

On the nature of the magnetization discontinuity in MnBi

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(Submitted December 21, 1972)

Zh. Eksp. Teor. Fiz. 65, 1454-1459 (October 1973)

By studying the rotational-hysteresis curves of MnBi particles it is found that sections with negative magnetic anisotropy exist. A method is found for separating the magnetization discontinuities due to the presence in the crystals of regions of a reduced positive anisotropy constant from regions with negative constant. It is shown that the regions with a negative anisotropy constant play a major role in the formation of the magnetization discontinuity in MnBi particles.

Increasing attention is paid of late to magnetic material with high uniaxial anisotropy. Of greatest interest for the understanding of the process of magnetization reversal is the intermetallic compound MnBi, in the particles of which the coercive force reaches 25-30% of the anisotropy field^[1]. Visual observations have shown that after the magnetization in fields exceeding the saturation field, the magnetization-reversal process in MnBi proceeds in jumps^[2], and the jump can be accompanied by the appearance of the domain structure^[3,4], the form of which differs significantly from the form of the domain structure obtained in the case of magnetization reversal in weaker fields.

No one has yet succeeded in discerning the mechanism of the magnetization jump. Only the following states are known: a initial single-domain state and a final multidomain state if the field of the jump is weaker than the saturation field, or a final single-domain state if the field jump exceeds the saturation field. As to the magnetization jump in MnBi particles, arguments have been advanced in favor of inhomogeneous rotation of the magnetization vector accompanied by the appearance of the domain structure in the entire volume of the crystal^[4], in favor of the onset of an individual magnetization-reversal nucleus^[5,9], and in favor of hysteresis in the growth of the magnetization-reversal nucleus^[6].

It was established recently that hysteresis of the nucleus formation is observed in many uniaxial ferromagnets, and in all materials the magnetization jump occurs not in the anisotropy field, as predicted by the theory, but in a much weaker field. Notice should be taken of the observation of this phenomenon in MnBi^[4], of the detailed investigation of this phenomenon in single crystals with low saturation magnetization^[8], and of the observation of the magnetization jump in bulky single crystals with high saturation magnetization^[9].

To understand the nature of the magnetization jump in real particles, it is necessary to know the character of the disturbances of the crystal lattice (the dislocation structure of the crystal, the presence of inclusions of another phase, etc.). Yet for uniaxial materials these data are either very scanty or entirely nonexistent. In the analysis of the influence of defect on the onset of the domain structure, it is important to distinguish between crystal sections and with decreased anisotropy constant (the easy magnetization axis coincides with the easy magnetization axis of the crystal, but the rotation of the magnetization vector is made easier), and sections with a negative anisotropy constant (the easy magnetization axis is perpendicular to the easy magnetization axis of the crystal or is at least inclined at an angle to it). Their influence on the nucleus-formation process will apparently be different. It can be assumed that the study

of the influence of the rotational field on the field of the magnetization jump in particles, and its comparison with the magnetization-jump field following a change in the direction of the magnetic field, will make it possible to distinguish between crystal sections with modified positive or negative anisotropy constants, and to reveal the role they play in the process of nucleus formation.

To observe the domain structure, the samples were prepared by sintering a pressed mixture of manganese and bismuth powders in a magnetic field^[10]. The sintered samples consisted of individual MnBi particles intruded into a nonferromagnetic matrix (mainly bismuth) and aligned with the hexagonal axis along the magnetic field direction during the sintering. The observations were made on a plane parallel to the hexagonal axis. The dimensions of the particles seen in the plane of the microsection fluctuated in rather wide limits. This is due not only to the difference between the dimensions of the crystals itself, but depends also on the location of the intersection of the particle with the plane of the microsection. The particle has the form of an oblate spheroid, so that the visible dimension of the particle still does not characterize its volume. The magneto-optical Kerr effect was used to observe the domain structure.

To study the rotational hysteresis, the samples were prepared from fine powders obtained by crushing the melt. The particles were aligned with the easy-magnetization axis along the magnetic field and were secured with paraffin, i.e., the samples were textured.

RESULTS OF INVESTIGATION AND DISCUSSION

1. Particle in a Field Parallel to the Easy Magnetization Axis

In the plane parallel to the hexagonal axis, all the particles in the demagnetized state reveal dark and light hairlines corresponding to domains with opposite orientation of the magnetization. Particle magnetization reversal without the magnetic field leading to a noticeable hysteresis of the magnetization-reversal nuclei were observed earlier^[2].

Irreversible annihilation of individual nuclei of magnetization reversal by the magnetic field is shown in Fig. 1. As seen from Fig. 1a, after magnetization by a 12.5 kOe magnetic field there are produced in the particle six magnetization-reversal nuclei. It suffices to increase the magnetic field by only 700 Oe ($H = 13.2$ kOe in Fig. 1b) to keep the four lower nuclei from appearing when the field is decreased. Further increase of the field to 24 kOe (Fig. 1) does not change the number of the produced magnetization-reversal nuclei. This

figure demonstrates clearly that the irreversible annihilation of the magnetization-reversal nuclei in different sections of the crystal is produced in unequal fields. This change of the number of produced magnetization-reversal nuclei as a result of a redistribution of the magnetic poles on the surface of the particle causes, according to our observations, a change in the form of the domain structure in the residual-magnetization state.

Following the irreversible annihilation of all the nuclei by the magnetic field, the magnetization reversal proceeds either with a breakdown into domains, i.e., an incomplete jump if the field of the jump is smaller than the saturation field, or else without a fixed breakdown into domains, i.e., a complete jump if the field of the jump is larger than the saturation field. Figure 2 shows the result of observation of the magnetization reversal on a particle in which, depending on the intensity of the magnetizing field, one observes three visually different magnetization-reversal mechanisms. In the demag-

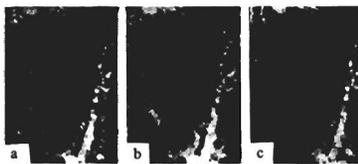


FIG. 1. Irreversible annihilation of nuclei of the reversed phase by the magnetic field. Form of domain structure at 2200 Oe after magnetization in fields of 12.5 (a), 13.2 (b), and 24 kOe (c); magnification 10000X.

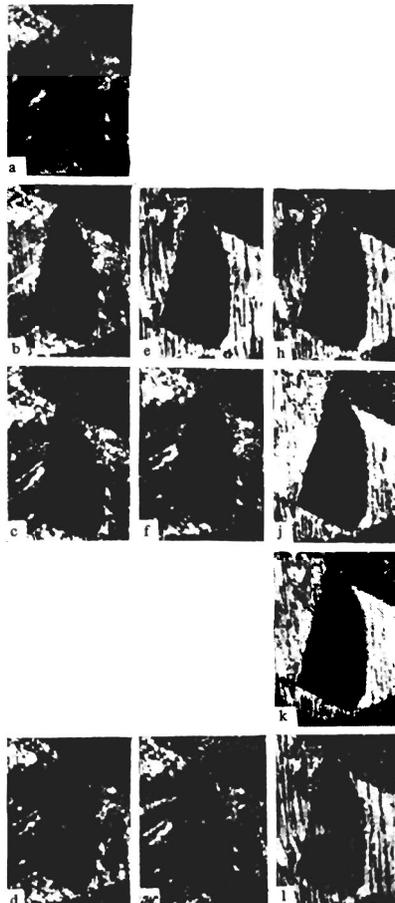


FIG. 2. Visual observation of a particle in which the domain structure is produced in a positive field (first column), in a negative field (second column), or is not produced at all (third column); a—demagnetized state; b, e, h—in magnetizing fields 5,000, 11,000, and 20,000 Oe; c, f, j—in states with residual magnetization; k—in negative field 4500 Oe prior to the magnetization jump, g, h, l—in the coercive force (-300, -1500, and -4800 Oe); magnification 1500X.

netized state (Fig. 2a) the particle is subdivided into the usual plane-parallel domains. After magnetization along the hexagonal axis, the last remainders of the domain structure disappear from the particle in a field of 4600 Oe. At magnetizing field intensities up to 8400 kOe, the magnetization-reversal nuclei are produced in fields 3400–3600 Oe. The domain structures in the residually-magnetized state and in a negative field -300 Oe are shown in Figs. 2c and 2d, respectively. In the magnetizing-field range from 9000 to 12000 Oe, the onset of the domain structure is subject to hysteresis, namely, the domain structure appears jumpwise in a negative field -1500 Oe (Fig. 2g). In the range of magnetizing fields from 14000 to 20000 Oe, the hysteresis is even more pronounced. In this case (third column) it is impossible to observe a domain structure in the magnetization-reversal process. The particle remains magnetized to saturation when the magnetic field is varied from 20000 (Fig. 2h) to -4500 Oe (Fig. 2k). In a field -4800 Oe (Fig. 2l) the particle magnetization is completely reversed in a single jump and the particle is again in the magnetic-saturation state.

The results of similar observations on other particles are shown in Fig. 3, where the abscissas represent the magnetizing field H_m and the ordinates the jump field H_j . The light circles denote incomplete jumps (with breakdown into domains) and the dark circles complete jumps (without breakdown into domains). As seen from Fig. 3, the jumplike appearance of the domain structure can take place in both positive (curve 1) and negative fields (curves 2, 3, 4). It is observed that the jump field in these particles depends on the magnetizing field. In a number of particles, however, a different regularity was observed, namely, the magnetization-jump field did not depend on the magnetizing field in the field interval from 6000 to 24000 Oe. Thus, after an irreversible annihilation of the nuclei by the magnetic field, the particle magnetization is reversed jumpwise, and the number of field jumps can be different for individual particles.

2. Particle in Rotating Magnetic Field

To determine the mechanism of the magnetization jump in uniaxial ferromagnets, it is of interest to study the behavior of the particle in a rotating magnetic field, since the onset of a domain structure is always connected with rotation of the magnetization vector.

The experiments whose results are shown in Fig. 4

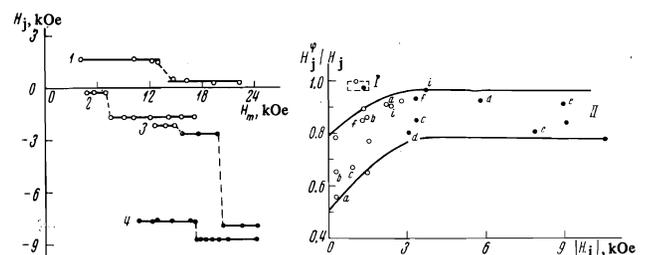


FIG. 3

FIG. 4

FIG. 3. Field in which the domain structure is produced jumpwise (○) or the particle magnetization is completely reversed (●) vs. the magnetizing field for four particles.

FIG. 4. Dependence of the value of H_j^2/H_j on the jump field: ○—incomplete jump, ●—complete jump. Identical letters designate jumps for one and the same particle.

were performed in the following manner: first, the particle was magnetized along the easy-magnetization axis by the field H_m and the jump field H_j was determined (negative); second, the particle was again magnetized by the same field H_m and the positive field was decreased to a value $H < |H_j|$, after which the particle was rotated in the field through 180° . We determined the minimal field H_j^ϕ during the course of rotation in which the particle magnetization was completely reversed or was broken up into domains. The ratio of these two quantities H_j^ϕ/H_j is shown in Fig. 4 as a function of the jump field. As seen from the figure, in some cases (region I) this ratio is equal to unity. In most cases (region II) we have $H_j^\phi/H_j < 1$.

Equality of H_j^ϕ and H_j should be expected in those cases when the principal role is played by hysteresis in the growth of the nucleus of magnetization reversal, or by the occurrence of a magnetization-reversal nucleus on the defect, which can be characterized by a certain effective anisotropy constant smaller than the natural crystallographic anisotropy but of the same sign. A difference between H_j^ϕ and H_j should be expected in those cases when the principal role in the magnetization-reversal process is played by sections with negative anisotropy constant, the presence of which facilitates the rotation of the magnetization vector when the particle is rotated in the magnetic field.

The presence of sections with negative anisotropy in the particles can be revealed by means of the rotational-hysteresis curves^[11]. To verify this assumption, we measured the rotational-hysteresis curves of a textured sample with particle dimension 8μ . The initial state was chosen to be the residually-magnetized state of the sample. The sample coercive force along the texture axis was 9400 Oe. As seen from Fig. 5, in fields much weaker than the coercive force (curve 1 in a field of 1000 Oe) the sample behaves like a permanent magnet, i.e., the single-domain state of the MnBi particles is preserved. In a field close to the coercive force of the sample (curve 2 in a field of 9000 Oe), the dependence of the torque on the field has a complicated character. The most important is the presence of hysteresis at angles close to 180° . This is evidence of the presence of sections with negative anisotropy in the particles. In a field equal to half the anisotropy field, the rotational hysteresis should exist in the angle ranges from 45 to 135° and 225 to 315° . As seen from curve 3, in this sample, in a field of 16 kOe (i.e., $H_A/2$), rotational hysteresis exists in a narrower angle interval, and this can take place when the summary anisotropy constant is decreased by the presence of regions with negative constants.

Thus, the existence of regions with negative anisotropy constants in MnBi particles can be regarded as proved.

The discussion of the nature of these regions encounters great difficulties, since we do not know of any studies of real defects, not only in MnBi but in general in materials having the NiAs structure.

Starting from general considerations of the defects of the crystal structure, we must confine ourselves only to certain general opinions. The vacancies and their clusters will not affect the magnetization-reversal process, not only in the case of domain-wall displacement, but also in the more general case of any rotation

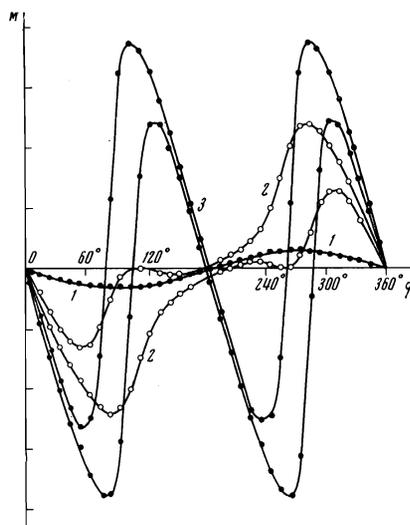


FIG. 5. Rotational hysteresis of textured samples made up of powder with particle dimension 8μ in fields of 1000 (1), 9000 (2), and 16000 Oe (3). Initial state—residually-magnetized, M —a quantity proportional to the torque.

of the magnetization vector. The stresses produced by the dislocations may exert a noticeable influence on the process of magnetization-vector rotation. The magnetostriction of MnBi is large^[12] and as a result the stresses may produce an additional anisotropy proportional to $\lambda_s \sigma$. Since, however, the real dislocation structure of MnBi has not been investigated, further discussion of this question is difficult.

We observed in the MnBi alloy the existence of a metastable phase having the same composition as the main phase, but with different lattice parameters, with a lower Curie point, with a lower saturation magnetization, and with a stronger anisotropy field^[13]. The most likely explanation of the nature of the regions with negative anisotropy constant is the presence inclusions of a metastable phase, with hexagonal axes rotated through a certain angle relative to the main-phase axis.

As seen from Figs. 3 and 4, several jump fields can be observed in one particle. In this case $H_j^\phi/H_j < 1$ for each of them (region II), i.e., the mechanism of the jump is connected with the existence of regions with negative anisotropy. The dependence of the jump field on the value of the magnetizing field is apparently due to the magnetization of the regions with negative anisotropy. It was demonstrated in^[4] that there exists in the particles a mechanism of multidomain-state production, wherein the magnetization jump begins with the rotation of the magnetization vector in the entire volume of the particle. The observation of regions with negative anisotropic constants explains this effect. Indeed, when the magnetic field is decreased, the magnetization vector begins to rotate in the sections with negative anisotropy, and if a coupling with the matrix exists it drags the magnetization vector in the entire volume of the particle.

¹E. V. Shtol'ts and Ya. S. Shur, Dokl. Akad. Nauk SSSR 95, 81 (1954) [Sov. Phys.-Doklady].

²Ya. S. Shur, E. V. Shtol'ts, G. S. Kandaurova, and L. V. Bulatova, Fiz. Met. Metallov. 5, 234 (1957).

³Ya. S. Shur, E. V. Shtol'ts, and V. I. Margolina, Zh. Eksp. Teor. Fiz. 38, 46 (1960) [Sov. Phys.-JETP 11,

- 33 (1960)]. E. V. Shtol'ts, A. A. Glazer, and Ya. S. Shur, *Izv. AN SSSR, ser. fiz.* **25**, 1445 (1961).
- ⁴E. V. Shtol'ts, *Fiz. Tverd. Tela* **7**, 3019 (1965) [*Sov. Phys.-Solid State* **7**, 2445 (1966)]; *Izv. AN SSSR, ser. fiz.* **30**, 1042 (1966).
- ⁵A. Aharoni, *J. Appl. Phys.*, **30**, 785 (1959).
- ⁶Ya. S. Shur and A. A. Glazer, *Fiz. Metal. Metallov*, **29**, 578 (1970).
- ⁷D. Chen, *J. Appl. Phys.*, **37**, 1486 (1966).
- ⁸Ya. S. Shur and V. I. Krabrov, *Zh. Eksp. Teor. Fiz.* **57**, 1899 (1969) [*Sov. Phys.-JETP* **30**, 1027 (1970)].
- ⁹B. Barbara, Ch. Becla, R. Lemaire, D. Paccard, and C. R. Paris, *Ch. Phys.*, **271**, Sec. B, 880 (1970).
- ¹⁰E. V. Shtol'ts and Ya. S. Shur, *Fiz. Metall. Metallov*, **13**, 359 (1962).
- ¹¹M. Prutton, transl. in: *Tonkie ferromagnitnye plenki (Thin Ferromagnetic Films)*, Leningrad, 1967, pp. 117-125.
- ¹²P. A. Albert and W. J. Carr, *J. Appl. Phys.*, **32**, 201 (1961).
- ¹³D. Chen and R. L. Paccard, *J. Appl. Phys.*, **41**, 2530 (1970).

Translated by J. G. Adashko
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