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# Ferromagnetic film on the surface of a superconductor: possible onset of inhomogeneous magnetic ordering

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A system consisting of a bulky superconductor and a thin ferromagnetic metallic film on its surface is considered. It is shown that under certain conditions the magnetic ordering in the film takes the form of a small-scale domain structure. This ordering corresponds to a minimum system energy if account is taken of the interaction between the magnetic moments and the superconducting electrons that penetrate into the film from the bulky superconductor.

## I. INTRODUCTION

The strong exchange field (exceeding as a rule by several orders the superconducting critical temperature  $T_c$ ) present in a ferromagnet suppresses superconductivity via the paramagnetic effect.<sup>1</sup> For the same reason, the superconducting order parameter is radically decreased near the contact of a superconductor with a ferromagnetic metal. The principal role is played here not by the proximity itself to the normal metal,<sup>2</sup> but precisely by the presence of a strong exchange field in the ferromagnet and by the electron penetration, due to the proximity effect, into this region of the strong exchange field.

The influence of the exchange field on the superconductivity is greatly weakened when a small-scale (compared with the superconducting correlation length  $\xi_0 = 0.18 v_F/T_c$ ) domain structure is produced in the ferromagnet—the exchange field is effectively averaged over Cooper-pair dimensions. Precisely such domain coexistence phases were predicted for ferromagnetic superconductors, and the experimental data agreed with this prediction.<sup>3</sup>

For this reason, inhomogeneous magnetic ordering, rather than ferromagnetism, should likewise occur in a thin ferromagnetic-metal film of thickness  $d$  (on the order of several dozen Angstroms) sputtered on the surface of a superconductor, provided that the Curie temperature  $\Theta < T_c$ . In the case  $Q \gg \xi_0^{-1}$ , where  $Q$  is the characteristic wave vector of such a magnetic structure, the oscillating magnetic field in the film suppresses less the superconductivity at the interface. The gain in the superconducting-condensation energy near the interface compensates here for the magnetic-energy loss due to formation of an inhomogeneous magnetic structure in the film.

A more interesting situation can be expected if  $\Theta > T_c$ . In this case, below the temperature  $T_c$ , in view of the weakness of the superconducting pairing, the film should be in a ferromagnetic phase and suppress strongly the superconductivity in a region of order  $\xi(T)$  near the boundary. With further decrease of the temperature, the superconducting condensation energy increases, as does also the energy loss due to the suppression of the superconductivity near the boundary. It may be expedient as a result to break up the films into minute domains: energy loss due to domain-wall formation is offset by the superconducting-condensation energy gain near the boundary (the domain phase suppresses the superconductivity more weakly). The temperature  $T_{DS}$  at which the film goes over into the domain phase decreases with increase of the film thickness and with decrease of the

superconducting correlation length  $\xi_0$  of the superconductor. The most suitable for observation of the transition are pure superconductors with large  $\xi_0$ , and also superconductors with high condensation energy (high  $T_c$ , high electronic-state density).

It is interesting to note that a suitable object for the study of the transition considered may be vanadium. Onset of surface ferromagnetism in vanadium was observed recently.<sup>4</sup> At low temperatures vanadium becomes superconducting and one can expect a domain magnetic structure rather than ferromagnetism to be produced on the surface.

The considered transition in a ferromagnetic film on the surface of a superconductor can be observed by magneto-optic methods or with the aid of tunnel measurements. The use of a ferromagnetic film as the weak-coupling link of a Josephson structure would make feasible a Josephson junction with variable coupling—the transparency should increase jumpwise when the film goes over into the domain phase.

## 2. BASIC EQUATIONS

We consider the case when a superconductor surface is coated with a thin ferromagnetic-metal film of thickness  $d$  that is small compared with the superconducting correlation length  $\xi(T=0)$ , and assume that the easy axis of the ferromagnet is parallel to the plane of the film (if the easy axis is perpendicular to the film plane a small-scale domain structure exists even in the absence of superconductivity). We assume that the film can have two magnetic states—ferromagnetic without domains (or with domains very large compared with  $\xi_0$ ), and one with small-scale domain structure, see Fig. 1. We designate these states by  $F$  and  $DS$ .

The details of the  $F$ - $DS$  transition in a ferromagnetic film on the surface of a superconductor depend on many parameters of the system: on the homogeneity of the film, on the conditions of the contact on the ferromagnet-superconductor interface, on the density of the electronic states in the film and in the bulk, and others. To illustrate the physics of the phenomenon, we confine ourselves therefore to a patently greatly simplified model. We assume that the dimensionless Cooper-pairing constant  $\lambda$  is the same in the film and in the bulk, and the electron mean free path in the film is  $l \ll \xi_0$ . We confine ourselves also to a situation in which the transition to the  $DS$  phase takes place near the superconducting transition. In the context of the BCS theory, the initial Hamiltonian of the system considered can be written in the form

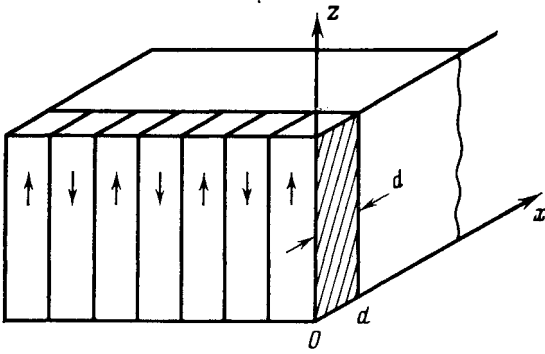


FIG. 1.

$$\mathcal{H} = \int d^3r \left\{ \psi^\dagger(\mathbf{r}) \frac{\hat{\mathbf{p}}^2}{2m} \psi(\mathbf{r}) + \Delta(\mathbf{r}) \psi^\dagger(\mathbf{r}) i\sigma_y \psi(\mathbf{r}) - \Delta^*(\mathbf{r}) \psi(\mathbf{r}) i\sigma_y \psi(\mathbf{r}) + \psi^*(\mathbf{r}) h(\mathbf{r}) \sigma \psi(\mathbf{r}) + |\Delta(\mathbf{r})|^2/\lambda \right\} + \mathcal{H}_{sc}, \quad (1)$$

where  $\Delta(\mathbf{r})$  is the superconducting order parameter,  $\psi(\mathbf{r})$  is a spinor,  $\sigma$  are Pauli matrices,  $h(\mathbf{r}) = h$  is the exchange field acting on the electrons in the film ( $h(\mathbf{r}) = 0$  inside a bulky superconductor), and the term  $\mathcal{H}_{sc}$  takes into account electron scattering by nonmagnetic impurities. The influence of the magnetic induction on the superconductivity is shown in Ref. 3 to be negligibly small under ordinary conditions compared with the exchange-field influence, and will be neglected here.

To describe the behavior of the order parameter in a superconductor, except in region near the film, we can use in lieu of (1) the Ginzburg-Landau function  $\mathcal{F}$  (see, e.g., Ref. 5), supplemented by the boundary condition

$$\frac{\Delta'(x)}{\Delta(x)} \Big|_{x=d} = \gamma. \quad (2)$$

The geometry of the problem is illustrated in Fig. 1, and the functional  $\mathcal{F}$  takes the usual form

$$\mathcal{F} = \int_a^\infty \left\{ \frac{\tau}{\eta} |\Delta|^2 + \frac{1}{4m} \left| \frac{\partial}{\partial x} \Delta \right|^2 + \frac{\beta}{2} |\Delta|^4 \right\} dx, \quad (3)$$

where  $\tau = (T - T_c)/T_c$  and, for example in a pure superconductor,<sup>6</sup>  $\eta = 0.12\varepsilon_F/T_c$ ,  $\beta = 1/\eta n$ , and  $n$  is the electron density.

The solution of the equation that follows from (3) for  $\Delta(x)$  is well known:

$$\Delta(x) = \Delta_0 \operatorname{th}(x/2^{1/2} + C), \quad \Delta_0 = |\tau|/\eta b, \quad (4)$$

where  $\xi^2 = \eta/4m|\tau|$ , and the constant  $C$  is determined by the condition (2), viz,  $\sinh(2C) = 2^{1/2}/\gamma\xi$  (we have taken into account that  $d \ll \xi$ ).

The influence of the magnetic structure of the film on the superconductivity is in fact determined in this case only by the parameter  $\gamma$ . For the ferromagnetic state  $F$  in the film, the superconducting order parameter (under the condition  $hd\xi_0/v_F l \gg 1$ ) is zero on the boundary,  $\Delta(d) = 0$ , i.e.,  $\gamma \rightarrow \infty$ . The solution (4) for the  $F$  phase is

$$\Delta(x) = \Delta_0 \operatorname{th}(x/2^{1/2}\xi).$$

The condensation-energy loss (per unit surface area)

due to the suppression of the superconductivity amounts in this case to<sup>5</sup>

$$\delta_F = n \frac{\tau^2}{\eta} \xi \frac{2^{1/2}}{3}. \quad (5)$$

If the  $DS$  phase is produced, the superconductivity of the interface is not so strongly suppressed (in view of the averaging of the exchange field), and the properties of the  $DS$  phase (period, exchange-field amplitude) are determined just by the parameter  $\gamma$  in (2). The condensation-energy loss is decreased and, as can be easily seen, equals

$$\delta_{DS} = n \frac{\tau^2}{\eta} \frac{2^{1/2}}{3} \xi (2 + \operatorname{th} C) (1 - \operatorname{th} C)^2, \quad (6)$$

where

$$\operatorname{th} C = \operatorname{sh} 2C / (1 + \operatorname{ch} 2C), \quad \operatorname{sh} 2C = 2^{1/2}/\gamma\xi.$$

A contribution to the total energy of a system with a  $DS$  phase is made also by the inhomogeneity of the magnetic structure in the film: the presence of a domain wall entails an energy loss  $E_w = \Theta nd(a/b)$ , where  $b$  is the domain thickness (half the  $DS$ -structure period),  $a$  is the magnetic correlation length and is of the order of the interatomic distance, and  $\Theta$  is of the order of the Curie temperature (the electron and magnetic-atom densities are assumed to be approximately equal). The expression for  $E_w$  indicates that the energy loss per magnetic atom in a domain wall of approximately atomic thickness is  $\sim \Theta$ , i.e., the magnetic anisotropy is regarded as strong—in the case of weak magnetic anisotropy the domain-wall energy is somewhat lower and a generalization to include this case is easy.

The total system energy in the presence of a  $DS$  phase in the film can be written in the form

$$E_{DS} = \Theta nd \left( \frac{a}{b} \right) + n \frac{\tau^2}{\eta} \xi \frac{(1 - \operatorname{th} C)^2 (2 + \operatorname{th} C) \cdot 2^{1/2}}{3} + E_m + E_s, \quad (7)$$

where  $E_m$  is the energy of the ferromagnetic ordering in the film, and  $E_s$  is the superconducting-condensation energy in the absence of a magnetic film on the surface. The values of  $E_m$  and  $E_s$  are the same for both the  $F$  and  $DS$  phases, are independent  $b$ , and their specific form is immaterial.

The total energy for the ferromagnetic state in the film is

$$E_F = n \frac{\tau^2}{\eta} \xi \frac{2 \cdot 2^{1/2}}{3} + E_m + E_s. \quad (8)$$

The period of the domain structure is determined from the condition  $\partial E_{DS} / \partial b = 0$ , and the condition for the transition from the ferromagnetic to the  $DS$  structure is the equality  $E_{DS} = E_F$ . The parameter  $C$  in (7) is determined by the boundary condition (2), where  $\gamma$  depends on  $b$ .

A complete solution of the problem calls thus for finding the boundary condition, i.e., for calculating the parameter  $\gamma$  as a function of  $b$ .

### 3. BOUNDARY CONDITION

Assuming the mean free path of the electrons in the ferromagnetic film to be small, we use the Usadel equations<sup>8</sup> to describe the superconductivity in the  $DS$ -phase region ( $0 < x < d$ , see Fig. 1).

In an approximation linear in the function  $F$  (the anom-

alous Green's function integrated over the energies and velocity directions on the Fermi surface), the Usadel equation in the presence of the exchange field  $h$  is

$$[\omega + ih(x, y)]F - D^{-1}D\nabla^2 F = \Delta(x, y), \quad (9)$$

where  $\omega = \pi T(2n + 1)$ ,  $D = v_F l / 3$  is the diffusion coefficient, and the approximation linear in  $F$  is used because we are considering the temperature region near  $T_c$ .

Rapid oscillations of the exchange field in the  $DS$  phase along the  $y$  coordinate cause a weak dependence of  $\Delta$  on  $y$ , which can be neglected, i.e.,  $\Delta \approx \Delta(x)$ . Using for  $F(x, y)$  the representation

$$F(x, y) = F_0(x) + \sum_k F_k(x) e^{ikh}, \quad (10)$$

where  $k \equiv k_n = \pi n / b$ ,  $n = 1, 2, \dots$ , and using the condition  $v_F / lk^2 \gg h \gg T_c$ , we obtain for  $F_0(x)$  the equation

$$\left(\omega + \frac{1}{\tau_s}\right)F_0 - \frac{D}{2} \frac{d^2 F_0}{dx^2} = \Delta(x), \quad (11)$$

where

$$\frac{1}{\tau_s} = \sum_k \frac{2|h_k|^2}{Dk^2} \approx 0.5 \frac{h^2 b^2}{v_F l}. \quad (12)$$

As seen from (11) the action of a  $DS$  structure on the superconductivity is analogous to that of magnetic scattering with a reciprocal time  $\tau_s^{-1}$  given by Eq. (12). This result is general for dirty superconductors, see Ref. 3.

From Eq. (1) and from the self-consistency condition we find that the behavior of the superconducting order parameter in the film is described by the equation

$$\begin{aligned} \Delta(x) \ln \left( \frac{T_c}{T} \right) \\ = \left[ \psi \left( \frac{1}{2} + \frac{1}{4\pi T} \left( \frac{2}{\tau_s} - D \frac{d^2}{dx^2} \right) \right) - \psi \left( \frac{1}{2} \right) \right] \Delta(x); \end{aligned} \quad (13)$$

here  $\psi(x)$  is a digamma function and it follows from (3) that the solution of interest to us, which satisfies the condition  $\Delta'(x) = 0$  on the interface with the vacuum, is of the form

$$\Delta(x) = \text{ch } qx, \quad q^2 = 2/\tau_s D. \quad (14)$$

The boundary condition (2) takes thus the form

$$\gamma = \frac{\Delta'(x)}{\Delta(x)} \Big|_{x=d} = q \text{th}(qd) \approx q^2 d = \frac{2d}{\tau_s D}. \quad (15)$$

#### 4. $F$ - $DS$ TRANSITION IN A MAGNETIC FILM

We determine the equilibrium energy in the case of a  $DS$  phase in the film. From the condition  $\partial E_{DS} / \partial b = 0$ , taking into account in (7) the dependence of  $C$  on the period  $b$  of the structure [Eqs. (12) and (15)], we get

$$n\Theta \frac{da}{b} = n \frac{\tau^2}{\eta} 2^{1/2} \xi \frac{\text{th } 2C}{\text{ch}^4 C}. \quad (16)$$

Using this condition, we find that at the transition point ( $E_{DS} = E_F$ ) the constant  $C$  is given by

$$\text{th}^2 C = 1 - 2/7^{1/2} \approx 0.24, \quad (17)$$

and since  $\sinh 2C = 2^{1/2} / \gamma \xi$ , we have

$$b = 2^{1/2} l v_F / h (\xi d \text{sh } 2C)^{1/2}.$$

Substituting this value of  $b$  in (16), we get the relative transition temperature

$$\tau_0^{-1/4} = \frac{v_F l}{da h} \frac{2^{1/2}}{\Theta \eta} \left( \frac{2\xi_0}{d} \right)^{1/2} \frac{\text{th } 2C}{\text{ch}^4 C \text{sh}^{1/2} 2C}. \quad (18)$$

It is taken into account here that  $\xi = \xi_0 \tau_0^{-1/2}$ . In order of magnitude, as follows from (18), we have

$$\tau_0^{1/4} \sim \Theta \frac{h}{T_c^2} \frac{d}{l} \left( \frac{d}{\xi_0} \right)^{1/2}. \quad (19)$$

Our analysis is qualitatively valid also at low temperatures, therefore the general condition for the transition to the  $DS$  phase

$$\frac{\Theta h}{T_c^2} \frac{d}{l} \left( \frac{d}{\xi_0} \right)^{1/2} \leq 1. \quad (20)$$

For a ferromagnet with RKKY interaction we have  $\Theta \approx h^2 / \varepsilon_F$ , and at  $\Theta \approx T_c$ ,  $h \sim 100$  K, and  $d \sim 10$  Å this condition is met for superconductors with  $\xi_0 \gtrsim 10^3$  Å.

The period of the  $DS$  structure at the transition point is of the order of  $d(\Theta/T_c)$ , and the destructive action of the exchange field in the  $DS$  phase turns out to be relatively weak ( $\tau_s^{-1} < T_c$ ). Note that we have disregarded magnetic scattering in the film. Below the Curie point in the presence of easy-axis magnetic anisotropy, magnetic scattering is strongly suppressed. In principle, however, its influence can be easily taken into account, and furthermore the foregoing conclusions remain qualitatively in force even for a reciprocal magnetic scattering time  $\tau_m^{-1} \sim \Theta$ .

It follows from our analysis that the most favorable conditions for the observation of the  $DS$ - $F$  transition are produced in superconductors with a large correlation length  $\xi_0$ . Furthermore, the effect is enhanced by the high density of the electronic states in the superconductor.

We have considered a system in which the magnetic film is on the surface of a superconductor. A similar situation can arise also in a superlattice consisting of thick superconductor slabs separated by thin ferromagnetic-metal layers. The  $F$ - $DS$  transition in the layer leads here to a first-order transition from two-dimensional to three-dimensional superconductor behavior.

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