Behavior of magnetic superconductors in a magnetic field

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We investigate the behavior in a magnetic field of magnetic superconductors in which the ferromagnetic and superconducting transition temperatures are close together. It is shown that as the temperature is lowered the order of the superconducting transition changes from second to first. The corresponding critical fields and the field and temperature dependences of the magnetization are determined. Attention is focused on a discontinuity in the magnetization in the vortex core in magnetic superconductors. This feature plus the relatively large scattering cross section make magnetic superconductors convenient objects for the study of the superconducting vortex lattice by neutron diffraction.

1. INTRODUCTION

Since the discovery of the first superconductors with a regular lattice of magnetic atoms, compounds with ferromagnetic ordering at low tempertures have attracted fundamental interest.[1] In the alternative case, where there is antiferromagnetic ordering, the interaction of the superconducting and magnetic subsystems is weak because of the absence of a constant exchange field and of a magnetic field, and these two transitions occur to a large extent independently of one another. A different situation occurs in superconducting ferromagnets: in the superconducting phase at the temperature T_M, which practically coincides with the Curie temperature Θ (as would be the case in the system in the absence of Cooper pairing), a nonuniform magnetic ordering can take place. Actually, this type of magnetic order takes the form of a domain structure with a characteristic wave vector $Q \sim (a\xi_0)^{-1/2}$ (Ref. 2) where a is the magnetic stiffness, which is of the order of the interatomic spacing, $\xi_0 = 0.18\, v_F/T_c$ is the superconducting correlation length, and T_c is the superconducting transition temperature. With further lowering of the temperature there is a first order phase transition from the domain-type superconducting phase into the normal ferromagnetic phase. At the present time two stoichiometric compounds of this type are known, $ErRh_4B_4$ and $HoMo_6S_8$.[1]

The properties of ferromagnetic superconductors show a number of singularities also above the magnetic transition temperature T_M: the critical field of the transition falls abruptly near T_M and the transition in the field is of first order,[3] whereas near the superconducting transition temperature one observes the ordinary behavior characteristic of a type II superconductor (we note that all known magnetic superconductors are type II). A theoretical description of the properties of a ferromagnetic superconductor requires that adequate account be taken of the exchange interaction,[4] and this substantially changes all the results that have been obtained within the framework of a model of an electromagnetic interaction between superconductivity and magnetism.[5]

The appropriate analysis of the critical magnetic fields in superconductors with $T_M \ll T_c$ has been presented in Ref. 6, and we note that in $ErRh_4B_4$ exactly this situation is realized ($T_c \approx 8.7$ K and $T_M \approx 1$ K). As shown in Ref. 6, in a magnetic field the superconducting transition becomes a first order transition near Θ eventhough the Ginsburg-Landau parameter $\varkappa \sim \lambda_L/\xi_0$ is practically unchanged. Moreover, in a magnetic field the existence of the Larkin-Ovchinnikov-Fulde-Ferrell (LOFF) nonuniform superconducting state is possible.[7,8] Because of the lack of detailed information on the properties of the LOFF nonuniform state it is difficult to determine completely the phase diagram of a ferromagnetic superconductor in the (H, T) plane, and in addition the temperature at which the transition changes from second to first order (the tricritical point) is unknown.

For ferromagnetic superconductors for which T_c and T_M are close together ($T_c > T_M$) it is found to be possible to simplify the full description of their specific behavior in a magnetic field and to determine the phase diagram and the tricritical point as well as the dependence of the magnetization on the external field. It is this range of questions that constitutes the subject of this investigation.

There are a large number of compounds for which T_c and T_M are close together; these are the magnetic alloys $(Er_{1-x}Ho_x)Rh_4B_4$ (Ref. 9), $(Ho_{1-x}Lu_x)Rh_4B_4$ (Ref. 10), $Ho(Ir_xRh_{1-x})_4B_4$ (Ref. 11), as well as many others in a specific range of concentration x. In all these alloys in the range of concentration of interest to us there is long range ferromagnetic order with an "easy-axis" type of magnetic anisotropy. The stoichiometric compound $Tm_2Fe_3Si_5$ also satisfies the necessary condition that the superconducting and ferromagnetic transition temperatures be close (Ref. 12) with $T_c \approx 1.7$ K and $T_M \approx 1.1$ K. In the description of the behavior of magnetic superconductors, anisotropy effects prove to be very important. Unfortunately, all the ferromagnetic superconductors that have been obtained up till now, except $ErRh_4B_4$, are polycrystalline, which makes a direct comparison of theoretical predictions and experimental data difficult.

2. FREE-ENERGY FUNCTIONAL OF A FERROMAGNETIC SUPERCONDUCTOR IN A MAGNETIC FIELD

To describe the behavior exhibited in a magnetic field by a ferromagnetic superconductor that has close values of T_c and Θ one can use a Ginzburg-Landau type functional of the superconducting order parameter $\Delta(\mathbf{r})$ and the magnetization $\mathbf{M}(\mathbf{r})$. It should be noted that in superconductors with

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close values of T_c and Θ, the formation of a magnetic domain structure is unfavorable; moreover, scattering of electrons by nonmagnetic impurities also destabilizes the domain phase, and the presence of irregularities in a magnetic sublattice plays a similar role.[13] As a result, at the point $T_M < \Theta$ a first-order transition should take place from the superconducting nonmagnetic phase to the normal ferromagnetic phase. The temperature of the transition is determined by the condition that the energy of superconducting condensation of electrons is equal to that of the ferromagnetic normal state. Experimental data on superconducting ferromagnetic alloys are in agreement with this conclusion. In the case of close T_c and Θ the energy of sueprconducting condensation is small and the first-order transition will take place into a ferromagnetic state with a small value of magnetic moment. As for the validity of an approach based on the Ginzburg-Landau functional, we note that easy-axis magnetic anisotropy and the long-range magnetic interaction in fact lead to a four-dimensional situation for magnetic fluctuations[14] and make it possible to use mean field theory for the description of the magnetic subsystem. This fact has been well confirmed experimentally (see, e.g., Ref. 15). There are no doubts as to the admissibility of expanding the superconducting functional in powers of $\Delta(\mathbf{r})$ near T_c.

Let us write down the free energy functional $\mathscr{F}(\Delta, \mathbf{M}, \mathbf{B})$ of a ferromagnetic superconductor in an external field $\mathbf{H}$ (which we shall assume parallel the easy axis z):

$$\mathscr{F}(\Delta,\mathbf{M},\mathbf{B}) = N(0)\left[-\tau\Delta^2 + \frac{b}{2}\cdot\frac{\Delta^4}{T_c^2} + g\xi_0^2 \times\left|\left(\nabla - \frac{2ie}{c}\mathbf{A}\right)\Delta\right|^2\right] + n\Theta_0\left[s^2\left(\varepsilon + \frac{\Theta_{em}}{\Theta_0}\right) + \frac{d}{2}s^4 + D(s_x^2+s_y^2)\right] - n\mu\,\mathbf{sB} + \frac{B^2}{8\pi} - \frac{\mathbf{HB}}{4\pi} + fN(0)\frac{\Delta^2 s^2 h_0^2}{2T_c^2}, \quad \mathbf{B} = \mathrm{rot}\,\mathbf{A}. \tag{1}$$

Here $N(0)$ is the electron density of states at the Fermi level, m is the density of localized moments, which, in the compounds we are considering, is of the order of the electron density n_e, $\tau = (T_c - T)/T_c$, $\varepsilon = (T - \Theta)/T_c$, $\mathbf{s} = \mathbf{M}/n$ is the magnetization normalized to the maximum value of $n\mu$, $\Theta_{\rm em} = 2\pi n\mu^2$, the value of Θ_0 is of the order of the magnetic energy per localized moment, i.e., of the order $\Theta_{\rm em}$ and Θ, and sh_0 is the exchange field acting on an electron. The constants b, g, and f which determine the superconducting functional are, in the general case, of the order unity (within the framework of the free electron gas model $b\approx 0.1$ and $f\approx 0.11$; for clean superconductors $g\approx 0.51$ and for dirty superconductors $\xi_0 \to 1.7(\xi_0 l)^{1/2}$, where l is the mean free path). In the case $T_c \approx \Theta$, the inverse magnetic scattering time is $\tau_m^{-1} \sim \Theta \sim T_c$, which, generally speaking, leads to a renormalization of the constants b, g, and f (without changing their order of magnitude); the appropriate expressions are given in, e.g., Ref. 16. We note that in writing down the interaction of the exchange field and superconductivity it is understood that the exchange field changes only slightly over a superconducting correlation length ξ_0. It follows from the results obtained below that this assumption is valid over the entire range investigated.

Both mechanisms contribute to the magnetic energy of the system: the electromagnetic mechanism ($\sim \Theta_{\rm em}$ per localized moment) and the exchange mechanism (the order of magnitude of this contribution is $\sim N(0)h_0^2/n = \Theta_{\rm ex}$). The corresponding Curie temperatures of the magnetic superconductors are quite low, and the situation arises where the exchange and electromagnetic mechanisms give the same order of magnitude contribution to the magnetic energy of the system: $\Theta_{\rm ex} \sim \Theta_{\rm em} \sim \Theta_0 \sim \Theta$ (we note that in ordinary ferromagnetic metals, as a rule, $\Theta_{\rm ex} \gg \Theta_{\rm em}$). The coefficient d in the magnetic part of the functional (1) is of the order unity.

As can be seen from (1), the magnetic subsystem acts on the superconducting subsystem via the orbital field $\mathbf{B}$ with vector potential $\mathbf{A}$ and the exchange field sh_0. Expression (1) is written under the assumption that the demagnetizing factor N_z of the sample is small. Below we shall consider just this case, since then all the characteristics of the behavior of magnetic superconductors in a magnetic field are most clearly apparent. It is easy to allow for the demagnetizing factor by adding to the functional (1) the term $N_z \Theta_{\rm em} s^2$ and making the appropriate changes in all the subsequent expressions.

3. CHARACTER OF THE TRANSITION AND CRITICAL MAGNETIC FIELD FOR THE SECOND-ORDER PHASE TRANSITION

To determine the critical magnetic field $H_{c2}(T)$ for the second-order phase transition, it is necessary to eliminate the variables $\mathbf{M}$ and $\mathbf{B}$ from the functional (1); then, in the resulting expression it is sufficient to retain terms up to second order in Δ:

$$\mathscr{F}(\Delta) = N(0)\left\{\Delta^2\left[-\tau + f\left(\frac{\mu H}{2\Theta_0\varepsilon}\right)^2\frac{h_0^2}{2T_c^2}\right] + g\xi_0^2\left|\left(\nabla - \frac{2ie}{c}\mathbf{A}\right)\Delta\right|^2\right\}. \tag{2}$$

As usual (see, e.g., Ref. 17), a determination of the upper critical field reduces to a determination of the smallest eigenvalue of a Schrödinger-type equation which has an oscillatory potential and which is obtained from (2) by a variation with respect to Δ. Thus, we may write immediately:

$$\tau = f\left(\frac{\mu H_{c2}}{\Theta_0\varepsilon}\right)^2\frac{h_0^2}{8T_c^2} + \frac{2eH_{c2}}{c}g\xi_0^2\left(1 + \frac{\Theta_{em}}{\varepsilon\Theta_0}\right). \tag{3}$$

From expression (3) it follows directly that near T_c the orbital effect plays a principal role, and that the dependence $H_{c2}(\tau) = \Phi_0\tau\varepsilon\Theta_{\rm em}/2\pi\xi_0^2 g\Theta_0$ has the usual linear character ($\Phi_0 = c\hbar\pi/e$ is the quantum of flux). The range of applicability of this linear temperature dependence is, however, limited to a narrow neighborhood of T_c: $\tau \ll (\xi_0^2/\Phi_0)^2\Theta_0\Theta_{\rm em}/\mu^2 \sim (\xi_0/\lambda_L^0)^2$, where $\lambda_L^{02} = mc^2/4\pi e^2 n_e$ is the London penetration depth at $T = 0$. Outside the limits of this small region near T_c the exchange effect plays the principal role in determining the critical field, and

$$H_{c2} = \left(\frac{2}{f}\right)^{1/2}\frac{2T_c\Theta_0\varepsilon\tau^{1/2}}{\mu h_0}. \tag{4}$$

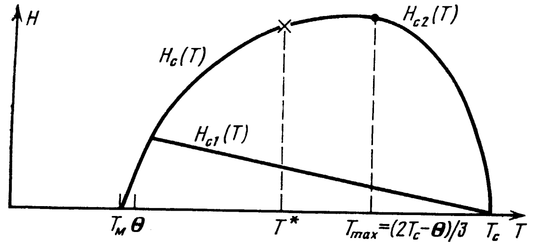

FIG. 1. Schematic temperature dependence of the critical fields of a ferromagnetic superconductor for the case $T_{\max} > T^*$.

The temperature dependence of the critical field is shown in Fig. 1. Over practically the entire range of existence of superconductivity, the transition from the superconducting state to the normal state is governed by the exchange field, and around T_c there should be a new characteristic dependence, $H_{c2} \sim \tau^{1/2}$. We note also that in this case the critical field H_{c3} for the onset of surface superconductivity coincides with H_{c2}.

Expression (4) gives the temperature dependence of the critical field of the second-order transition. An interesting feature of ferromagnetic superconductors is the change of the transition from second to first order at some temperature T^* that lies between Θ and T_c. This temperature corresponds to the vanishing of $\tilde{b}$, the coefficient of Δ^4 in the functional that is obtained from (1) after the elimination of M and B. The essence of the situation is that in the presence of superconductivity the exchange field is weakened, and it is given by the expression

$$h=h_0 s=\frac{h_0 \mu H}{2\Theta_0 \varepsilon+\Theta_{ex} f(\Delta/T_c)^2},$$

in the derivation of which we can neglect screening due to the orbital effect, since, as is well known,[17] this screening only gives a correction $(\xi_0/\lambda_L^0)^2$ to the coefficient b. Let us note that near T^* we assume that $H_{c2} \gg H_{c1}$; the correctness of this inequality will follow directly from the results obtained. Finally, the superconducting functional has the form

$$\mathcal{F}_s(\Delta)=N(0)\left[-\tau\Delta^2+\frac{b}{2}\frac{\Delta^4}{T_c^2}\right]-\frac{H^2}{8\pi}\left[1+\frac{\Theta_{em}}{\Theta_0\varepsilon+\Theta_{ex}f\Delta^2/2T_c^2}\right]. \quad (5)$$

Expanding in terms of Δ^2, we find

$$\tilde{b}=b/2-H^2\Theta_{ex}^2\Theta_{em}f^2/32\pi\Theta_0^3T_c^2\varepsilon^3N(0)$$

and, using (4) we can determine the temperature T^*:

$$T^*=T_c(1+a\Theta/T_c)(1+a)^{-1}=T_c-(T_c-\Theta)a/(1+a),$$
$$a=\Theta_0 b/\Theta_{ex}f\sim 1. \quad (6)$$

From (6) we can conclude that T^* in general lies somewhere halfway between Θ and T_c (see Fig. 1) i.e., there is a first-order transition that takes place in an appreciable region of existence of the superconducting phase. We call attention to the fact that $H_{c2}(T)$ has a maximum at a temperature $T_{\max}=T_c-(T_c-\Theta)/3$, which is determined only by the Curie temperature and T_c and is independent of the coefficients of the functional (1)! If this maximum falls in the region of the second-order transition ($T_{\max} > T^*$), a direct experimental verification of this result is possible.

Before going on to a determination of the first-order transition curve $H_c(T)$, let us clarify the character of the screening of a weak field in ferromagnetic superconductors and determine the vortex penetration field H_{c1}.

4. LOWER CRITICAL FIELD H_{c1}

In determining the field H_{c1} we can assume that the order parameter Δ is independent of the exchange field and is equal to its equilibrium value $\Delta^2=\tau T_c^2/b$. The equation for the vortex field **B**, derived by a variation of (1) with respect to **B** has the form

$$\text{rot}\,\mathbf{B}=\frac{4\pi}{c}\mathbf{j}_s+4\pi\,\text{rot}\,\mathbf{M}. \quad (7)$$

The magnetization is $M=\mu^2 nB/[2\Theta_0\varepsilon+2\Theta_{em}+\Theta_{ex}(\Delta/T_c)^2]$ (**M** and **B** are directed along the z axis). Substituting this expression for **M** into (7) we find that

$$\text{rot}\,\mathbf{B}=\frac{4\pi}{c}\mathbf{j}_s\frac{\Theta_{em}}{\Theta_0(\varepsilon+\Theta_{ex}\Delta^2/2\Theta_0T_c^2)}. \quad (8)$$

As follows from (8), the difference in the screening of the field from that in the case of an ordinary superconductor consists of a renormalization of the London penetration depth

$$\lambda_L^2\to\tilde{\lambda}_L^2=\lambda_L^2(\varepsilon\Theta_0+\tau\Theta_{ex}/2b)/\Theta_{em}, \quad (9)$$

where λ_L is the London penetration depth in the absence of magnetic atoms. Calculating the energy J of a vortex, we obtain (Ref. 6)

$$J=(\Phi_0/4\pi\lambda_L)^2\ln(\tilde{\lambda}/\xi),$$

which differs from the usual expression by the substitution $\lambda_L\to\tilde{\lambda}_L$ only in the argument of the logarithm. Thus, the lower critical field

$$H_{c1}=4\pi J/\Phi_0=(\Phi_0/4\pi\lambda_L^2)\ln(\tilde{\lambda}/\xi)$$

varies weakly in comparison with the corresponding value of H_{c1}^0 in a nonmagnetic superconductor (Fig. 1) and, as follows from (9), the increase of H_{c1} with temperature can be either stronger or weaker than that of $H_{c1}^0(T)$ depending on the parameter $\Theta_{ex}/2\Theta_0 b$ of the system. Let us point out that in the expression for H_{c1} obtained in Ref. 18 λ_L in the argument of the logarithm is differently defined; with the exchange interaction ignored.

It is important to note that the Ginsburg-Landau parameter $\tilde{\varkappa}=\tilde{\lambda}_L/\xi\gg 1$ over the entire range of existence of the superconducting phase. This result again emphasizes the specific property—the exchange mechanism for the onset of a first order transition near the Curie temperature Θ.

5. FIRST-ORDER TRANSITION CRITICAL FIELD H_c

To determine the first-order transition critical field it is necessary to equate the free energy $\mathcal{F}_N$ of the normal state to $\mathcal{F}_s$, that of the superconducting phases. Since $H_{c2}(T^*)=H_c(T^*)\gg H_{c1}$, the transition will occur into the vortex phase with a high vortex density over almost the entire temperature range from T^* to Θ, and this allows us to consider the field B uniform. The orbital effect in the parameter $\varkappa^{-2}$ can be neglected in this case and in fields $H\gg H_{c1}$

the difference in the magnetization in the superconducting phase from that in the normal phase will be determined by the exchange interaction. For the functional $\mathscr{F}_s(\Delta)$ we can use expression (5), which is valid everywhere except for a small region in the neighborhood of Θ, where it is necessary to keep terms proportional to s^4 in the magnetic functional.

The first-order transition field will, therefore, be determined by the following system of equations:

$$-2+x+\frac{H_c^2}{H_{c0}^2}\frac{\Theta_{em}A}{(\Theta_0\varepsilon+Ax)\Theta_0\varepsilon}=0, \tag{10}$$

$$-2+2x+\frac{H_c^2}{H_{c0}^2}\frac{\Theta_{em}A}{(\Theta_0\varepsilon+Ax)^2}=0, \tag{11}$$

$$x=\frac{b}{\tau}\left(\frac{\Delta}{T_c}\right)^2,\quad \frac{H_{c0}^2}{8\pi}=N(0)\frac{T_c^2\tau^2}{2b},\quad A=\frac{f\tau\Theta_{ex}}{2b}.$$

Solving the system of equations (10) and (11) we obtain the transition field H_c:

$$H_c^2=H_{c0}^2(\tau)\frac{\Theta_0\varepsilon}{\Theta_{em}}\left(1+\frac{\Theta_0\varepsilon b}{f\Theta_{ex}\tau}\right)^2. \tag{12}$$

This dependence is shown in Fig. 1. The order parameter Δ_1, that occurs in the transition is given by the expression

$$(\Delta_1/T_c)^2=\tau b^{-1}(1-\Theta_0\varepsilon b/f\tau\Theta_{ex}). \tag{13}$$

Near the Curie temperature, where the transition field is smaller than H_{c1}, the transition takes place directly from the normal ferromagnetic phase to the superconducting Meissner phase. Equating the energies of these phases, we find the transition field

$$-\frac{H_c^2}{8\pi}\left(1+\frac{\Theta_{em}}{\Theta_0\varepsilon}\right)=-\frac{H_{c0}^2}{8\pi},$$

i.e., $H_c\approx H_{c0}(\varepsilon\Theta_0/\Theta_{em})^{1/2}$ and near Θ one should thus observe a square root dependence on the temperature.

In the transition in fields $H\gg H_{c1}$ the field dependence of the magnetization in the compounds we are investigating is unusual for superconductors. In this case a principal role is played by a decrease in the polarizability of the magnetic subsystem with an increase in the superconducting order parameter, and not by the screening of the superconducting current, as is usually the case. The magnetic moment is equal to

$$M=\mu^2 nH/(\Theta_0\varepsilon+\Theta_{ex}\Delta^2/2T_c^2), \tag{14}$$

while the parameter Δ is determined from the equation (11), where the field H takes the place of H_c. The dependence $M(H)$ is shown schematically in Fig. 2. Similar dependences were obtained by numerical methods in Ref. 19.

In the above discussion we have not taken into account terms proportional to s^4 in the magnetic functional. This is valid eveyrwhere except for a small range of temperature $|\varepsilon|\sim\tau(T_c/\varepsilon_F)^{1/2}$ near Θ. It is not difficult to obtain the appropriate expressions for H_c in this region as well, but because the region is so narrow we shall not do so. We only note that the $H_c(T)$ curve terminates at the point T_M corresponding to the first-order transition from the superconducting state to the normal ferromagnetic state in the absence of a magnetic field, this point being determined by the equality of the free energy in these phases:

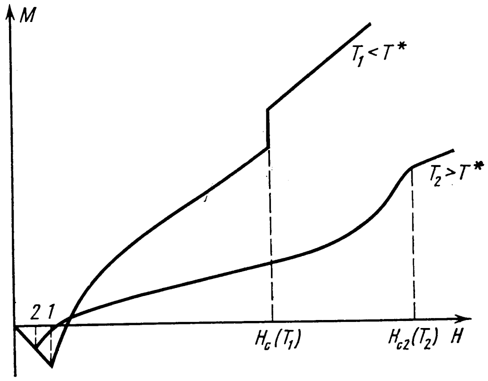

FIG. 2. Field dependence of the magnetization. The points 1 and 2 correspond, respectively, to the fields $H_{c1}(T_1)$ and $H_{c1}(T_2)$.

$$-N(0)T_c^2\tau^2/2b=-n\Theta_0\varepsilon^2/2d;$$

$$T_M=\Theta+(T_c-\Theta)(T_c^2N/(0)d/nb\Theta_0)^{1/4},$$

$$\Theta-T_M\sim(T_c-\Theta)(T_c/\varepsilon_F)^{1/4}.$$

6. CONCLUSION

The behavior of ferromagnetic superconductors in a magnetic field thus differs substantially from the corresponding behavior of ordinary superconductors. For the case where the temperatures T_c and Θ are close together it is possible to obtain a complete description within the framework of a Ginsburg-Landau type functional. The transition changes from second to first order as the temperature is lowered towards Θ, and this effect has nothing to do with a decrease in the Ginsburg-Landau parameter $\varkappa$. Near the temperatures Θ and T_c square root temperature dependences of the critical field should be observed. A particular feature is also the substantially nonlinear field dependence of the magnetic moment in the superconducting phase.

Concerning a comparison of the theory presented here with experiment, we note that detailed measurements of the magnetic properties of ferromagnetic superconductors with close values of T_c and Θ have not yet been made. Existing data on the single crystal $ErRh_4B_4$ agree qualitatively with the description presented here, but because in $ErRh_4B_4$, $\Theta\ll T_c$, it is not appropriate to speak of a more complete comparison with experiment. Furthermore, all ferromagnetic superconductors (with the exception of $ErRh_4B_4$) have so far been obtained only in polycrystalline form. Polycrystalline samples will contain crystallites of various orientations and shapes, i.e., with various demagnetizing factors. As a result each crystallite will have its own transition field. As a consequence, the transition in terms of the resistance of the magnetic superconductors will have a typically percolation character.[20] The lower critical field can also in principle be determined by percolation effects: if the crystallite size is less than λ_L, then for Meissner currents to arise a closed superconducting path must be formed through the crystallites.

In conclusion let us point out that magnetic superconductors may be suitable objects for the study of superconducting vortex lattices by neutron scattering. In ordinary superconductors in the vortex phase, neutron scattering occurs because of the nonuniform distribution of the magnetic field. In magnetic superconductors the appearance of a magnetization $M \sim B$ leads to a substantial contribution from scattering by magnetic atoms, where the scattering cross section is much larger than the cross section for scattering by the field. Let us also call attention to the interesting circumstance that at the core of a vortex sueprconductivity is suppressed; this leads to an increase in the spin susceptibility and, as a result, in the center of the vortex there should be a magnetization discontinuity $\Delta M \sim N$ which is superimposed on the smooth $\mathbf{M}(\mathbf{r})$ dependence in the vortex lattice. The vortex lattice can also point at an angle to the field H if the field is not accurately oriented along the easy axis of the crystal.

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