) 104 K; d) 185 K; e) 250 K.

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mined in various magnetic fields in the course of heating of a sample. Both hysteresis loops and magnetization curves were recorded after demagnetization of a sample at the measurement temperature.

Cooling induced magnetic order in these samples. Figure 1 shows the hysteresis loops and magnetization curves of $Er_{26}Fe_{74}$ obtained at various temperatures. It was found that a magnetic hysteresis loop expanded greatly as a result of cooling and the coercive force rose strongly. Similar hysteresis loops were obtained also for the other alloys.

Figure 2 illustrates the temperature dependence of the magnetization σ of Er_xFe_{100-x} alloys determined during heating in various magnetic fields, as well as the temperature dependence of the magnetic susceptibility χ. It is clear from this figure that the $\sigma(T)$ curves had maxima due to the appearance of a magnetic order. The Curie temperatures T_C deduced from the magnetization curves by the method of thermodynamic coefficients[4] amounted to 223, 173, and 118 K for the alloys with $x = 26$, 32, and 54, respectively. Maxima of the $\chi(T)$ dependence were located 3–4 K below the Curie temperatures of the corresponding alloys and were found (insets in Fig. 2) by the thermodynamic method from the H/σ dependence.

The $\chi(T)$ curves obtained near the Curie temperatures for $Er_{32}Fe_{68}$ and $Er_{54}Fe_{46}$ had two or more maxima. The more complex nature of the dependence $\chi(T)$ exhibited by amorphous alloys near the Curie temperature was explained in Ref. 5 by the existence of additional maxima because of a magnetic resonance and because the temperature dependence of the anisotropy constant passed through zero. The absence of additional information prevented us from accounting for the additional maxima exhibited by the $\chi(T)$ curves.

Lowering the Curie temperature as the Er content increased indicated that the main contribution to the exchange interactions came from the *d–d* exchange. A smooth variation of the specific magnetization of the alloys near the Curie temperature indicated that the phase transition was of the "smeared-out" type, which was known to be a characteristic feature of amorphous R–TM alloys where local fluctuations of the magnetic anisotropy energy near T_C are quite large.[6]

Cooling below T_C in the temperature range 30–100 K revealed maxima of the $\sigma(T)$ curves of all the Er_xFe_{100-x} alloys (Fig. 2). The temperatures of these maxima depended strongly on the magnetic field in which a sample was heated. The insets in Fig. 2 show the temperature dependence of the

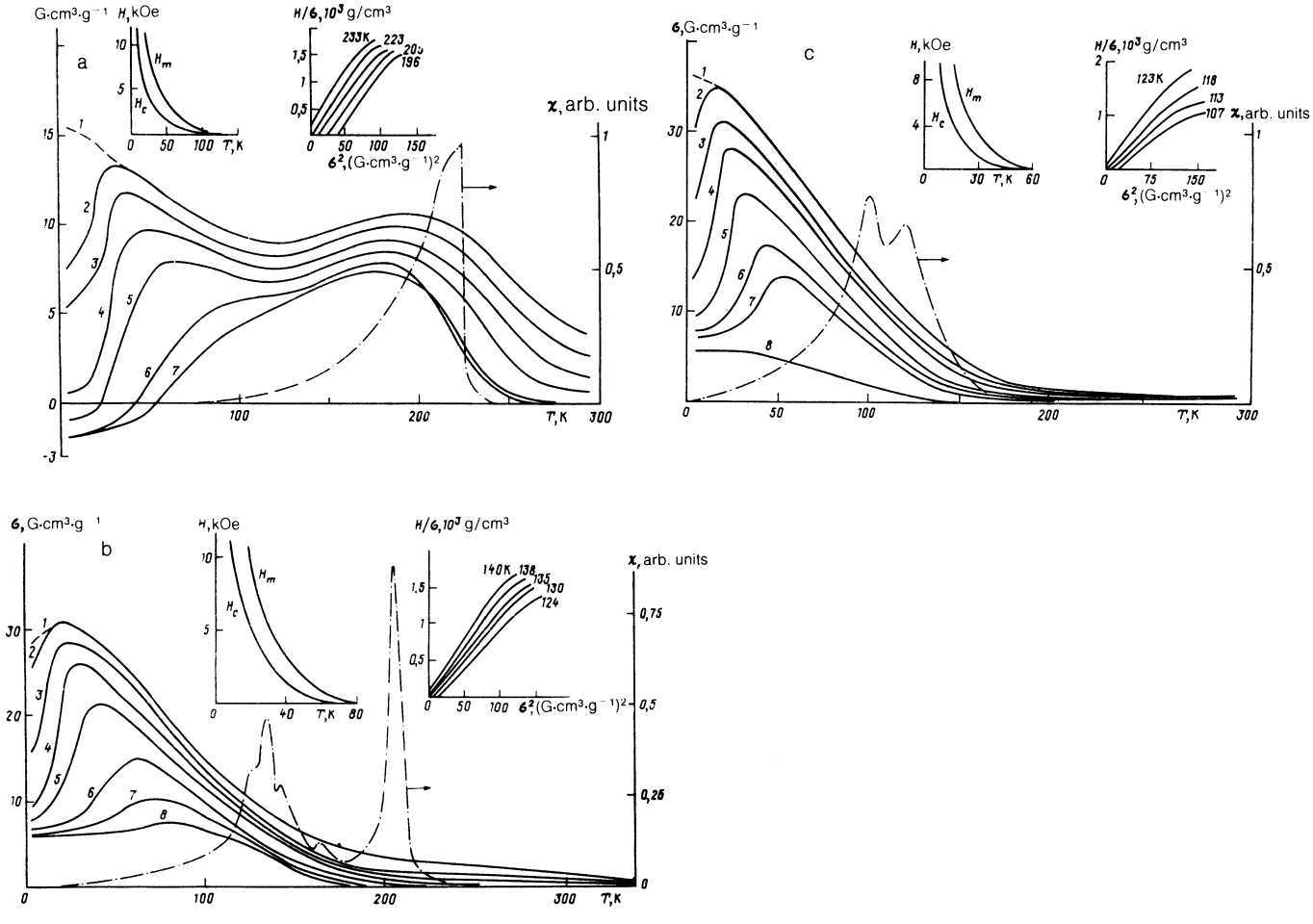

FIG. 2. Temperature dependences of the specific magnetization of amorphous $Er_{26}Fe_{74}$ (*a*) $Er_{32}Fe_{68}$ (*b*), and $Er_{54}Fe_{46}$ (*c*) alloys. Curves labeled 1 represent measurements carried out during heating in a field of 14 kOe after cooling to $T = 4.2$ *K* in a field of 14 kOe. Curves 2–7 represent the results obtained after cooling in zero field and heating in the following fields: 2) $H = 14$ kOe; 3) 10 kOe; 4) 6 kOe; 5) 3 kOe; 6) 1 kOe; 7) 0.4 kOe (*a*, *c*) and 0.5 kOe (*b*); 8) 0.2 kOe (*b*) and 0.05 kOe (*c*). The chain curves are the temperature dependences of the initial magnetic susceptibility $\chi(T)$ at $f = 200$ Hz in a field $H = 5$ Oe. The insets show the temperature dependence of H_c and H_m, as well as the dependence of H/σ on σ^2.

fields H_m at which $\sigma(T)$ reached maximum values. For comparison, the insets in Fig. 2 showed also the temperature dependence of the coercive force H_c. It is clear from the figures that throughout the investigated temperature range the inequality $H_m > H_c$ was obeyed. The temperature and field dependences of the magnetization can be explained employing the following model.

4. THEORY

It is difficult to provide a satisfactory microscopic description of the investigated structurally disordered magnetic materials. Studies of the influence of the field of the immediate environment on the energy spectrum of a magnetic ion, selection among the various mechanisms of the exchange interaction, determination of the signs and magnitudes of the exchange integrals all require detailed information on the structure and properties of the environment. However, such information cannot be deduced directly from the experiments. We can only measure the quantities which are averaged over the whole volume of a sample or a part of it.

It is therefore necessary to use simple but detailed model representations. In the continuum approximation the problem of determining the magnetic state of an amorphous magnetic material reduces to a variational problem and the coefficients of the density of the thermodynamic potential are random functions. The moments (average values, variances, correlation functions, etc.) of these quantities should also be specified in model or phenomenological forms. The situation is complicated by the absence of small parameters, the use of which can linearize or simplify the Euler–Lagrange nonlinear differential equations. Therefore, we make certain simplifying assumptions which reduce the problem to the algebraic form, as shown below.

It is known that the initial magnetic state of amorphous R–TM alloys represents a macroscopically disordered structure. Nevertheless, if we consider sufficiently small regions of a sample, we find that they are magnetically ordered and form as a whole a characteristic domain structure. We assume that the domain structure in our compounds is random in the sense that the orientation of the magnetization vectors in the domains is random. The possibility that such structures appear in strongly fluctuating systems was pointed out by Imry and Ma.[6]

The dimensions of these domains are determined by the competition between the magnetostatic exchange interactions and by the anisotropic effects governing primarily the orientation of the domain magnetization. The application of an external field induces a change in the domain structure, which gives rise to a macroscopic magnetization. We assume that the processes of rotation of the magnetization predominate over the processes of displacement of domain walls. This hypothesis is supported by the high coercivity and low mobility of domain walls in amorphous magnetic materials, demonstrating the presence of a large number of scattering and pinning centers, and also by the observation that such walls are not generally of the 180° type. The thermodynamic potential of such a system can therefore be written in the form

$$\Phi=\Phi_0+\sum_i \Phi_i+\sum_{i,j}\Phi_{ij}, \tag{1}$$

where the last term describes the interaction between domains and transition regions between them (domain "walls"). It follows from the above discussion that we can ignore this term. The summation is carried out over all the domains and the thermodynamic potential of the ith domain can be represented in the form

$$\Phi_i=-M'(T)H\cos\varphi_i-K(T)\cos^2(\varphi_i-\psi_i), \tag{1a}$$

where $M'(T)$ is the magnetization of a domain, $K(T)$ is the anisotropy constant, φ_i is the angle between the direction of magnetization of the ith domain and the external field **H**, and ψ_i is the angle between the direction of **H** and the anisotropy axis of a domain.

The magnetic state of this system can be found by minimization of Eq. (1) with respect to the angles φ_i, allowing for the conditions of stability of the resultant solutions. In fact, the problem reduces to a study of the process of magnetization of a ferromagnet in a tilted field[7] followed by summation over all the domains or by averaging over the angle ψ, which governs the direction of the easy magnetization axes of the domains. If we assume that the magnetization vectors of the domains fill a sphere with a uniform probability density (in the absence of an external field), we find that the magnetization per chemical formula unit can be written in the form

$$I=\frac{M'(T)}{2}\int_0^{\pi}\cos[\varphi(\psi,H,T)]\sin\psi\,d\psi, \tag{2}$$

which is analogous to the Tikazumi model of "magnetic needles." Obviously, such a stochastic structure may appear in amorphous films of considerable thickness (40–60 μm) when the "thin film" properties (transverse magnetic anisotropy, "columnar" structure, etc.) are absent.

The temperature dependences of the magnetization and the anisotropy constant can be used in a quantum-mechanical description of the system of the 4*f* ions. Usually the 4*f* ions, apart from the S ions of Gd^{3+} and Eu^{2+}, are strongly anisotropic. The Hamiltonian of the crystal field acting on an *f* ion in an amorphous host material can be represented in the following form on the basis of the Harris–Plischke–Zuckermann model:[1]

$$\mathcal{H}_{cr}=-D\sum_i (\hat{J}_{z_i}^{(i)})^2, \tag{3}$$

where $\hat{J}^{(i)}$ is the magnetic moment of the ith ion, the summation is carried out over all the *f* ions, the constant satisfies $D>0$ and is the same for all positions, and the orientation of the z_i axis varies at random from one position to another. This model provides a satisfactory description of the properties of heavy rare earths. As pointed out already, the *d–d* exchange interaction in R–TM alloys predominates over the *f–d* exchange and is much stronger than the *f–f* exchange. Therefore, in the first approximation we can regard the *f* subsystem as an ideal paramagnet which is in an effective field $\mathbf{H}_{\text{eff}}=\mathbf{H}-\Lambda\mathbf{M}$, where Λ is the *f–d* exchange interaction constant and **M** is the magnetization of the 3*d* subsystem which we treat as "saturated" and dependent only on temperature. The characteristic splitting of a multiplet in the crystal field is $\Delta_{cr}\sim 100\ \text{cm}^{-1}$, whereas the Zeeman splitting is $\Delta_H\sim 1-10\ \text{cm}^{-1}$, so that the Zeeman interaction with the field $\mathbf{H}_{\text{eff}}$ can be regarded as a perturbation compared

with the Hamiltonian (3). If we calculate the thermodynamic potential of an R ion, we find that to first order of perturbation theory

$$\Phi_R=-k_B T\ln\left[2\,\mathrm{ch}\left(\frac{\mu n_\alpha H_{eff}}{k_B T}\right)\right], \tag{4}$$

where k_B is the Boltzmann constant, $\mathbf{n}_\alpha$ is a unit vector along the z_i axis, and μ is the magnetic moment of an ion. The thermodynamic potential of a "domain" is then

$$\Phi_i=-\mathbf{MH}\cos\varphi-\frac{1}{V_0}\sum_{V_0}\Phi_R-\Phi_{an}, \tag{5}$$

where the first term is the energy of the interaction of the $3d$ subsystem with the field $\mathbf{H}$, V_0 is the volume of a domain, and Φ_{an} describes the anisotropic effects. If the orientation of the z_i axes within a domain has a specific direction, then the anisotropy appears when the second term is discussed. Introducing the anisotropic probability density of orientations of the z_i axes

$$\rho(\theta)=\frac{1+\gamma\cos^2\theta}{1+\gamma/3}, \tag{6}$$

where $\gamma\ll 1$ is the nonisotropy parameter and θ is the angle between the vector representing the preferred direction and $\mathbf{n}_\alpha$, we find that

$$\frac{1}{V_0}\sum\Phi_R=\langle\Phi_R\rangle=x\int_0^{\pi/2}\Phi_R(\theta)\rho(\theta)\sin\theta\,d\theta, \tag{7}$$

where x is the relative concentration of the f ions.

Expanding the thermodynamic potential of Eq. (4) in powers of the parameter $\xi=\mu n_\alpha H_{\mathrm{eff}}/k_B T$ and retaining only terms of second order, we find the anisotropy constant is

$$K(T)=\frac{\gamma x\mu^2\Lambda^2M^2}{15k_B T}=K'(0)\frac{M^2(T)}{T}=K_0\frac{M^2(T)}{1-\tau}, \tag{8}$$

where $\tau=1-T/T_c$.

The temperature dependence of the magnetization of the d subsystem of a domain in an amorphous alloy can be represented in the form

$$M'(T)\sim M(T)=M_0\tau^\delta, \tag{9}$$

where δ is a dimensionless parameter. The temperature dependence of the magnetization of the $3d$ subsystem and of the alloy as a whole can be approximated by the expression $M(T)\propto M'(T)\propto\tau^\delta$, where $M'(T)=M(T)(1-\chi_R(T))$. The parameter δ will be regarded as variational and its value should be obtained from the experimental data.

We divide the expression for the thermodynamic potential of Eq. (1a) by Eq. (8), which yields

$$\Phi_i/K(T)=-\beta\cos\varphi-\cos^2(\varphi-\psi), \tag{10}$$

where

$$\beta=\frac{H}{K_0M_0}\frac{1-\tau}{\tau^\delta}. \tag{11}$$

An important feature of this model is that the parameters representing an amorphous alloy (δ, K_0, M_0, and T) occur as a universal combination characterized by the dimensionless parameter β. Minimization of Eq. (10) yields the following equation for the determination of the angle:

$$\beta\sin\varphi=\sin 2(\varphi-\psi), \tag{12}$$

which makes it possible to find the solution comparable with the stability condition.

Equation (12) can generally have multivalued solutions. For example, if $H=0$, there are two stable solutions: $\varphi_1=\psi$ and $\varphi_2=\psi+\pi$, whereas in the presence of a field the solution φ_1 (if $\psi>\pi/2$) or the solution φ_2 (if $\psi<\pi/2$) becomes metastable and loses its stability at a value of β deduced from the condition[7]

$$\cos^{2/3}\psi+\sin^{2/3}\psi=(2\beta^{-1})^{2/3}. \tag{13}$$

An analysis of the stability condition (13) shows that the magnetic state of the system can be divided into three regions:

a) $\beta<1$ is the range of weak fields or low temperatures; in this case the magnetization process involves continuous rotation of the magnetization vectors of the domains (see Fig. 5 below);

b) $1<\beta<2$ is the intermediate range where reorientation of some of the domain magnetization vectors with $\psi>\pi/2$ occurs; the process becomes irreversible and a steep rise of the magnetization is observed;

c) $\beta>2$ corresponds to completion of the abrupt reorientation processes and represents "pulling" of the magnetization vectors toward the external field; the magnetization falls as the Curie point is approached.

Approximate analytic expressions can be obtained for all three regions. This can be done by linearizing the equation for the extremum given by Eq. (12) in the regions a and b, assuming that the difference $\delta=\psi-\varphi$ is a small parameter, while in the region c it can be done by considering the angle φ itself as a small parameter. The asymptotic expression for the rotation region ($\beta<1$) is

$$I(\beta)=\frac{M'(T)}{2}\left(\frac{1}{4}+\frac{1}{\beta^2}\right)\ln\left[\left(\frac{\beta+2}{2-\beta}\right)^2-\frac{2}{\beta}\right], \tag{14a}$$

while in the "pulling" region ($\beta>2$), the corresponding expression is

$$I(\beta)=\frac{M'(T)}{3}\left[4-\mathrm{arctg}\frac{1}{(\beta-2)^{1/2}}\right]\left[(\beta-2)^{1/2}+\frac{1}{3(\beta-2)^{1/2}}\right]. \tag{14b}$$

In the region b the expressions for $I(\beta)$ are complicated and will not be given here. Using Eqs. (14a) and (14b), we can determine the numerical values of the phenomenological parameters employed in the model by measuring the initial susceptibility, which can be deduced from Eq. (14a), or by investigating the law governing the approach of the magnetization to saturation [Eq. (14b)]. The expressions given by Eqs. (14a) and (14b) are difficult to analyze and, moreover, they are asymptotic. We therefore obtained a numerical solution of Eq. (12) allowing for the loss of the stability described by Eq. (13). A typical calculated result is presented in Fig. 5 (curve 3). A comparison of the numerical solution with the asymptotic expressions demonstrated good agreement between the system (14) and the results of numerical analysis, with the exception of the region $\beta\sim 2$, because the

asymptote given by Eq. (14b) then diverges. It is clear from Fig. 5 that the best quantitative agreement was observed in the intermediate range of temperatures $0 < T < T_C$. The discrepancy at low temperatures could easily be explained by noting that one should then allow more rigorously for the temperature dependence of the anisotropy constant. In fact, in accordance with the microscopic model put forward above, the expression (8) for the anisotropy constant is invalid at low temperatures. Therefore, it may be necessary to allow for the higher terms in the expansion of the thermodynamic potential given by Eq. (4). The deviations observed at high temperatures can be explained, using a not very satisfactory description of a two-sublattice magnetic material with a complex temperature dependence of the sublattice parameters, by a simple power-law dependence τ^{δ}.

We now consider the temperature dependence of the coercive force H_c and of the field at the maxima H_m. The process of magnetization reversal can easily be discussed on the basis of the same model. The limiting hysteresis loop corresponds to the residual magnetization, which represents the orientations of the domain magnetization vectors filling a hemisphere. When the field is applied in the opposite direction, a numerical calculation can yield that field in which the magnetization vanishes:

$$H_c = 0{,}96 K_0 M_0 \tau^{\delta}/(1-\tau) \qquad (15)$$

($\beta_c = 0.96$), which corresponds to the very onset of the magnetization switching process.

The field at the maxima $H_m(T)$ can readily be shown to correspond to $\beta = 2$ (completion of the magnetization switching processes) at moderate temperatures when the temperature dependence $M(T)$ "swamps" the rise of the magnetization because of the rotation. Therefore, we have

$$H_m(T) \approx 2H_c(T) \qquad (16)$$

and the experimental results shown in the insets in Fig. 2 can be explained satisfactorily. When temperature is increased, the $H_c(T)$ and $H_m(T)$ curves approach one another because of the more rapid fall of H_m with increasing temperature, as is indeed observed experimentally (Fig. 2).

5. DISCUSSION OF RESULTS AND CONCLUSIONS

The relationship (16) is independent of the parameters of the compounds and should be universally valid. Nevertheless, in the case of different compositions exhibiting similar dependences $\sigma(T)$, $H_c(T)$, and $H_m(T)$ [Tb–Co, Tb–Fe, (Tb–Dy)Fe], the ratio $\varepsilon = H_m/H_c$ has been found to vary from 1.5 to 3. Such deviations clearly result because the Imry–Ma domain structure may be suppressed gradually (or the spherical symmetry of the distribution of the domain magnetization vectors may be distorted) as the content of the $3d$ metal and the difference between the $4f$ and $3d$ metals increase, becoming an antiparallel distribution of the magnetization in neighboring domains. Equation (15) is derived on the assumption of an equiprobable orientation of the easy magnetization axes of the domains (corresponding to total chaos). Under realistic conditions and particularly in the case of sufficiently thin films we need to allow for possible deviations from the hypothesis of total chaos of easy axes. In the case of amorphous films we can expect primarily two types of a regular domain structure: a planar orientation of the domain magnetization vectors as well as domains with the magnetization vectors oriented at right-angles to the plane of the amorphous film. It is therefore reasonable to assume the existence of a preferential orientation of the domain easy magnetization axes either at right-angles to the plane of the amorphous film or mainly in the plane of this film. In the former case the values of the field at the maxima and of the coercive force differ most from one another ($\varepsilon \sim 3$) because of a reduction in the coercive force; in the latter case the difference between H_c and H_m should decrease because of an increase in H_c. The field at the maxima H_m is practically independent of the geometry of the structure. We therefore assume that, having determined ε, we can

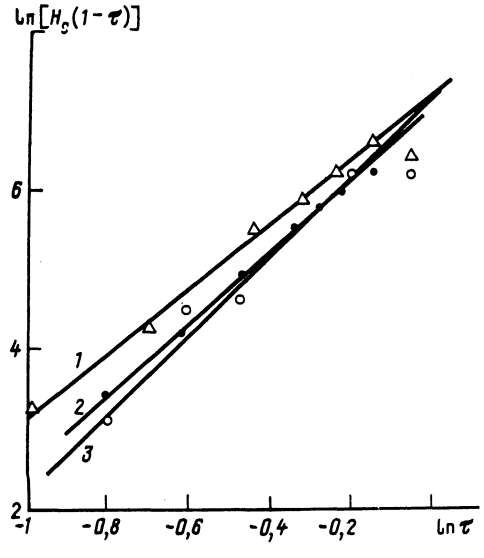

FIG. 3. Dependence of $\ln[H_c(1-\tau)]$ on $\ln\tau$, where $\tau = (T_C - T)/T_C$ are plotted for the following alloys: 1) $Er_{32}Fe_{68}$; 2) $Er_{26}Fe_{74}$; 3) $Er_{54}Fe_{46}$.

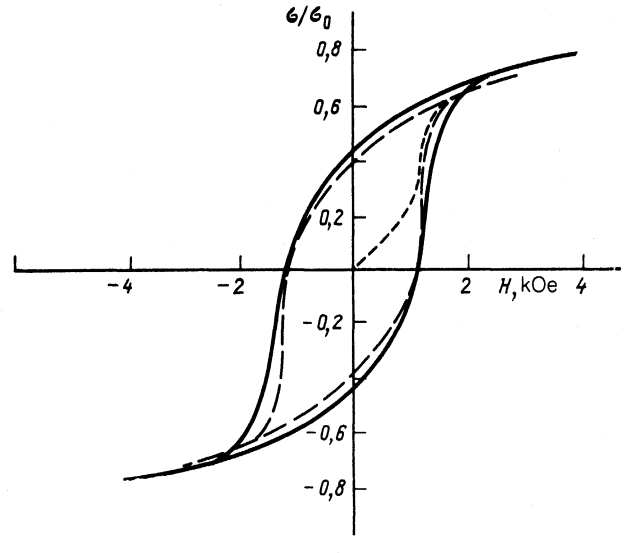

FIG. 4. Experimental (continuous curves) and theoretical (dashed curves) hysteresis loops of $Er_{32}Fe_{68}$ at $T = 37$ K.

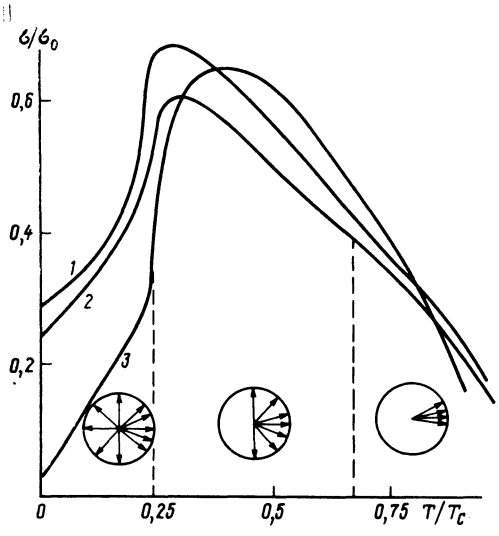

FIG. 5. Experimental (curve 1 for $Er_{32}Fe_{68}$ and curve 2 for $Er_{26}Fe_{74}$); and theoretically calculated (curve 3) temperature dependence of the magnetization (after cooling in zero field) recorded during heating in a field of 3 kOe. The distribution of the domain magnetic moments in various temperature ranges are shown schematically at the bottom of the figure.

draw the conclusion that either chaos is present or that a regular domain structure is distributed; the film thickness and the conditions during sputtering are then important control parameters.

Figure 3 shows the dependence of $\ln[H_c(1-\tau)]$ on $\ln \tau$ for amorphous Er_xFe_{100-x} alloys. It is clear from this figure that the experimental points fit a straight line quite well and thus confirm the relationship (15). Hence, we can calculate the parameters K_0, M, and $\delta \approx 4$. Using the numerical values of these parameters, we plotted hysteresis loops (one of which is shown in Fig. 4) and the temperature dependence of the magnetization (Fig. 5). The same figures give the experimental $\sigma(H)$ and $\sigma(T)$ curves. It is clear from these figures that the experimental and theoretical results are in satisfactory quantitative agreement.

We can summarize our results as follows. A new model is proposed for the description of the magnetic state of amorphous magnetic materials based on a hypothesis of the existence of a stochastic domain structure (of the Imry–Ma type) and the concept of a random crystal field acting on the f ions (considered in the Harris–Plischke–Zuckermann approximation). A microscopic description of the f subsystem makes it possible to determine the temperature dependence of the anisotropy constants of domains for one of the important mechanisms, which is the anisotropy of the pair ordering of atoms. Other mechanisms (for example, magnetoelastic) can easily be allowed for on the basis of the same model.

An experimental investigation of the processes of magnetization of Er–Fe amorphous alloys is reported. The experimental results are in good agreement with the theoretical model in which it is assumed that the processes of rotation of the domain magnetization vectors predominate over the displacement processes.

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