Theory of remagnetization curves of dilute random magnets

D. V. Berkov and S. V. Meshkov

Institute for Solid State Physics, Academy of Sciences, USSR

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The hysteresis behavior of a system of dipole-interacting uniaxial ferromagnetic particles of low concentration is investigated. A number of new results are obtained for an ensemble of noninteracting particles. The application of the local-field approximation to remagnetization of dilute magnetics with random or parallel orientation of the easy axis is substantiated. The local field-distribution density is found. Analytic expressions are presented for the field dependence of the magnetization. The remagnetization curves calculated numerically for ensembles of varying concentration are also given.

Systems of classical magnetic moments randomly distributed in space and separated by a relatively large distances, which interact with each other primarily through the dipole interaction, are the subject of this study. The introduced model applies, in the first place, to such experimental objects as ensembles of macroscopic ferromagnetic particles in a solid suspension of the type used in a magnetic recording, but it is also useful for the study of hysteretic properties of dipolar spin glasses.

The energy of an ensemble of interacting dipoles is a complicated function of the angular coordinates of all the moments and, as in other disordered systems, has a large number of local minima—valleys. Equilibrium magnetic properties are studied numerically for spin glass systems mainly in weak magnetic fields, and the system relaxation toward thermodynamic equilibrium is the most interesting problem. For large values of elementary magnetic moments the barriers between valleys are high and, therefore, tunneling and (at low temperatures) excitation transitions are not likely to occur. In this case the question of a thermodynamic equilibrium and relaxation towards it becomes a secondary one, while the system evolution caused by fact that the valleys of potential energy lose stability and vanish when the external magnetic field **H** changes becomes primary. If one completely disregards transitions between valleys and assumes relatively high dissipation, one can ignore the moments dynamics after loss of stability. The problem then becomes one of tracking the minima of potential energy of an ensemble of interacting dipoles. The magnetization **M**(**H**) averaged over the entire system then exhibits hysteretic behavior, which is of principal interest.

When the elementary moments do not have an internal anisotropy, each of them is not collinear with the local magnetic field acting on it, and the hysteresis of **M**(**H**) exists only as collective interaction between the moments. When an internal anisotropy (of crystallographic origin or caused by particle shape) leads to magnetization hysteresis we have a much simpler solution even for noninteracting particles. In this case one can consider the interaction between the moments as a perturbation and look for corrections to the ideal hysteresis loop by expanding in the moment concentration in the magnetic material.

The first calculations for an ensemble of noninteracting single domain ferromagnetic particles were made by Stoner and Wohlfarth.[1] The properties of remagnetization curves for such ensembles with uniaxial, as well as with more complicated anisotropy of particle magnetic characteristics are, in principle, known now (see for example Ref. 2). At the same time a rigorous remagnetization theory for an ensemble of noninteracting particles is still in the initial stage, in spite of long-term research efforts. A successive expansion in the particle concentration in the local-field approximation was used only for random Ising magnetic systems with dipole-dipole interaction under thermodynamic equilibrium.[3] Applied studies that employ various approximations are dominant in the existing literature.[4–6] There is now an effort to model the remagnetization processes numerically using methods of molecular dynamics[7] and semi-empirical systematization of experimental results.[8]

The magnetization hysteresis of a dilute system of identical spherical ferromagnetic particles with an easy anisotropy axis is studied in this paper. The situations with random axes orientation and with axes parallel to an applied field are considered. The equilibrium equation for an elementary moment and its main properties are studied in the first section; the remagnetization curves for an ensemble of noninteracting particles are derived in the second section; in the third section we discuss the justification for the use of the local field approximation. The Holtsmark method for calculation of a three-dimensional local magnetic field distribution function, which turns out to be close to a Lorentzian, is calculated in the fourth section, and on the basis of this expression the remagnetization curves for real dilute magnetic systems with random or parallel easy-axes orientation are constructed numerically in the last two sections. It is possible to derive analytical expressions for a number of characteristic points and regions of these curves, in particular for the value of remanent magnetization and (in an oriented ensemble) coercive force.

1. SINGLE PARTICLE

For simplicity let us assume that all ferromagnetic particles have spherical shapes and identical radii a. In the low temperature region (we limit ourself to only these temperatures) the particle magnetic moment **m** rotates without changing its value $|\mathbf{m}| = I_s V_0$, where I_s is the saturation magnetization, and $V_0 = 4\pi a^3/3$ is the particle volume. The magnetic field will be measured in units of βI_s (β is the magnetic anisotropy constant[9]), and the magnetic moment in units of $I_s V_0$ (i.e., $|\mathbf{m}| = 1$). The energy will correspondingly be measured in units of $\beta I_s^2 V_0$.

In this section we consider only one particle of an en-

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semble in an arbitrarily directed magnetic field **h**. Let us introduce a coordinate system with z' axis parallel to the magnetic field and x' axis in the plane formed by the magnetic field and the particle easy axis. The easy axis direction in the $x'Oz'$ plane is given by the spherical angle θ_0, the direction of the magnetic moment (which, in case of the single-axis anisotropy, lies in the sample plane) by the angle θ. We can assume, without limiting the universality, that θ_0 changes from 0 to $\pi/2$. The particle energy per unit volume is

$$U=-h_z\cos\theta+{}^1/_2\sin^2(\theta-\theta_0). \quad (1)$$

The equilibrium value of the angle θ is determined by

$$\partial U/\partial\theta=h_z\sin\theta+{}^1/_2\sin 2(\theta-\theta_0)=0, \quad (2)$$

which must be complemented by the stability condition

$$\partial^2U/\partial\theta^2=h_z\cos\theta+\cos 2(\theta-\theta_0)>0. \quad (3)$$

Analysis of Eq. (2) shows that for any angle θ_0 there exists a region of weak fields $|h_z|<H_0(\theta_0)$ where two solutions for θ exist, and a region of strong fields $|h_z|>H_0(\theta_0)$ where the solution is unique. This agrees with a physical fact that in a weak magnetic field anisotropy determines two equivalent moment directions, while in a high field the influence of anisotropy is suppressed and the only possible direction for a moment is along the magnetic field. For $h_z=\pm 1/2$ Eq. (2) has a trivial form and two solutions linear in θ_0:

$$\theta_1=\frac{2}{3}\theta_0,\quad \theta_2=\begin{cases}\pi+2\theta_0, & 0\leqslant\theta_0\leqslant\pi/4\\ 2(2\pi+\theta_0)/3, & \pi/4\leqslant\theta_0\leqslant\pi/2\end{cases}. \quad (4)$$

For other field values Eq. (2) requires a numerical solution. The resulting dependence $\theta(h_z)$, given for example in Ref. 2, possesses an obvious physical symmetry: $\theta(h_z)=\theta(-h_z)\pm\pi$.

The critical value of the field $H_0(\theta_0)$ at which one solution vanishes is determined by the set of equations:

$$H_0\sin\theta+{}^1/_2\sin 2(\theta-\theta_0)=0,\quad H_0\cos\theta+\cos 2(\theta-\theta_0)=0,$$

which follows from (2) and from the stability loss condition (3). The solution for this system gives (compare with Ref. 2) an $H_0(\theta_0)$ dependence, which is symmetrical about $\theta_0=\pi/4$:

$$2[(1-H_0^2)/3]^{3/2}=H_0^2\sin 2\theta_0. \quad (5)$$

For $\theta_0=\pi/4$ the $H_0(\theta_0)$ dependence has a minimum $H_0(\theta_0)=1/2$ (at the boundary of the stability range the angle of deflection of the particle moment from the easy axis is $\pi/2$). Near the minima $H_0(\theta_0)$ is given by

$$H_0(\theta_0)={}^1/_2+3(\theta_0-\pi/4)^2. \quad (6)$$

The maximum value of $H_0(\theta_0)$ reached at $\theta_0=0$ and at $\theta_0=\pi/2$ is 1. The asymptotic form of $H_0(\theta_0)$ near $\theta_0=0$ follows from (5):

$$H_0(\theta_0)=1-{}^3/_2\theta_0^{2/3}. \quad (7)$$

A similar form can be obtained for θ_0 close to $\pi/2$.

In order to apply these results to calculations of the remagnetization of an ensemble of particles it is necessary to relate the direction of the moment **m** to the vector of the magnetic field **h** for an arbitrary direction of the particle easy axis, which is given by a unit vector **n**. This relation is determined by the rotation of the initial coordinate system xyz (in which **h, m** and **n** are defined) to the coordinate system $x'y'z'$ used above to derive the equation of state for the moment, which is conveniently expressed as $\theta(h_z,\cos\theta_0)$. Let us skip simple calculations and show the final expression:

$$\mathbf{m}(\mathbf{h},\mathbf{n})=\hat{\mathbf{h}}\cos\theta(h,\hat{\mathbf{h}}\mathbf{n})+\frac{\mathbf{n}-\hat{\mathbf{h}}(\hat{\mathbf{h}}\mathbf{n})}{|[\hat{\mathbf{h}}\mathbf{n}]|}\sin\theta(h,\hat{\mathbf{h}}\mathbf{n}), \quad (8)$$

from which it is easy to obtain the projection of the particle moment on the z axis of the initial coordinate system (the caret here and in ensuing expressions will mean a vector renormalized to unit length). Equation (8) solves, in principle, the problem of possible allowed states of an arbitrarily oriented particle in an arbitrary magnetic field **h**.

The existence of a region where the dependence $\theta(h,\cos\theta_0)$ does not have a unique solution leads to a similar non uniqueness in the expression (8). In order to construct the remagnetization curve for an ensemble of particles this expression must be complemented (for those **h** for which it is necessary) by a prescription for choosing one of the two values of m_z. The universal rule is that as the applied magnetic field changes continuously, the moment **m** also changes continuously so long as the stability condition (3) is not violated. Whenever this condition is violated, the moment discontinuously assumes a new value, which is, in turn, stable for the given field value. In the future we will refer to these transitions as to moment flips. Therefore, the value m_z for a given particle depends on the sequence of change of the applied magnetic field under the specific remagnetization conditions.

2. NONINTERACTING PARTICLES

In our study of remagnetization curves we will assume, as always, that the applied external magnetic field **H** is uniform and is always parallel to a certain direction, which we choose as the z axis in our coordinate system. Then components H_x and H_y are zero and H_z changes from $-\infty$ to $+\infty$ and back. The magnetization **M** of an ensemble depends not only on the value of the magnetic field, but on the direction of its change as well, and can be expressed in terms of the moment **m** averaged over all particles. It is convenient to relate the magnetization not to a unit volume of a magnetic material, as usual, but only to one particle

$$\mathbf{M}(H_z)=\langle\mathbf{m}(H_z)\rangle. \quad (9)$$

Because of the symmetry of the problem, the vector **M** will always be directed along the z axis and we will omit the magnetization-component subscript. We will understand that $M_0(H_z)$ and $M(H_z)$ are z-components of the magnetization of an ideal (noninteracting particles) and a real magnetic material, respectively. In accordance with the definition (9) the limiting values of $M_0(H_z)$ and $M(H_z)$ as $H_z\to\pm\infty$ are ± 1.

It is not necessary to use the general expression (8) when an ideal magnetic material is studied. In the absence of interaction each particle is acted upon only by the external magnetic field directed along the z-axis and therefore the initial coordinate system coincides (up to the rotation around the z-axis) with the one used in the derivation of the equation of state of the moment. The magnetization $M_0(H_z)$ can be obtained by direct averaging of $\cos\theta$ over the directions of the particle axes:

$$M_0(h_z) = \langle \cos(\theta(H_z, \cos\theta_0))\rangle. \qquad (10)$$

If the external field changes monotonically strictly along the z-axis, we choose one of the two values of the projection of the moment (8) according to following simple recipe: the smaller value of $m_z = \cos\theta$ is chosen for H_z increasing from $-\infty$ and the larger for decreasing from $+\infty$.

As H_z increases all moments are at first antiparallel to the z-axis. Flipping takes place for positive values of the critical field $H_z = H_0(\theta_0)$, which depend on the orientation of the easy axis. For H_z decreasing from $+\infty$, flipping takes place for negative values $H_z = -H_0(\theta_0)$. From now on we always mean that the field increase from $-\infty$, so that moment flips occur in the region of positive H_z only.

For an ensemble oriented along the z-axis the equilibrium equation has a trivial form. All particles flip simultaneously at $H_z = 1$, changing the projection of the moment m_z from -1 to $+1$. Therefore the hysteresis loop shown in Fig. 1 has a rectangular shape.

For a random distribution of the axes the particle flipping occurs consecutively, beginning from the field value $H_z = 1/2$, at which only particles that form an angle θ_0 close to $\pi/4$ flip. Near the minima the dependence $H_0(\theta_0)$ has the quadratic form (6) and thus the interval of the angles θ_0 to which the axes of flipped particles belong, for H_z close to 1/2, increases as a square root. The particles with axes parallel and perpendicular to the external field flip last (at $H_z \approx 1$).

When $\theta(H_z, \cos\theta_0)$ is the solution of Eq. (2) with allowance for particle flips (i.e., the correct branch of θ is picked), the averaging (10) over the orientations of easy axes gives the $M_0(H_z)$, dependence shown in Fig. 1. Let us discuss the form of this dependence for various limiting cases that allow analysis.

For weak fields, $H_z \ll 1$, the angle between the moment of each particle and its easy axis is small: $|\theta - \theta_0| \ll 1$. According to Eq. (2), accurate to third order in H_z,

$$\theta_0 - \theta \approx H_z \sin\theta_0 (1 - H_z \sin\theta_0), \qquad (11)$$

which after substitution into (8) gives

$$M_0(H_z) = -{}^1/_2 + {}^2/_3 H_z + {}^3/_8 H_z^2, \quad |H_z| \ll 1, \qquad (12a)$$

i.e., for an ideal ensemble the remanent magnetization $j_R^0 = M_0(0) = 1/2$, and the static susceptibility $\chi_0(H_z) = \partial M_0(H_z)/\partial H_z$ has the initial value $\chi_0(0) = 2/3$.

In strong fields, $H_z \gg 1$, the angle θ is small. Expanding (2) in powers of H_z^{-1} we get, to first order, $\theta \approx \sin(2\theta_0)/2H_z$. The limiting case of $-H_z \gg 1$ is treated similarly and as a result we have

$$M_0(H_z) = \mathrm{sign}(H_z)(1 - {}^1/_{15} H_z^{-2}), \quad |H_z| \gg 1. \qquad (12b)$$

The points $H_z = 1/2$ and $H_z = 1$ in which flipping begins and ends are of particular interest. It was noticed in many numerical calculations (see Refs. 2, 6, and 7) that susceptibility $\chi_0(H_z)$ for ($H_z = 1/2$ is very large, but the values quoted for $\chi_0(1/2)$ varied greatly. Actually, the susceptibility becomes infinite to the right of $H_z = 1/2$ because, according to (12), the fraction of flipped particles has a

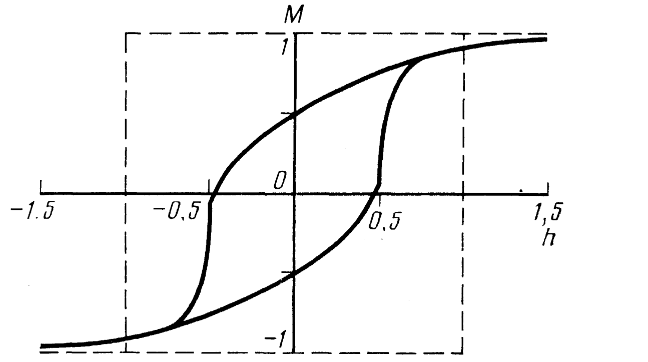

FIG. 1. Remagnetization curve for an ensemble of noninteracting particles with a random (solid line) and collinear with the external field (dashed line) easy-axis orientation.

square-root increase after passage through this point:

$$M_0(H_z) \approx C + {}^3/_2 (2H_z - 1)^{1/2}, \quad \chi_0(H_z) \approx {}^3/_2 (2H_z - 1)^{-1/2}. \quad (13)$$

The singularity turns out to be weaker to the left of $H_z = 1/2$ and the susceptibility goes to infinity logarithmically

$$M_0(H_z) \approx C - 2^{-1/2}(1 - 2H_z)\,|\ln(1 - 2H_z)|,$$

$$\chi_0 \approx \frac{1}{\sqrt{2}} |\ln(1 - 2H_z)|. \qquad (14)$$

The constant C in (13) and (14) is equal to $C = (3^{5/2} + 5 - 7\times 2^{3/2})/15 \approx 0.053$. To the left of $H_z = 1$, where flipping of the moments ceases, the calculation of the fraction of nonflipped particles according to (7) gives an extremely weak singularity which corresponds to a discontinuity in the third derivative of $M_0(H_z)$.

3. THE LOCAL-FIELD APPROXIMATION

Allowance for the interaction of the moments to lowest order in particle concentration in magnetic materials means that each moment experiences the combination of the external field and the field produced by all other moments. The self-action of the moment through its influence on the directions of the neighboring moments is an effect of the second order in concentration. Therefore the field applied to a particle by all other particles can be viewed as a random field not correlated with the spatial position of the particle and with the orientation of its moment. Knowledge of the density distribution of this field $F(\mathbf{h})$ allows us to write the magnetization of a weakly nonideal ensemble as

$$M(H_z) = \int M_0(\mathbf{h}) F(\mathbf{h} - \mathbf{H})\, d^3h, \qquad (15)$$

where $M_0(\mathbf{h})$ is the magnetization of the ideal ensemble.

This approach, which is usually called the local-field approximation, was applied earlier to an equilibrium thermodynamic situation.[3] We consider the problem of the remagnetization of a system of dipoles, and the main difference of this problem from earlier studies is nonuniqueness of magnetization as a function of field. When a certain branch of $M(H_z)$ is calculated in the integral (15), the corresponding branch of the ideal hysteresis loop $M_0(H_z)$ must be used in its three-dimensional form (i.e., valid for any $\mathbf{h}$ direction)

$$M_0(\mathbf{h}) = \langle m_z(\mathbf{h}, \mathbf{n})\rangle. \qquad (16)$$

This form is more universal than (10).

It may seem that for using the local field approximation it is sufficient to consider a sufficiently diluted magnetic material. But the question is complicated by the magnetization hysteresis. The neighboring-particles field acting on a certain particle during the remagnetization, changes in steps that correspond to the field produced by these particles. Because of that the local magnetic field at a particular particle has, in general, a complicated trajectory of motion toward its instant value **h**. This trajectory can sometimes leave the region of stability of the considered branch even if **h** itself is still within this region (we will call these return trajectories). The existence of return trajectories must lead ultimately to earlier moment flips, i.e., to the narrowing of the real hysteresis loop. A rigorous analysis of this situation would have demanded the consideration of the dependence of the magnetization on the trajectory of the local field and introduction of a probability distribution of these trajectories. The problem would become so complex that the very concept of a local field would be meaningless.

The hysteresis loop refinements are determined mainly by a small number of closely located particles rather than by a large number of particles far away, and therefore the problem is not eliminated by the abundant number of particles in an ensemble. In order to be able to neglect the influence of the return trajectories the interaction field between particles that are separated from each other by a minimum distance $2a$ must be small, i.e., the anisotropy constant must be large: $\beta \gg 1$. In this case even the flip of a closely located particle will cause a relatively minor jump of the local field. In the case of a random distribution of the axes this jump will not usually exceed the critical field. As a consequence, the probability for return trajectories turns out to be relatively small and all particle flips can be considered independent. Let us emphasize that this conclusion is directly related to the random-axes distribution and the situation is much more complicated for an ensemble with parallel axes.

The recipe for choosing one of the two values of the function $\theta(h,\cos\theta_0)$ in the expression (8) remains practically the same as for an ideal magnetic material (see Sec 2): for H_z increasing from $-\infty$ the value of θ that gives the larger m_z in the integral (16) is chosen.

4. THE LOCAL-FIELD DENSITY DISTRIBUTION

When one calculates the random field distribution density it is convenient to replace the averaging over all particles by the field averaging over the spatial positions and the moment directions of all particles except the test particle at the origin. Taking into consideration the dilute character of the magnetic material we will assume that particles are distributed in space randomly and neglect the correlation associated with the finite particle size. We must realize, however, that the moments interacting with the test particle cannot be at a distance less than the particle diameter $2a$ from its center. We will denote the average over the direction of the momentum **m** by the angle brackets ($\langle \ldots \rangle$). In the local-field approximation the probability density of the direction of the moment **m** must correspond to that of an ideal ensemble at a certain value of the external field H.

First, let us calculate the average value and the variance, i.e., the mean-square deviation of the random field. The fields created by different particles are independent, and a particle with a moment **m** located at a point **r** creates at the origin a field

$$\mathbf{h}(\mathbf{m},\mathbf{r}) = \frac{V_0}{\beta}\tilde{\mathbf{h}}(\mathbf{m},\hat{\mathbf{r}})/r^3, \quad \tilde{\mathbf{h}}(\mathbf{m},\hat{\mathbf{r}}) = 3\hat{\mathbf{r}}(\mathbf{m}\hat{\mathbf{r}}) - \mathbf{m}. \tag{17}$$

Averaging over the particle positions in the sample and over the direction of its moments **m** and then multiplying by the total number of particles in the system we have

$$\langle h_\alpha \rangle = \frac{nV_0}{\beta}\left\langle \int \tilde{h}_\alpha(\hat{\mathbf{r}}) r^{-3} d^3r \right\rangle = -\eta \frac{J_{\alpha\beta}}{\beta}\langle m_\beta \rangle, \tag{18a}$$

$$\langle h_\alpha h_\beta \rangle = \frac{nV_0^2}{\beta^2}\left\langle \int_{2a}^{\infty} \tilde{h}_\alpha(\hat{\mathbf{r}})\tilde{h}_\beta(\hat{\mathbf{r}}) r^{-6} d^3r \right\rangle$$

$$= \frac{2\pi^2\eta}{15\beta^2}\left[\delta_{\alpha\beta} + \frac{1}{3}\langle m_\alpha m_\beta \rangle\right]. \tag{18b}$$

Here and below η stands for the volume fraction of the ferromagnetic substance, $\eta \equiv nV_0$. The integral (18a) is taken over all scales, from the particle size to the sample boundaries, and constitutes the demagnetization field, while $J_{\alpha\beta}$ is the tensor of the demagnetization coefficients.[9] In the integral (18b), unlike in (18a), only the small distances are essential, infinity is the upper limit, and the particle size is the lower.

Many authors[10,11] deduce a Gaussian distribution density of the local field from the central limit theorem. Should these speculations be correct, the calculated parameters (18) would have fully described the distribution. In fact the above-mentioned theorem claims that any random quantity which is the sum of a large number N of other random quantities which all have identical distributions which are independent of N has in the limit as $N \to \infty$ a Gaussian distribution with a variance equal to the sum of the variances of all parts. In our case N can be increased only by increasing the system volume and the variance of the field distribution created by one particle depends (inversely) on the system volume. Therefore there are no real grounds to believe that the local field distribution is Gaussian. This effect is physically due to the steep decrease of the dipole field with distance.

We will use the Holtsmark technique [12] for a rigorous calculation of the distribution density of the local field. The field distribution created at the origin by N particles distributed randomly over the volume V of the sample can be expressed as

$$F(\mathbf{h}) = \frac{1}{V^N}\int \delta\left(\mathbf{h} - \sum_{i=1}^{N} \mathbf{h}(\mathbf{m}_i, \mathbf{r}_i)\right)\prod_{i=1}^{N} d^3r_i \,.$$

Here averaging over all moments $\mathbf{m}_i$ is assumed. Using the Fourier transform

$$F_\mathbf{k} \equiv \int F(\mathbf{h}) e^{-i\mathbf{k}\mathbf{h}} d^3h = I^N(\mathbf{k}) \tag{19}$$

the distribution density is expressed through the integral for one particle:

$$I(\mathbf{k}) = \frac{1}{V}\left\langle \int \exp(-i\mathbf{k}\mathbf{h}(\mathbf{m},\mathbf{r}))\, d^3r \right\rangle.$$

For large systems this expression is close to unity:

$$I(\mathbf{k})=1-\frac{1}{V}\left\langle \int [1-\exp(-i\mathbf{k}\mathbf{h}(\mathbf{m},\mathbf{r}))]d^3r \right\rangle. \quad (20)$$

For not very small $k \gg a^3$ the integral transforms into

$$\int d\Omega \int_{2a}^{R} r^2\,dr\left\{1-\exp\left(-i\frac{V_0\mathbf{k}\tilde{\mathbf{h}}(\mathbf{m},\hat{\mathbf{r}})}{\beta r^3}\right)\right\}$$

$$=\frac{1}{3}\int d\Omega \int_{R^{-3}}^{(8a^3)^{-1}} [1-\exp(-iV_0\beta^{-1}\mathbf{k}\tilde{\mathbf{h}}(\mathbf{m},\hat{\mathbf{r}})\xi)]\xi^{-2}\,d\xi$$

$$\approx\frac{\pi V_0}{6\beta}\int [|\mathbf{k}\tilde{\mathbf{h}}(\mathbf{m},\hat{\mathbf{r}})|-i\mathbf{k}\tilde{\mathbf{h}}(\mathbf{m},\hat{\mathbf{r}})\ln|\mathbf{k}\tilde{\mathbf{h}}(\mathbf{m},\hat{\mathbf{r}})|]d\Omega \quad (21)$$

$$\equiv V_0\beta^{-1}|\mathbf{k}|A(\hat{\mathbf{k}}\mathbf{m}).$$

Here the sample is approximated by a sphere of an infinitely large radius R. In the general case the integration at large distances adds in (21) a specific imaginary term $-ik_\alpha J_{\alpha\beta} m_\beta$ which is related to the demagnetization and depends on the sample shape only.

Substituting (20) and (21) into (19) we have, in the limit $V\to\infty$, for a fixed particle concentration $N/V=n$

$$F_\mathbf{k}=\exp\left\{-\frac{\eta}{\beta}[\langle A(\hat{\mathbf{k}}\mathbf{m})\rangle|\mathbf{k}|-ik_\alpha J_{\alpha\beta}\langle m_\beta\rangle]\right\}. \quad (22)$$

The unknown distribution density $F(\mathbf{h})$ can be expressed as the inverse Fourier transform of (22)

$$F(\mathbf{h})=\int \exp\left\{i\mathbf{k}\mathbf{h}-i\frac{\eta}{\beta}[k_\alpha J_{\alpha\beta}M_\beta-|\mathbf{k}|\langle A''\rangle]-\frac{\eta}{\beta}|\mathbf{k}|\langle A'\rangle\right\}\frac{d^3k}{(2\pi)^3}, \quad (23)$$

where A' and A'' denote real and imaginary parts of A. The distribution of the projections of the random field $\mathbf{h}$ on an arbitrary direction $\hat{\mathbf{h}}$ can be derived from the three-dimensional distribution (23). It turns out to be a displaced Lorentzian distribution:

$$F(\hat{\mathbf{h}}\mathbf{h})=\frac{1}{\pi}\frac{(\beta/\eta)\langle A'(\hat{\mathbf{h}}\mathbf{m})\rangle}{[\langle A'(\hat{\mathbf{h}}\mathbf{m})\rangle]^2+[(\beta/\eta)\hat{\mathbf{h}}\mathbf{h}-\hat{h}_\alpha J_{\alpha\beta}M_\beta-\langle A''(\hat{\mathbf{h}}\mathbf{m})\rangle]^2}. \quad (24)$$

Let us note that the displacement is due not only to the demagnetization but also to the existence of the additional imaginary term $-i\eta\beta^{-1}\langle A''(\hat{\mathbf{k}}\cdot\mathbf{m})\rangle$ in the exponent of (22).

By virtue of the approximations used in transforming (21), the expression (23) is valid for fields much smaller than the maximum possible field of interaction between the particles $h\ll\beta^{-1}$. It is not necessary to calculate the integral (20) exactly for calculation of $F(\mathbf{h})$ in the $h\sim\beta^{-1}$ range. The distribution for a dilute ferromagnet in this range is determined by only the one particle closest to the test particle. As a result, $F(\mathbf{h})$ can be calculated directly:

$$F(\mathbf{h})=n\left\langle \int \delta[\mathbf{h}-\mathbf{h}(\mathbf{m},\mathbf{r})]d^3r \right\rangle. \quad (25)$$

The practical use of (23) and (25), which describe the local field distribution in the whole actual range $h\ll\beta^{-1}$, is difficult as the integrals in these expressions can not be calculated explicitly. The numerical analysis, however, shows that for any distribution of moment directions the function $F(\mathbf{h})$ is almost isotropic. This is revealed by the behavior of the function $A(\hat{\mathbf{k}}\mathbf{m})$: the real part is even and is located between $A'(0)=4\pi/3\approx4.19$ and $A'(1)=8\pi^2\times3^{-5/2}\approx5.06$, the imaginary part is odd, its absolute value is less or equal to

$$A''(1)=4\pi^2\{1-3^{-1/2}\ln[(3^{1/2}+1)/(3^{1/2}-1)]\}/9\approx1.05.$$

Thus the isotropic distribution density $F_0(\mathbf{h})$, obtained for a case of uniform momentum distribution over a sphere (or a hemisphere), is thus a good quantitative approximation. Corresponding to this distribution is

$$A(\hat{\mathbf{k}}\mathbf{m})\equiv A=\pi^2[2\cdot3^{1/2}+\ln(2+3^{1/2})]/6\cdot3^{1/2}\approx4.54.$$

The three-dimensional distribution that follows from (23) and (25) in the range $h\ll1/\beta$ is

$$F(\mathbf{h})=\frac{\beta}{\pi^2\eta}\frac{A}{[A^2+(\beta h/\eta)^2]^2}, \quad (26a)$$

and in the range $h\approx\beta^{-1}$

$$F(h)=(\eta/2\cdot3^{1/2}\beta h^4)\Phi(6\beta h/\pi), \quad (26b)$$

where

$$\Phi(\xi)=\begin{cases}2\cdot3^{1/2}+\ln(2+3^{1/2}), & \xi<1\\ 2\cdot3^{1/2}-\xi(\xi^2-1)^{1/2}+\ln\dfrac{2+3^{1/2}}{\xi+(\xi^2-1)^{1/2}}, & 1<\xi<2.\\ 0, & \xi>2\end{cases}$$

For $h>\pi/3\beta$ the function $F(\mathbf{h})$ is zero with an accuracy of up to second order in concentration. The average value and the variance of the local field are, according to (18)

$$\langle\mathbf{h}\rangle=0,\quad \langle h_\alpha h_\beta\rangle=\frac{8\pi^2\eta}{45\beta^2}\delta_{\alpha\beta}. \quad (27)$$

We will use this distribution for numerical calculations and assume, in further analysis, that the statistical properties of the local field do not depend on the external field $\mathbf{H}$. We will also ignore the demagnetization. Let us remark that the influence of the demagnetization is missing for a spherical sample ($J_{\alpha\beta}\equiv0$) and a disk with its plane parallel to the z axis ($J_{\alpha z}\equiv0$).

We conclude thus that the local field distribution has a complicated form and actually has a Lorentzian cutoff at large fields. It has three characteristic scales: the field $\tilde{h}_1\approx\eta/\beta$ created by a particle on distances of the average order $n^{-1/3}$, the field of a nearest neighbor $\tilde{h}_3\approx\beta^{-1}$ and the square averaged field $\tilde{h}_2\approx\eta^{1/2}/\beta$.

5. RANDOM ORIENTATION OF THE AXES

In order to calculate the hysteresis loop in the local field approximation for an ensemble of interacting particles one has to substitute the distribution density of the local field (26) and an ideal three-dimensional hysteresis loop (16) into the integral (15). The ideal hysteresis loop (16) is in fact the expression (8) for projections of the moment m_z averaged over the easy axes orientations. For a random axes orientation this leads to

$$M(H_z)=\int_0^{2\pi}\frac{d\varphi_0}{2\pi}\int_0^{\pi/2}\sin\theta_0\,d\theta_0\int m_z(\mathbf{h},\mathbf{n})F(\mathbf{h}-\mathbf{H})\,d^3h. \quad (28)$$

Figure 2 shows hysteresis loops for ensembles with different particle volume fractions η, obtained by numerical integration of (28). Figure 3 gives the concentration dependence of the coercive force for these ensembles.

The Lorentzian character of the distribution $F(\mathbf{h})$ in the range of small values of the random field is especially important near the singularities of the ideal curve $M_0\,(H_z)$ at $H_z = 1/2$ and $H_z = 1$. However, for external fields that are not extremely close to the singularities ($|H_z - 1/2| \gg 1/\beta, |H_z - 1| \gg 1/\beta$), one can use the local character of the distribution density $F(\mathbf{h})$ and consider the quadratic expansion $M_0(\mathbf{H})$ only. In that range the result is determined by the average value of the random field and its variance, which were calculated in the beginning of the previous section. For small H_z this does not help much because the derivatives of the magnetization $M_0(\mathbf{H})$ and the curve $M_0(\mathbf{H})$ itself can be obtained numerically only from the equations derived in Sec. 1. The only exception is the zero value of the external field. For $\mathbf{H} = 0$ in the absence of interaction between particles (see Sec. 2) the moments uniformly fill a hemisphere and give $M_0(0) = 1/2$. The influence of the interaction can be best accounted for by considering separately groups of particles with the same easy-axis orientation. The deflection δ of a particle moment from the easy axis is equal [see (11)] to the component of the local field perpendicular to this axis. The average moment of a particle in each group remains directed along an easy axis in the case of a spherically symmetric random field distribution. The influence of the local field is reduced to the reduction in its value which is now equal to

$$\langle\cos\delta\rangle\approx 1-\frac{\langle\delta^2\rangle}{2}=1-\frac{1}{2}\langle h_\perp^2\rangle=1-\frac{1}{3}\langle h^2\rangle.$$

The remanent magnetization is equal to the half of this moment:

$$j_k=M(0)=\frac{1}{2}-\frac{1}{6}\langle h^2\rangle=\frac{1}{2}-2\pi^2\eta/30\beta^2.$$

Thus the interaction between particles reduces the remanent magnetization.

6. PARALLEL ORIENTATION OF THE AXES

When we considered (Sec. 2) an ideal ensemble with the easy axes of all particles collinear to the external field (z axis) we saw that the particle moments do not deviate from the field direction but rather change sign only during flips (all particles simultaneously) when $H_z = 1$. The influence of the random fields created by other particles leads to deviation of the moments from z and to a certain scatter of the flipping field.

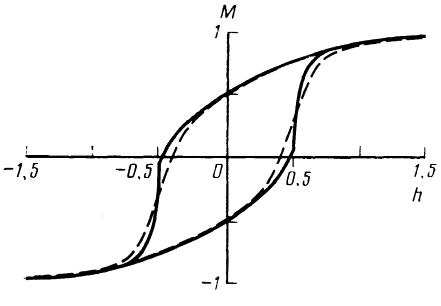

FIG. 2. Remagnetization curves for a randomly oriented ensemble with a volume particle concentration $\eta = 0.2$ with (dashed line) and without (solid line) interaction between particles.

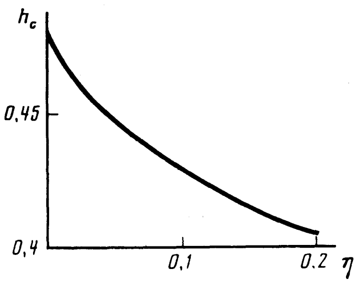

FIG. 3. Dependence of the coercive force for a randomly oriented ensemble on the volume particle concentration.

The random field component which is perpendicular to the axis of the random field is the most important one and that becomes the main criterion for the use of the local field approximation as applied to an oriented ensemble. Far from the flipping range $H_z \approx 1$ the influence of the random field reduces to deflection of the particle moment from the easy axis by an angle (see Sec. 1)

$$\delta=\begin{cases} h_\perp/(1-H_z), & H_z<1\\ h_\perp/(1+H_z), & H_z>1\end{cases}$$

which is in the first approximation proportional to the perpendicular component of the random field. This deflection leads to a decrease, quadratic in $h_\perp$, of the absolute value of the magnetization $M(H_z)$:

$$M(H_z)=\begin{cases} -1+\langle h_\perp^2\rangle/2(1-H_z)^2, & H_z<1\\ 1-\langle h_\perp^2\rangle/2(1+H_z)^2, & H_z>1\end{cases}. \quad (29)$$

In the flip region the displacement of the critical particle field under the influence of the field produced by other particles contributes more to changes of the ensemble magnetization. The critical displacement of the flipping point due to a random field component which is parallel to the z axis is weaker than the deflection of the local field from the particle axis. According to the asymptotic behavior (7), this deflection decreases the value of the particle's critical field by

$$1-H_c(h_\perp)={}^3/_2 h_\perp^{2/3}. \quad (30)$$

The assumption that the jump of the local field caused by the neighboring particle flip is not likely to change the stability range for a given particle is essential for validity of the local field concept (see Sec. 3). This assumption might seem not to hold in an oriented ensemble where all particles flip at almost identical field values. In fact, however, when a neighboring particle flips, the field created by it on a given particle is replaced by a practically opposite field. The change of sign of the random-field component parallel to the axis is negligible in this case, while the absolute value of the perpendicular component (see (29) and (30)) does not change.

Thus the existence of the return trajectories that are unfavorable for the local field approximation can be ignored in the case of the oriented ensemble as well. Figure 4 shows

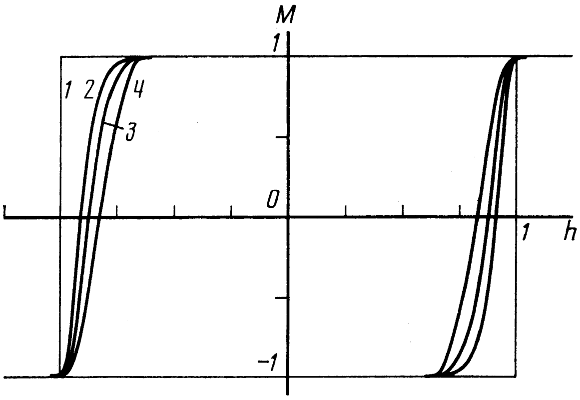

FIG. 4. Remagnetization curves of a collinear ensemble for various values of the volume particle concentration: 1) $\eta \to 0$, 2) $\eta = 0.02$, 3) $\eta = 0.04$, 4) $\eta = 0.08$.

for an oriented ensemble with a different particle concentration the remagnetization curves obtained numerically as the convolution

$$M(H_z) = \int m_z(\mathbf{h}) F(\mathbf{h}-\mathbf{H})\, d^3h$$

of the three dimensional remagnetization curve of an oriented ensemble

$$m_z(\mathbf{h}) = \hat{h}_z \cos\theta(h, \hat{h}_z) + \hat{h}_\perp \sin\theta(h, \hat{h}_z),$$

which is derived from the general expression (8) after recognizing that the director n is parallel to the z axis, with the local field distribution (26).

The analytic form for the asymptotic behavior of $M(H_z)$ outside the flipping region where the magnetization changes are caused mainly by the moment deflections (29) from the z-axis, can be obtained by making use of expansions derived above, which are valid for small random field values which in turn correspond to a large anisotropy $\beta \gg 1$. The behavior of $M(H_z)$ in the range $H_z \simeq 1$ where, on the contrary, only changes of critical fields caused by the random field (30) are important, can be obtained in a similar fashion. In the first case it is necessary to calculate, according to (18), the average square of the perpendicular field component

$$\langle h_\perp^2 \rangle = 4\pi^2\eta/15\beta^2,$$

and in the second case it is necessary to derive from the three-dimensional distribution (23), by means of integration over h_z, the distribution of the random field component that is perpendicular to the axis

$$F_2(\mathbf{h}_\perp) = \frac{\beta}{2\pi\eta} \frac{A'(0)}{[(\beta h_\perp/\eta)^2 + A'(0)^2]^{3/2}} .$$

Here we neglect demagnetization effects. As a result we have

$$M(H_z) = \begin{cases} 1 - \dfrac{2\pi^2\eta}{15\beta^2} \dfrac{1}{(1+H_z)^2}, & H_z > 1 \quad (31b) \\ -1 + \dfrac{2\pi^2\eta}{15\beta^2} \dfrac{1}{(1-H_z)^2}, & 1 - H_z \gg \hbar_3^{2/3}. \\ -1 + 2A'(0) \Big/ \left[\dfrac{8\beta}{27\eta^2} (1-H_z)^3 + A'(0)^2 \right]^{1/2}, & \end{cases}$$

$$0 < (1-H_z) \ll \hbar_3^{2/3} \qquad (31a)$$

These relations allow us to calculate, in particular, the correction to the remanent magnetization (from (31b))

$$j_R = -M(0) = 1 - 2\pi^2\eta/15\beta^2$$

and to the coercive force (from (31b))

$$H_c = 1 - \frac{3^{4/3}}{2} \left(\frac{\eta}{\beta} A'(0) \right)^{2/3} \approx 1 - 5{,}6 \left(\frac{\eta}{\beta} \right)^{2/3} .$$

The remanent magnetization and coercive force decrease under the influence of the moments' interaction, similar to the case of a random-axes distribution.

In conclusion let us remark that the practical uses of oriented ensembles include studies of distributions in the anisotropy constants. In this case the moments' interaction, which, as we have shown, deforms the hysteresis loop significantly (see Fig. 4) even for identical particles, is not taken into account.

[1]E. P. Wohlfarth and E. C. Stoner, Philos. Trans. R. Soc. London. Ser. A **240**, 599 (1948); E. P. Wohlfarth, Research, **8**, S42 (1955); J. Appl. Phys. **29**, 595, (1958).
[2]S. Tikadzumi (Physics of Ferromagnetism. Magnetic Characteristics and Applications (transl. from Japanese), Mir, 1987.
[3]M. W. Klein, Phys. Rev. B **14**, 5018 (1976).
[4]L. Néel, Compt. Rend. **224**, 1550 (1947).
[5]E. I. Kondorskiĭ Dokl. Akad. Nauk SSSR **63**, 507 (1948).
[6]I. S. Jacobs and C. P. Been, Phys. Rev. **100**, 1060 (1955).
[7]I. S. Lyberatos and E. P. Wohlfarth, J. Magn. Magn. Mater. **59**, L1 (1986).
[8]A. R. Caroli and E. P. Wohlfarth, IEEE Trans. 1978, MAG-14, p. 861.
[9]L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media*, Pergamon, 1987.
[10]D. Eldridge, J. Appl. Phys. Suppl. **32**, 247S (1961).
[11]D. Walton and D. Dunlop, Sol. St. Comm. **53**, 359 (1984).
[12]S. Chandrasekhar, Rev. Mod. Phys. **15**, 1 (1943).

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