Contribution to the theory of inhomogeneous states of magnets in the region of magnetic-field-induced phase transitions. Mixed state of antiferromagnets

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We demonstate the existence of an extensive group of easy-axis antiferromagnets in which an inhomogeneous state similar to the mixed states of type-II superconductors is realized in a wide range of fields and angles. A phenomenological theory of the mixed state in easy-axis antiferromagnets is developed.

1. It is known that two homogeneous states of essentially different type can be realized in superconductors in the region of a magnetic-field-induced first-order phase transition (FPT) to the normal state. The character of these inhomogeneous states is determined by the sign of the surface tension (σ) on the interface of the normal and superconducting phases.[1-3] In type-I superconductors ($\sigma > 0$) there is produced a thermodynamically stable domain structure made up of domains of the normal and superconducting phases—the intermediate state (IS) of the superconductor.[4] In type-II superconductors ($\sigma < 0$) there is produced the so-called mixed state, which constitutes a system of superconducting (Abrikosov) vortices.[2,3]

It is shown in Refs. 5 and 6, with the spin-flop (SF) transition in easy-axis antiferromagnets as the example, that the same arguments as used in Ref. 4 to corroborate the formation of the IS of a superconductor lead to the following conclusion: a thermodynamically stable domain structure made up of domains of competing phases is produced in the region of spin-reorientation-induced FPT. By analogy with superconductors, this domain structure was named in Ref. 5 an intermediate state of the antiferromagnet. Shortly thereafter the IS of an antiferromagnet was observed in MnF_2 (Refs. 7 and 8). It was proved in Refs. 9 and 10 that the necessary condition for formation of all thermodynamically stable domain structures in magnets is the presence of an FPT induced by an external field in the system. This situation permits the entire manifold of thermodynamically stable domain structures (including the IS of a superconductor) to be treated in the framework of a single theory (see the review[11]).

Since inhomogeneous states ($\sigma > 0$) are not energywise favored, a domain structure in an IS has as a rule a regular character, viz., regions with a homogeneous magnetization distribution (domains) are separated by thin transition layers—domain walls (DW). A substantially different situation is realized in type-II superconductors. Here the inhomogeneous states have lower energy than the homogeneous ones ($\sigma < 0$). The structure realized in the mixed state of a superconductor has therefore a maximally extended inhomogeneity region, or a lattice of Abrikosov vortices, in which the decrease of the vortex sizes is limited only by the magnetic-flux quantization condition.[3]

Can magnets have states similar to the mixed state of a type-II superductor? The only magnet known at present in which there was apparently observed a mixed state is the metamagnet $FeCO_3$ (Ref. 12). As shown there, owing to the competing character of the exchange interaction in the multisublattice metamagnet $FeCO_3$, the energy density of the DW between the anitferromagnetic and paramagnetic phases turns out to be negative in the case of a metamagnetic transition. This produces in $FeCO_3$, in a definite range of magnetic fields, a mixed state that constitutes according to Ref. 12 a triangular lattice of "magnetic vortices." It must be borne in mind that in the case of metamagnetic phase transition that leads to formation of a mixed state in $FeCO_3$ the inequality $\sigma < 0$ is due not to the magnetic symmetry of the crystal, but to the specific character of the exchange interaction, i.e., it is realized for definite relations between the magnitudes of the exchange interactions that form the magnetic structure of the crystal.

We show in the present paper that there exists a large group of easy-axis antiferromagnets in which the possible existence of a mixed state is due to symmetry factors. We develop a phenomenological theory of the mixed state in easy-axis antiferromagnets. We determine on the field–angle phase diagram the boundaries of the existence of a mixed state, and calculate the magnetization and the magnetic susceptibility.

2. The nonequilibrium thermodynamic potential of a two-sublattice antiferromagnet can be expressed, accurate to terms quadratic in the components of the total-magnetization vector $\mathbf{m} = (\mathbf{M}_1 + \mathbf{M}_2)/2M_0$ and the antiferromagnetism vector $\mathbf{l} = (\mathbf{M}_1 - \mathbf{M}_2)/2M_0$ ($\mathbf{M}_i$ is the magnetization of the ith sublattice and $M_0 = |\mathbf{M}_i|$), in the form

$$W = \int dV \left(\frac{1}{2} \alpha_{ik}^{(l)} \frac{\partial l}{\partial x_i} \frac{\partial l}{\partial x_k} + \frac{1}{2} \alpha_{ik}^{(m)} \frac{\partial m}{\partial x_i} \frac{\partial m}{\partial x_k} + w_0 + w' \right), \quad (1)$$

where $\alpha_{ik}^{(l)}$ and $\alpha_{ik}^{(m)}$ are the inhomogeneous-exchange-interaction constants, w_0 is the homogeneous part of the energy, and w' contains terms linear in the first spatial derivatives of m_i and l_i. The possible existence of such invariants in the thermodynamic potential of a magnet was first pointed out in Ref. 13. The actual form of w' is determined by the crystal symmetry. In crystals without inversion center, the nonequilibrium potential contains invariants of the type

$$l_\alpha \frac{\partial l_\beta}{\partial x_\gamma} - l_\beta \frac{\partial l_\alpha}{\partial x_\gamma}. \quad (2)$$

Table I lists certain easy-axis antiferromagnets without inversion centers (the data were taken from Ref. 14). For example, in uniaxial antiferromagnets [crystallographic classes C_n, C_{nv}, D_n, S_4, D_{2d} ($n = 3, 4, 6$)] invariants of type (2) enter in w' in the following form:

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TABLE I.

Antiferromagnet	Symmetry group	T_N^*, K	Antiferromagnet	Symmetry group	T_N^*, K
$RbFeF_4$	C_{2v}^5	–	$CuFeS_2$	D_{2d}^{12}	815
β-$FeNaO_2$	C_{2v}^9	723	$Ba(Fe_xGa_{1-x})_2O_4$	D_6^6	100–600
$BaFe_2O_4$	$C_{2v}^{11(14)}$	880	$FeNb_3S_6$	D_6^6	–
$BaFeF_4$	C_{2v}^{12}	60	$Fe_2Mo_3O_8$	C_{6v}^4	59.5 **
$BaNiF_4$	C_{2v}^{12}	60	$Co_2Mo_3O_8$	C_{6v}^4	40.8 **

*Néel temperatures

**From data of Ref. 29

$$C_{nv}:\ w'=\alpha' w_1;\quad D_n:\ w'=\alpha' w_2;\quad D_{2d}:\ w'=\alpha' w_2';$$
$$C_n:\ w'=\alpha_3' w_1+\alpha_4' w_2;\quad S_4:\ w'=\alpha_1' w_1'+\alpha_2' w_2', \tag{3}$$

where

$$w_1=l_z\frac{\partial l_x}{\partial x}-l_x\frac{\partial l_z}{\partial x}+l_z\frac{\partial l_y}{\partial y}-l_y\frac{\partial l_z}{\partial y},$$
$$w_2=l_z\frac{\partial l_x}{\partial y}-l_x\frac{\partial l_z}{\partial y}-l_z\frac{\partial l_y}{\partial x}+l_y\frac{\partial l_z}{\partial x},$$
$$w_1'=l_z\frac{\partial l_x}{\partial x}-l_x\frac{\partial l_z}{\partial x}-l_z\frac{\partial l_y}{\partial y}+l_y\frac{\partial l_z}{\partial y}, \tag{4}$$
$$w_2'=l_z\frac{\partial l_x}{\partial y}-l_x\frac{\partial l_z}{\partial y}+l_z\frac{\partial l_y}{\partial x}-l_y\frac{\partial l_z}{\partial x}$$

(the symmetry axis is parallel to the z axis). Note that classes D_n and C_n admit of the existence of the invariants

$$l_x\frac{\partial l_y}{\partial z}-l_y\frac{\partial l_x}{\partial z},$$

which can lead to formation of a helicoidal structure in antiferromagnets with $\mathbf{l}\perp z$.

We express the homogeneous part of the nonequilibrium potential for the investigated antiferromagnets in the following standard form[11,15]:

$$w_0=\left[2\lambda\mathbf{m}^2-\frac{2\mathbf{Hm}}{M_0}-B_1l_z^2-B_2l_z^4-(B_1-\beta)m_z^2\right]M_0^2, \tag{5}$$

where λ is the intersublattice exchange-interaction constant, B_1 and β are the second-order anisotropy constants, B_2 is the fourth-order anisotropy constant, and $\mathbf{H}$ is the magnetic field. Usually $\lambda\gg(\beta, B_1)\gg B_2$. In addition, all the calculations in this paper are for low temperatures, when we can put $\mathbf{m}^2+\mathbf{l}^2=1$ and $\mathbf{m}\cdot\mathbf{l}=0$ (Ref. 15).

If $B_1+B_2>0$, the z axis is the easy-magnetization axis. We present now the results, needed later on, of a calculation of the equilibrium states of an antiferromagnet having an energy (5), in a magnetic field inclined to the easy-magnetization axis.[11,15–19] After minimizing with respect to m, we have in the leading approximation in B_1/λ

$$m=\frac{H}{2\lambda M_0}\sin(\theta-\psi) \tag{6}$$

(ψ is the angle between $\mathbf{H}$ and the z axis and θ is the angle between $\mathbf{l}$ and z); the nonequilibrium potential (5) takes the form

$$w_0=H_n^2[{}^1/_2K\sin^2 2\theta-(h^2\cos 2\psi-1)\cos 2\theta-h^2\sin 2\psi\sin 2\theta], \tag{7}$$
$$\mathbf{h}=\mathbf{H}/H_T,\quad K=B_2/B_1+\beta/\lambda,$$
$$H_T=(2\lambda B_1)^{1/2}M_0. \tag{8}$$

First-order phase transitions take place on the H_x–H_z diagram in a narrow vicinity of the point $H_z=H_T$, $H_x=0$. The unique features of the ground state in this region have been investigated in detail in Refs. 15–19. For $K>0$, in particular, a first-order phase transition takes place at the point $H_z=H_T$, $H_x=0$ from the antiferromagnetic (AF) phase ($\theta=0$) to the spin-flop (SF) phase (SF transition).

Let us clarify the role played by invariants of type (2) in the formation of inhomogeneous states of an antiferromagnet in an SF transition. To this end we calculate the energy of a plane domain wall (DW) between the AF and SF phases (90-degree DW). Since we have $m\ll1$ in the field H_T (8), the DW energy is connected mainly with the rotation of $\mathbf{l}$. We assume, to be specific, that $\mathbf{l}$ rotates in the xz plane. For the energy w' in (1) we write

$$w'=-\alpha'\left(l_z\frac{\partial l_x}{\partial\xi}-l_x\frac{\partial l_z}{\partial\xi}\right)=-\alpha'\frac{d\theta}{d\xi}, \tag{9}$$

where ξ is the coordinate in the direction perpendicular to the DW plane. By virtue of the condition $m\ll1$ we assume next that $\mathbf{l}^2=1$. To calculate the energy of a solitary DW in an SF transition we must solve a variational problem for the functional

$$W=\int dV\left[\frac{1}{2}\alpha\left(\frac{d\theta}{d\xi}\right)^2-\frac{1}{2}|K|B_1\sin^2 2\theta-\alpha'\frac{d\theta}{d\xi}\right]M_0^2 \tag{10}$$

with boundary conditions $\theta(\infty)=0$, $\theta(-\infty)=\pi/2$, $d\theta/d\xi(\pm\infty)=0$. The Euler equation for this problem does not contain a term with α'. This enables us to calculate the $\theta(\xi)$ dependence by a standard procedure.[20] For the DW considered there are two directions of rotation of l in the xz plane. Reversal of the rotation direction changes the sign of the integral of the last term in (10), without changing the remaining ones. This is most important: for any sign of α' there is in the DW a rotation direction for $\mathbf{l}$ such that the last term of (10) makes a negative contribution to the energy. Integration yields the following expression for the DW energy density per unit surface:

$$\sigma_{SF}=2(\alpha|K|B_1)^{1/2}-|\alpha'|\pi/2. \tag{11}$$

Invariants of type (2) describe an exchange-relativistic interaction, so that in order of magnitude we have

$|\alpha'| \sim (\lambda B_1)^{1/2} a_0$ (a_0 is the lattice constant) and $\alpha \sim \lambda a_0^2$. Substituting these relations in (11) we see that

$$|\alpha'|\pi/2 \approx (\lambda B_1)^{1/2} a_0 \gg 2(\alpha|K|B_1)^{1/2} \approx (\lambda B_1)^{1/2}|K|^{1/2} a_0,$$

i.e., the energy of a DW in an SF transition is negative. Thus, the presence of invariants of type (2) in the thermodynamic potential leads perforce to formation of inhomogeneous states in the vicinity of the SF transition. By analogy with type-II superconductors, we call this state a *mixed state of a ferromagnet.*

Before dealing with the structure of the mixed state, let us determine the limits of its existence. To this end we consider a plane DW between states with opposite directions of **l** (180-degree DW). Outside a narrow vicinity of the SF transition $\alpha_{ik}^{(l)}$ and $\alpha_{ik}^{(m)}$ the first term of (7) can be neglected; minimization of (7) leads then to the known Néel formula[20]

$$\mathrm{tg}\, 2\theta_0 = \frac{h^2 \sin 2\psi}{1 - h^2 \cos 2\psi}. \qquad (12)$$

Substituting (12) in (1) we get

$$W = \int dV \left[\frac{1}{2} \alpha \left(\frac{d\theta}{d\xi} \right)^2 - B^* \cos^2(\theta - \theta_0) - \alpha' \frac{d\theta}{d\xi} \right], \qquad (13)$$

where

$$B^* = B_1 [(1 - h^2 \cos 2\psi)^2 + (h^2 \sin 2\psi)^2]^{1/2}$$

plays the role of the effective anisotropy. The variational problem $\delta W = 0$ with boundary conditions $\theta(-\infty) = \theta_0$, $\theta(\infty) = \theta_0 + \pi$, $d\theta/d\xi(\pm\infty) = 0$ leads to the first integral of the Eugler equation; this integral coincides functionally with the first integral of the corresponding problem for an easy-axis ferromagnet (see Ref. 21):

$$\frac{1}{2} \alpha \left(\frac{d\theta}{d\xi} \right)^2 = B^* [C - \cos^2(\theta - \theta_0)]. \qquad (14)$$

$C = 1$ for the above boundary conditions. The absence of a term with α' from (14) leads to an important conclusion: interactions described by the invariant (9) do not influence the $\theta(\xi)$ distribution in the DW.

Integration in (13) with respect to (14) leads to

$$\sigma = 2(2\alpha B_1)^{1/2} [(1 - h^2 \cos 2\psi)^2 + (h^2 \sin 2\psi)^2]^{1/4} - |\alpha'|\pi. \qquad (15)$$

Just as in (11), we have chosen here for **l** a rotation direction for which the DW energy is lower, When the SF-transition field is approached, the first term tends to zero and the second remains constant. It follows from (14) that the inequality $\sigma \leqslant 0$ defines on the H_x–h_z phase diagram a mixed-state region bounded by the closed curve (Fig. 1)

$$(1 - h^2 \cos 2\psi)^2 - (h^2 \sin 2\psi)^2 = \nu^2,$$
$$\nu = \left(\frac{\alpha'\pi}{2} \right) \frac{1}{2\alpha B_1} = \frac{h_2^2 - h_1^2}{h_2^2 + h_1^2}, \qquad (16)$$

where h_1 and h_2 are the limits of the region with $\sigma \leqslant 0$ in a field parallel to the z axis:

$$h_{1,2}^2 = 1 \mp \nu. \qquad (17)$$

The maximum angle for which the condition $\sigma \leqslant 0$ is satisfied

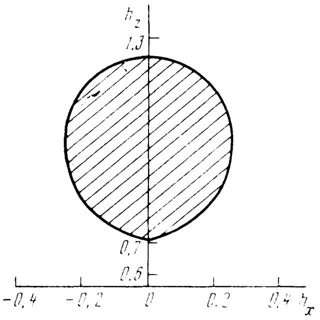

FIG. 1. Phase diagram of easy-axis antiferromagnet with mixed state. The region in which mixed state exists is hatched.

is

$$\psi_{max} = {}^1/_2 \arcsin \nu. \qquad (18)$$

Since $\alpha' \sim (\lambda B_1)^{1/2} a_0$, we have $\nu \sim 1$. This means that the field interval in which $\sigma \leqslant 0$ is comparable with the SF-transition field H_T. Thus, in the antiferromagnets considered there exists on the h_x–h_z phase diagram an extensive region where a mixed state is energywise favored. This region is hatched in Fig. 1 [the boundary (16) was calculated for $\nu = 0.5$; in this case $h_1 = 2^{-1/2}$, $h_2 = (3/2)^{1/2}$, $\Delta h = h_2 - h_1 = 0.517$, $\psi_{\max} = 15°$].

The domain walls in the antiferromagnets considered are due to competition of two opposing tendencies. On the one hand, DW formation increases the system energy, since states with energies higher than the equilibrium value are realized in the DW. On the other hand, interactions describable in the thermodynamic potential by invariants of type (2) tend to produce in the system inhomogeneous distributions of **l** and **m**, therefore their contribution lowers the DW energy. To describe these counteracting factors in the formation of an antiferromagnet DW, we can introduce parameters with the dimension of length. We make the change of variable $\xi = L_1 \tau$, where

$$L_1 = (\alpha/B^*)^{1/2}. \qquad (19)$$

We obtain then for W from (13)

$$W = (\alpha B^*)^{1/2} \int_{-\infty}^{\infty} d\tau \left[\frac{1}{2} \left(\frac{d\theta}{d\tau} \right)^2 + \sin^2(\theta - \theta_0) - \frac{2^{3/2}}{\pi} \frac{L_2}{L_1} \left(\frac{d\theta}{d\tau} \right) \right]. \qquad (20)$$

We have introduced here the parameter

$$L_2 = \alpha'\pi/2^{3/2} B^*, \qquad (21)$$

with the dimension of length. The parameter L_1 [Eq. (19)] has the meaning of the DW thickness. The interaction describable by invariants (2) and (3) lowers the DW energy without changing its structure. The parameter L_2 [Eq. (21)] describes the relative contribution of the interaction described by the invariant (9). It follows from (15) that the inequality $\sigma < 0$ is reached at

$$L_2 > L_1. \qquad (22)$$

The inequality (22) is similar to the known criterion for the production of a mixed state in a superconductor

$$2^{1/2}\lambda > \xi,$$

where ξ is the coherence length and λ is the depth of penetration of the magnetic field.

3. We consider now the structure of the mixed state of an antiferromagnet. In contrast to the IS, where the inhomogeneous states are as a rule concentrated in narrow regions occupying an insignificant part of the magnet volume, in the mixed state the distribution of the magnetization is essentially inhomogeneous in the entire sample. It is clear even from the symmetry of the problem that a magnetization distribution having axial symmetry (single vortex, vortex lattice) can be realized in the mixed state of an antiferromagnet if there is no strong anisotropy in the basal plane. The easy-axis antiferromagnets having invariants of type (2) and satisfying this condition are those with symmetry higher than rhombic, and also rhombic ones with anomalously low anisotropy in the basal plane. It was shown in Ref. 22 that in easy-axis ferromagnets without inversion center, with symmetry higher than rhombic, a system of non-interacting vortices has in a definite field interval a lower energy compared with the homogeneous state and a spiral structure. We emphasize that, in contrast to the vortical states of a magnet (topological solitons), which are intensively investigated in the theory (see, e.g., Refs. 23–25), Ref. 22 deals with stable magnetic vortices. In the region where the mixed state exists, it is natural to expect formulation of one-dimensional inhomogeneous structures in rhombic antiferromagnets with sufficiently strong anisotropy in the basal plane.

In particular, for a one-dimensional structure in a field parallel to the easy-magnetization axis, the distributions of **m** and **l** are determined by solving the variational problem for the potential $W(\theta)$ [Eqs. (10) and (13)], which coincides functionally with that obtained in Ref. 13 in a calculation of a spiral in an easy-plane antiferromagnet. The $\theta(\xi)$ distribution is obtained by integrating (14) and, as is well known,[13] $\theta(\xi)$ is expressed in terms of elliptic functions. Without dwelling on an analysis of the field dependences of $\theta(\xi)$ (this distribution is investigated in detail in Ref. 13, as well as in Izyumov's monograph[27]), we present expressions for the magnetization of an antiferromagnet in a mixed state with a one-dimensional structure:

$$M_x = M_y = 0,$$

$$M_z = \frac{H_T}{2\chi}\begin{cases} [1+\nu k^2E^{-1}(k)]^{1/2}\left[1-\dfrac{1}{k^2}+\dfrac{E(k)}{k^2K(k)}\right], & h_1<h<1,\\[2ex] [1-\nu k^2E^{-1}(k)]^{1/2}\left[\dfrac{1}{k^2}-\dfrac{E(k)}{k^2K(k)}\right], & 1<h<h_2.\end{cases} \quad (23)$$

$$h = \begin{cases} [1+\nu k^2E^{-1}(k)]^{1/2}, & h_1<h<1,\\ [1-\nu k^2E^{-1}(k)]^{1/2}, & 1<h<h_2.\end{cases} \quad (24)$$

Equations (23) and (24) specify the function $M_z(h)$ with k (the elliptic-integral modulus) as a parameter, while $K(k)$ and $E(k)$ are complete elliptic integrals of the first and second kind, respectively.

Figure 2 shows a plot of $M_z(h)$ according to Eqs. (23) and (24) for $\nu = 0.5$ [$M(h)$ is measured in units of the magnetization jump $H_T/2\lambda$ in an SF transition]. The dashed line shows the magnetization curves for an antiferromagnet in which no mixed state is produced. In the SF-transition region the field dependence (23) of M_z is linear:

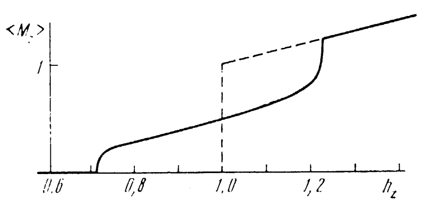

FIG. 2. Magnetization curve of an easy-axis antiferromagnet with mixed state in a magnetic field parallel to the easy-magnetization axis. The dashed line shows the magnetization curve for an antiferromagnet in which no mixed state is produced.

$$M_z = \frac{H_\Pi}{2\lambda}\left[\frac{1}{2}+\left(\frac{1}{2}-\frac{\pi}{16\nu}\right)(h-1)\right], \quad (25)$$

and the static susceptibility is

$$\chi_{zz} = \frac{1}{4\lambda}\left[1-\frac{\pi}{8\nu}\right]. \quad (26)$$

In the vicinity of the limiting values (17) of the fields h_1 and h_2 the variation of $M_z(h)$ is abrupt and $\chi_{zz}(h)$ increases without limit as $h\to h_1$ (h_2).

In the calculations above, **h** has the meaning of the internal magnetic field and differs, by the values of the demagnetizing fields, from the external magnetic field $\mathbf{h}^{(e)}$ that can be monitored and measured. Figure 3 shows the internal magnetic susceptibility $\chi_{zz}^{(i)}$ as a function of the internal field h_z^i (a) and also the field dependence of the magnetic susceptibility in an external field

$$\chi_{zz}^{(e)} = [(\chi_{zz}^{(i)})^{-1}+4\pi N]^{-1},$$

where N is the magnetizing factor along the z axis.

In the region of the limiting fields we have $\chi_{zz}^{(i)}\to\infty$ and $\chi_{zz}^{(e)}$ approaches $1/4\,N$, while $h_1^{(e)}$ and $h_2^{(e)}$ are equal to

$$h_1^{(e)} = h_1, \qquad h_2^{(e)} = h_2\left(1+\frac{2\pi N}{\lambda}\right). \quad (27)$$

The width of the region in which the mixed state exists, in terms of the external-field components, is

$$\Delta h^{(e)} = h_2^{(e)} - h_1^{(e)} = h_2 - h_1 + \frac{2\pi N}{\lambda}h_2. \quad (28)$$

By virtue of the above estimates we have $(h_2-h_1)\approx 1 \gg 2\pi N/\lambda$, i.e., in contrast to IS magnets, a

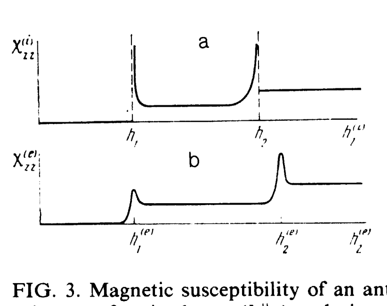

FIG. 3. Magnetic susceptibility of an antiferromagnet in the region of existence of a mixed state ($\mathbf{h}\| z$) vs the internal field (a) and in an external field (b).

mixed state exists in a much wider range of fields and angles, and its width depends little on the sample shape.

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