Thermodynamically stable "vortices" in magnetically ordered crystals. The mixed state of magnets

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It is shown that in magnetically ordered crystals belonging to the crystallographic classes C_n , C_{nv} , D_n , D_{2d} , and S_4 (n = 3, 4, 6), in a certain range of fields, a thermodynamically stable system of magnetic vortices, analogous to the mixed state of superconductors, can be realized.

It is known that in a superconductor two essentially different types of inhomogenous states can be realized in the region of a magnetic-field-induced first-order phase transition to the normal state. In type-I superconductors an intermediate state is realized,¹ while in type-II superconductors a so-called mixed state—a system of Abrikosov vortices²—is realized in a certain range of fields.

In magnets in the region of a first-order phase transition induced by an external field, a thermodynamically stable domain structure consisting of domains of competing phases is formed and is analogous, both in the physical reasons leading to its formation and in its basic properties, to the intermediate state of a superconductor.^{1,3–5} In particular, the domain structure of a ferromagnet is such a structure.

We shall show that in magnetically ordered crystals belonging to the crystallographic classes C_n , C_{nv} , D_n , S_4 , and D_{2d} (n = 3, 4, 6) a thermodynamically stable system of magnetic vortices, analogous to the mixed state of superconductors, can be realized in a certain range of fields.

A standard symmetry analysis shows that for ferromagnets belonging to the above symmetry classes the energy of the system in a magnetic field parallel to the symmetry axis (the z axis) has the form (to within terms quadratic in the components of the magnetization vector \mathbf{M})

$$W = \int \left\{ \frac{1}{2} \alpha \left(\frac{\partial \mathbf{M}}{\partial x_i} \right)^2 - \frac{1}{2} \beta M_z^2 - H M_z - \frac{1}{2} \mathbf{M} \mathbf{H}_{\mathbf{M}} + w' \right\} dV,$$
(1)

where α is the constant of the inhomogeneous exchange interaction, β is the anisotropy constant (for $\beta > 0$ the z axis is the axis of easy magnetization), \mathbf{H}_M is the magnetostatic field, satisfying the equations curl $\mathbf{H}_M = 0$ and div $\mathbf{H}_M = -4\pi \operatorname{div} \mathbf{M}$, and w' is the energy of the inhomogeneous relativistic-exchange interaction. Depending on the symmetry of the system, w' has the form

$$C_{nv}: w' = \alpha' w_{1}, \quad D_{n}: w' = \alpha_{1}' w_{2},$$

$$D_{2d}: w' = \alpha_{2}' w_{2}', \quad C_{n}: w' = \alpha_{3}' w_{1} + \alpha_{4}' w_{2},$$

$$S_{4}: w' = \alpha_{5}' w_{1}' + \alpha_{6}' w_{2}',$$
(2)

where

$$w_{1} = M_{z} \frac{\partial M_{x}}{\partial x} - M_{x} \frac{\partial M_{z}}{\partial x} + M_{z} \frac{\partial M_{y}}{\partial y} - M_{y} \frac{\partial M_{z}}{\partial y},$$

$$w_{2} = M_{z} \frac{\partial M_{x}}{\partial y} - M_{x} \frac{\partial M_{z}}{\partial y} - M_{z} \frac{\partial M_{y}}{\partial x} + M_{y} \frac{\partial M_{z}}{\partial x},$$

$$w_{1}' = M_{z} \frac{\partial M_{z}}{\partial x} - M_{z} \frac{\partial M_{z}}{\partial x} - M_{z} \frac{\partial M_{y}}{\partial y} + M_{y} \frac{\partial M_{z}}{\partial y},$$

$$w_{2}' = M_{z} \frac{\partial M_{x}}{\partial y} - M_{z} \frac{\partial M_{z}}{\partial y} + M_{z} \frac{\partial M_{y}}{\partial x} - M_{y} \frac{\partial M_{z}}{\partial x}.$$
(3)

We note also that the classes D_n and C_n admit the invariants $M_x \partial M_y / \partial z - M_y \partial M_x / \partial z$, which can lead to the formation of a spiral structure with propagation vector along the z axis. However, their role is not discussed in the present paper [we assume that the constants corresponding to them are small in comparision with (2)].

The possibility of the existence of invariants linear in the first spatial derivatives in the expansion of the nonequilibrium thermodynamic potential of a system was first pointed out in Ref. 6 (see also Refs. 7–9).

We shall consider first of all a planar isolated domain wall, separating states with $\mathbf{M}_1 \| z \text{ and } \mathbf{M}_2 = -\mathbf{M}_1 (\mathbf{H} = 0)$. For definiteness, let the system belong to the crystallographic class C_{nv} . Then it is not difficult to show that for any orientation in the xy plane of the normal **n** to the domain wall the energy density of the domain wall will be given by the expression

$$=2M_0^2(\alpha\beta)^{\frac{1}{2}} \tag{4}$$

for a Bloch wall (M rotates in the plane perpendicular to n), and

$$\sigma = 2M_0^2 (\alpha \beta^*)^{\frac{1}{2}} \pm \alpha' \pi M_0^2, \quad \beta^* = \beta + 4\pi$$
(5)

for a Néel wall (*M* rotates in the nz plane). The signs \pm in (5) pertain to the right and left directions of rotation, respectively.

It can be seen from (5) that for

σ

$$\pi |\alpha'| > 2(\alpha \beta^*)^{\frac{\gamma_1}{2}} \tag{6}$$

the formation of domain walls becomes energetically favored, i.e., the uniform ferromagnetic state becomes unstable. The important point is that the cause of the instability is the inhomogeneous-interaction energy (2), (3), and not the magnetostatic energy. By analogy with type-II superconductors, it is natural to assume that a mixed state in the form of a system of magnetic vortices should arise in a ferromagnet when the criterion (6) is fulfilled.

The inequality (6) has a simple physical meaning. The formation of an inhomogeneous state, on the one hand, leads to an increase of the energy associated with the inhomogeneous exchange interaction and the anisotropy, and, on the other hand, lowers the energy of the inhomogeneous relativistic-exchange interaction. The inequality (6) is analogous to the well known relation between the coherence length ξ and penetration depth $\lambda: \xi < 2^{\frac{1}{2}}\lambda$ (Ref. 2), which determines the condition for formation of a mixed state of a superconductor. We have not succeeded in obtaining an exact solution for the mixed state of a magnet, and therefore we shall confine ourselves to the following proof of the possibility of its existence. Let the ferromagnet be situated in an external magnetic field $\mathbf{H} || z$ that is strong enough for the state of the ferromagnet to become uniform. The energy density of the uniform state is equal to

$$w_{0} = -\frac{1}{2} \beta M_{0}^{2} - M_{0} H + 2\pi N_{z} M_{0}^{2}.$$
 (7)

We assume that the ferromagnet has the shape of an ellipsoid of revolution with its principal axis along $z(N_z)$ is the corresponding demagnetization factor). With decrease of the field a nonuniform state should arise. We shall consider at what value of the field the formation of isolated vortices of cylindrical shape becomes energetically favored. We shall study a nonuniform magnetization distribution $\mathbf{M}(\mathbf{r})$ localized at the coordinate origin. For the vector \mathbf{M} we introduce spherical coordinates:

$$\mathbf{M} = M_0(\sin\theta\cos\psi, \ \sin\theta\sin\psi, \ \cos\theta), \tag{8}$$

while for the spatial variables we introduce cylindrical coordinates (ρ , φ , z). The variational problem for the energy (1) admits the solution $\psi = \varphi$, $\theta = \theta(\rho)$.¹⁾ After integrating over φ , we obtain for the energy (1) the following expression:

$$W = 2\pi M_0^2 \int \rho \, d\rho \left\{ \frac{\alpha}{2} \left[\left(\frac{d\theta}{d\rho} \right)^2 + \frac{1}{\rho^2} \sin^2 \theta \right] \right. \\ \left. + \alpha' \left[\frac{d\theta}{d\rho} + \frac{1}{\rho} \sin \theta \cos \theta \right] \right. \\ \left. - \frac{1}{2} \beta^* \cos^2 \theta - h \cos \theta \right\},$$
(9)

where $h = (H - 4\pi N_z M_0)/M_0$, and the internal magneticdipole energy of a vortex is calculated in the Winter approximation.

The function $\theta(\rho)$ minimizing the functional (9) with the boundary conditions $\theta(0) = \pi$ and $\theta(\infty) = 0$ describes the magnetization distribution in an isolated magnetic vortex. We have not succeeded in finding the exact solution for $\theta(\rho)$. Therefore, to calculate the energy of a magnetic vortex we use the trial function

$$\theta(\rho) = \pi (1 - \rho/\rho_0), \quad \rho < \rho_0; \quad \theta(\rho) = 0, \quad \rho > \rho_0.$$
 (10)

Integration of (9) with $\theta(\rho)$ (10) leads to the following expression for the energy of a magnetic vortex:

$$\Phi = W - W_{unit} = \pi M_0^2 [A - B\rho_0 + C\rho_0^2],$$

$$A = 6.1\alpha, \quad B = \pi \alpha', \quad C = (\beta + 2, 38h)/4.$$
(11)

The last term in (11)—the energy associated with the homogeneous part of the exchange energy of the ferromagnet—is proportional to the potential barrier $\Delta \Phi$ separating the equilibrium states. In the expression (11) the characteristic size ρ_0 of the vortex plays the role of the variational parameter. An important point is that the equilibrium values of ρ_0 do not depend on the magnitude of the energy of the inhomogeneous exchange interaction. Substituting into (11) the equilibrium values ρ_0^* :

$$\rho_0^* = \frac{B}{2C} = \frac{2\pi\alpha'}{\beta^* + 2.38h},$$
(12)

we find that the region of thermodynamic stability of the magnetic vortex is specified by the condition $\Phi \leq 0$ and is reached in fields

$$|H| \leq H_{c} = \left[\frac{\pi^{2}}{14.76} \frac{(\alpha')^{2}}{\alpha} - 0.42\beta^{2} + 4\pi N_{z}\right] M_{0}.$$
(13)

It follows from the latter relation that for $\alpha' > 0.8(\alpha\beta^*)^{1/2}$, in a certain range of fields (13), a nonuniform state in the form of a system of magnetic vortices (a mixed state) will be energetically more favored than the uniform state. We note that in the calculation of the energy of an isolated vortex we have used the trial function (10) instead of the true distribution $\theta(\rho)$. Therefore, the criterion given above for the formation of a mixed state of a magnet is an overestimate in comparison with (6). We shall estimate the sizes of the magnetic vortices. It follows from (12) that in the boundary field H_c the equilibrium size of a vortex is equal to

$$\rho_0^* \approx \alpha/\alpha' \approx \alpha/(\alpha\beta)^{\frac{1}{2}} \sim (\alpha/\beta)^{\frac{1}{2}} \sim 10^1 - 10^2 a_0, \qquad (14)$$

where a_0 is the lattice constant.

We note that when the condition $(\alpha')^2/\alpha \gg 4\pi$ is fulfilled the region of existence of the mixed state of magnets will be considerably greater than the region of existence of the intermediate state.

The formation of spiral magnetic order is usually attributed to the presence of invariants of the type (3).^{6–9} Therefore, we shall compare the energies of the mixed state considered by us and a spiral structure.

When the field is lowered below the saturation region a spiral magnetic structure can arise. By analogy with Ref. 9 it is natural to assume that near the nucleation field the spiral structure will be a system of noninteracting planar 360-degree domain walls—a soliton lattice.

Substituting $M_z = M_0 \cos \theta$ and $M_n = M_0 \sin \theta$ into (1), we find that the energy density of a 360-degree domain wall (per unit area perpendicular to the normal **n** to the wall) is equal to

$$\sigma = \int_{-\infty}^{\infty} dx [w(\theta) - w_0] + \int_{-\infty}^{\infty} dx w'(\theta), \qquad (15)$$

where

$$w(\theta) = \frac{1}{2} \alpha M_0^2 \left(\frac{\partial \theta}{\partial x}\right)^2 - \frac{1}{2} \beta M_0^2 \cos^2 \theta - H M_0 \cos \theta, \quad (16)$$

$$w'(\theta) = -\alpha' M_0^2 \frac{d\theta}{dx}, \qquad (17)$$

 w_0 is defined as in (7), and x is the coordinate along **n**.

By making use of the fact that the last term in (15) does not give a contribution to the variational equation $\delta \sigma = 0$ determining the distribution $\theta(x)$, we transform the first term in (15) in the standard way.¹⁰ We have

$$\frac{1}{M_0^2}\sigma = (2\alpha)^{\frac{\gamma_2}{\gamma_0}} \int_0^{2\pi} \left[H(1-\cos\theta) + \frac{1}{2}\beta\sin^2\theta \right]^{\frac{\gamma_0}{\gamma_0}} d\theta - \alpha' \int_0^{2\pi} d\theta.$$
(18)

After the integration we obtain

$$\sigma = 2M_{\mathfrak{o}^{2}}(\alpha\beta^{*})^{\nu_{h}} \left[2(\tilde{\hbar}+1)^{\nu_{h}} + \tilde{\hbar} \ln \frac{(\tilde{\hbar}+1)^{\nu_{h}} + 1}{(\tilde{\hbar}+1)^{\nu_{h}} - 1} \right] - 2\pi M_{\mathfrak{o}^{2}} |\alpha'|.$$
(19)

where $\tilde{h} = h / \beta^*$. It follows from this that the spiral structure becomes energetically favored when

$$H < H_{\bullet} = \left[\frac{\pi^{2}}{16} \frac{(\alpha')^{2}}{\alpha} - 0,33\beta^{*}\right] + 4\pi N_{\bullet} M_{\bullet}.$$
 (20)

This formula was obtained in the limit $(\alpha')^2/\alpha \gg \beta^*$.

From comparison of (20) and (13) it can be seen that when the condition

$$\alpha' > 1.32 (\alpha \beta^*)^{\frac{1}{2}} \tag{21}$$

is fulfilled the mixed state nucleates at higher fields (i.e., earlier) than the spiral state, and this means that there exists a range of external magnetic fields in which the mixed state is energetically the more favored.

The criterion (21) was obtained in a rather crude model calculation of the vortex energy. It is clear that the true criterion for the existence of a mixed state is less stringent than (21). It is entirely probable that when the criterion (6) itself is fulfilled the mixed state will possess the lowest energy. (We note that the criterion $\xi < 2^{1/2} \lambda$ for the existence of a mixed state of superconductors is also derived from consideration of the energy of a planar interphase boundary.)

We have considered vortices in ferromagnets belonging to the crystallographic class C_{nv} . In this case the magnetization distribution (8) with $\psi = \varphi$ corresponded to the extremum of the potential. It can be shown that in the other classes the following distributions are realized:

$$D_{n}: \psi = \varphi - \pi/2; \quad D_{2d}: \psi = -\varphi + \pi/2; \quad C_{n}: \psi = \varphi + \psi_{1};$$

$$S_{4}: \psi = -\varphi + \psi_{2}, \quad (22)$$

with

$$\operatorname{tg} \psi_{1} = -\alpha_{4}'/\alpha_{3}', \ \operatorname{tg} \psi_{2} = -\alpha_{6}'/\alpha_{5}'. \tag{23}$$

We note also that the presence of terms analogous to (3) in the energy of many-sublattice magnets (ferrites, antiferromagnets, etc.) can lead, under certain conditions, to the formation of a mixed state in them. The conditions for the formation of a mixed state become less stringent in the region of spin-reorientation transitions, both spontaneous $(\beta \rightarrow 0)$ and induced (e.g., in antiferromagnets near the spin-flop transition).

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¹⁾ Here and below we assume for definiteness that $\alpha' > 0$.

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