

Kent, Ohio, 12-16 August 1968 (to be published).

<sup>4</sup>For a survey of the continuum theory of liquid crystals, see J. L. Ericksen, *Appl. Mech. Rev.* **20**, 1029 (1967). The basic formulation may also be found in F. M. Leslie, *Proc. Roy. Soc. (London), Ser. A* **307**, 359 (1968).

<sup>5</sup>A theory of boundary layers has been developed by F. M. Leslie, *Arch. Rational Mech. Anal.* **28**, 265 (1968).

<sup>6</sup>M. Miesowicz, *Nature* **158**, 27 (1946).

<sup>7</sup>A detailed study of flow in circular capillaries is

planned to be published elsewhere.

<sup>8</sup>See, e.g., G. W. Gray, *Molecular Structure and the Properties of Liquid Crystals* (Academic Press, Inc., New York, 1962). From the green appearance we estimate  $P=3000 \text{ \AA}$ , assuming the refractive index to be 2.

<sup>9</sup>W. Helfrich, *J. Chem. Phys.* **50**, 100 (1969). In the notation of that paper  $\kappa_1 + \kappa_2$  stands for  $-\lambda_1$ .

<sup>10</sup>The slight bending of the planes of equal alignment does not contradict and is in fact identical with the rather strong distortion of the orientation pattern in the planes normal to the helical axis (Ref. 7).

## COUPLING BETWEEN FERRIMAGNETIC INSULATORS THROUGH A SUPERCONDUCTING LAYER

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In agreement with a recent theoretical prediction of de Gennes, the transition temperature of In (or another superconductor) films sandwiched between two magnetic films depends on the relative orientation of the magnetization of these ferrites. The highest transition temperature (nearly the  $T_c$  of bulk In, 3.4°K) is obtained when the two ferrites are deposited in antiparallel magnetic fields, while the lowest ( $\approx 1^\circ\text{K}$ ) occurs when the ferrites are deposited in parallel magnetic fields. The depression of the transition temperature of an indium film sputtered or evaporated between two magnetic films magnetized in parallel fields is inversely proportional to the thickness of the In film.

Recently, de Gennes<sup>1</sup> predicted theoretically that two ferromagnetic insulating layers with respective magnetizations  $M_1$  and  $M_2$  are coupled when a superconducting film of thickness smaller than 1 coherence length  $\xi_0$  is placed inbetween. The most striking conclusion was that the transition temperature  $T_c$  of the superconducting film would be highest if  $M_1$  and  $M_2$  were antiparallel and lowest if  $M_1$  and  $M_2$  were parallel. In agreement with the theory it is found that a 3000- $\text{\AA}$  In film sandwiched between two  $\text{Fe}_3\text{O}_4$  films with parallel magnetizations has its  $T_c$  reduced by 1.5°K while the same film sandwiched between two  $\text{Fe}_3\text{O}_4$  films deposited in antiparallel magnetic fields displays a reduction of about 0.3°K. Furthermore, a 3000- $\text{\AA}$  In film sandwiched between two randomly magnetized  $\text{Fe}_3\text{O}_4$  films (thus partly antiparallel) showed a reduction of 0.8°K. If a 500- $\text{\AA}$   $\text{Al}_2\text{O}_3$  layer is deposited between the In and two  $\text{Fe}_3\text{O}_4$  layers magnetized in parallel fields, the In film displays the bulk  $T_c$  of 3.4°K, thus ruling out closure fields as the cause of the depression of  $T_c$ . It was also established, in agreement with de Gennes's theory,<sup>1</sup> that the depression of  $T_c$  of such triple layers is inversely proportional to the thickness of the superconducting film  $d$ . These experiments demonstrate that the reduction in  $T_c$  is due to the exchange field produced by the two magnetic layers and not by

the closure field of magnetic domains. Finally, it was estimated from such experiments that the exchange integral  $\Gamma$  was approximately 0.3 eV.

When a superconducting film is in contact with one or two insulating ferrimagnets three effects can be taking place: firstly, a magnetic field effect either from the closure field of domain walls or from the component of magnetization normal to the film surface; secondly, the first-order effect discussed by de Gennes<sup>1</sup> which is caused by the exchange field of the magnetic layers; and thirdly, a second-order effect which can be called an impurity effect and is similar to the one found in dilute alloys.

Most of the experiments described in this study were performed with films of magnetite ( $\text{Fe}_3\text{O}_4$ ). The magnetite films were prepared as follows: A powder mixture (8 parts Fe powder to 92 parts  $\text{Fe}_2\text{O}_3$  powder) was pressed into the form of a button and sintered for 2 h at 700°C in an argon atmosphere; after pressing an iron stem into this button, this target was used in conjunction with the getter sputtering technique<sup>2</sup> to deposit a film usually 2000  $\text{\AA}$  thick. The  $\text{Fe}_3\text{O}_4$  films had the spinel cubic structure of bulk  $\text{Fe}_3\text{O}_4$  and a room-temperature resistivity of 0.01  $\Omega \text{ cm}$  which is close to that of bulk magnetite ( $7 \times 10^{-3} \Omega \text{ cm}$ ); the resistivity of the  $\text{Fe}_3\text{O}_4$  films increased to 100  $\Omega \text{ cm}$  at 77°K which again is in agreement

with bulk behavior. Some triple layers were made and kept at 77°K until the  $T_c$  was measured; the triple layers were then warmed up to room temperature and remeasured. It was found that the  $T_c$  and the resistivity ratio of the indium film remained unchanged. Furthermore, these quantities remained unchanged if the triple layer remained at room temperature for two days. It can therefore be concluded that Fe does not diffuse from magnetite into In and for this reason, most of the sandwiches studied were taken out at room temperature. The evaporated In films ranged in thickness from 1700 to 44 000 Å with a resistivity ratio varying from 15 to 175. The sputtered In films ranged in thickness from 1500 to 4000 Å with resistivity ratio varying from 4 to 10.

The first experiments on the influence of an insulating ferromagnetic material on a superconducting film<sup>3,4</sup> were performed by evaporating Sn or In on a polished yttrium iron garnet (YIG) crystal and on FeNi ferrites prepared by oxidation of a NiFe alloy on a quartz substrate. These experiments had the following drawbacks: Firstly, only one ferrite was used thereby diminishing the exchange interaction; secondly, the ferrite was always prepared outside the vacuum system, and the present study has revealed that if the bell jar is opened between the deposition of the ferrites and the In film, the reduction in  $T_c$ , although not eliminated, is greatly reduced. Thirdly, a large portion of the effect reported was due to the magnetic field of magnetic domains. Indeed, even if all the magnetizations of the domains lie in the plane of the substrate there still can be field effects if the substrates have scratches, which was certainly the case in

YIG and in quartz. Further details will be given on this magnetic field effect in a later publication; it is sufficient to say here that in order to eliminate such field effects one must deposit the superconducting film on a ferrite film which has been deposited on a glass microslide or some other smooth surface. A recent attempt to study the coupling between ferromagnetic layers<sup>5</sup> used the surface oxidation of FeNi and Ni as insulating layers. However, as the junction resistance between the ferromagnetic layer and the superconductor was  $10^{-3} \Omega$ , it is more than likely that many pinholes were present and that an appreciable portion of the effect is due to the proximity effect of the conducting ferromagnet.<sup>6</sup>

Figure 1 shows the  $T_c$  of In films both sputtered and evaporated as a function of inverse In film thickness. Although the results are shown for ferrites grown in a parallel field of 400 G, triple layers deposited in fields of 100 G or 2500 G give identical results. It is clear from Fig. 1 that except for some scatter especially present in the thinnest sputtered films, the depression of  $T_c$  of triple layers deposited in parallel fields is inversely proportional to the In film thickness. Furthermore, a 500-Å  $Al_2O_3$  layer deposited between the In and the magnetite films completely eliminates the effect shown in Fig. 1: Varying the In film thickness between 1270 and 3380 Å resulted in a constant  $T_c$  of 3.4°K thereby proving that one is truly dealing with the contact interaction discussed by de Gennes.<sup>1</sup> Table I shows the  $T_c$  for In films evaporated between two ferrites deposited in 100-G antiparallel fields. Again in agreement with de Gennes's theory, one can see that when the magnetizations of the two ferrites are predominantly antiparallel the reduction in  $T_c$  is very small, 0.35°K at most. If on the other hand, the In films are evaporated between random ferrites, i.e., ferrites deposited in the absence of an applied magnetic field (crosses in

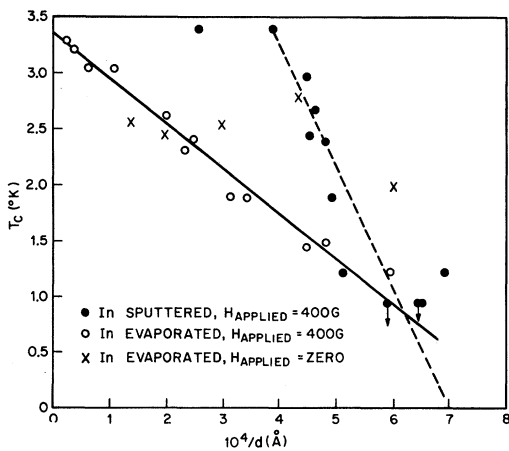


FIG. 1. Transition temperature of  $Fe_3O_4$ -In- $Fe_3O_4$  sandwiches as a function of inverse In film thickness  $d$ .

Table I. Transition temperatures of evaporated In films sandwiched between two ferrite films deposited in antiparallel magnetic fields.

$T_c$ (°K)	In thickness (Å)	$\frac{\rho(RT)}{\rho(4.2°K)}$
3,314	2540	24
3,046	3460	26
3,146	3720	37
3,354	4220	26
3,289	4740	32
3,110	5850	47

Fig. 1), the  $T_c$ 's fall between those obtained on triple layers composed of ferrites with parallel and antiparrel magnetizations. The scatter for the random ferrites is understandable from the fact that the relative orientations of the magnetizations of two random ferrites will change from run to run. As mentioned above, de Gennes<sup>1</sup> discussed two effects (exchange field and impurity) which are contact effects. The strength of each effect is inversely proportional to the superconduction-film thickness. But the fact that, as shown in Fig. 1 and Table I, the  $T_c$  of an In film depends on the relative orientation of the magnetizations of the ferrites clearly demonstrates that the greater part of the effect reported here is due to the exchange field of the two ferrite films. In a random ferrite the magnetizations lie in all directions within the plane of the ferrite film as a result of the low magnetocrystalline anisotropy of  $\text{Fe}_3\text{O}_4$  and the high shape anisotropy of the film. Therefore, when parallel and antiparallel configurations are mentioned, it is only meant to represent a trend and by no means implies that all magnetizations are parallel or antiparallel.

The sputtered films always satisfy the condition  $l < d$ , while the evaporated films correspond to  $l \simeq d$ , where  $l$  is the electron mean free path. As the exchange field should scale with  $\xi_0/d$ ,<sup>1</sup> and as when  $l < \xi_0$  the pertinent parameter becomes  $\xi_{\text{eff}} = (\xi_0 l)^{1/2}$ , this suggests a plot of the transition temperature as a function of  $\xi_{\text{eff}}/d$ , which is shown in Fig. 2. The first point to be made about Fig. 2 is that the difference between sputtered and evaporated films is appreciably re-

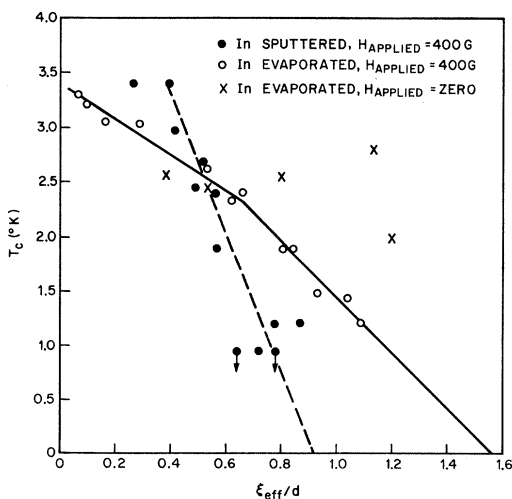


FIG. 2. Transition temperature of  $\text{Fe}_3\text{O}_4$ -In- $\text{Fe}_3\text{O}_4$  sandwiches as a function of  $\xi_{\text{eff}}/d$ , where  $\xi_{\text{eff}} = (\xi_0 l)^{1/2}$  with  $\xi_0 = 2700 \text{ \AA}$  and  $l = (84 \text{ \AA}) \times \rho(\text{RT})/\rho(4.2^\circ\text{K})$ .

duced from that shown in Fig. 1. Indeed, at a given value of  $\xi_{\text{eff}}/d$  the  $T_c$  difference between sputtered and evaporated films is at most  $0.75^\circ\text{K}$ . The curve for the evaporated films shows a kink at  $\xi_{\text{eff}}/d = 0.7$  which is about the film thickness where  $\xi_0/l = 1$ . Consequently, it is believed that the lower slope for the thicker evaporated films (where  $l > \xi_0$ ) is predominantly caused by the impurity effect. This is also supported by the fact that in that region the  $T_c$  is almost independent of the relative orientation of the magnetization of the ferrites. It follows from de Gennes's<sup>1</sup> model that the curve close to  $T_c = 0^\circ\text{K}$  can be approximated by the relationship

$$T_c/T_{c0} = 1 - 10(\Gamma S/E_F)(\xi_0/d), \quad (1)$$

where  $T_c$  is the transition temperature of the triple layer and  $T_{c0}$  the transition temperature of the pure superconductor.  $\Gamma$  is the exchange integral from the coupling between a ferromagnetic spin  $S$  and a conduction-electron spin  $S_e$ , and  $E_F$  is the Fermi energy of the superconductor. The exchange integral can therefore be evaluated either from the slope or from the intercept at  $T_c = 0^\circ\text{K}$ . The value of  $\Gamma$  thus obtained using  $E_F = 5 \text{ eV}$  and  $S = 2$  is  $(0.2-0.16) \text{ eV}$  for evaporated films and  $(0.45-0.27) \text{ eV}$  for sputtered films. Although this estimate of  $\Gamma$  is smaller or equal to the real  $\Gamma$  because of a possible decoupling between the ferrites and the In, it is in good agreement with the values reported on alloys. The lower value of  $\Gamma$  obtained with evaporated films is most probably due to the fact that a sputtered film makes better contact with a substrate than an evaporated film.

The depression of  $T_c$  of In films on a single ferrite is also inversely proportional to the In film thickness. A complete report including the effect of a single ferrite on a superconducting film as well as tunneling studies will be published later. I would like to thank W. H. Haemerle for his technical help, and J. H. Condon, R. C. Sherwood, and H. C. Theuerer for useful discussions.

<sup>1</sup>P. G. de Gennes, *Phys. Letters* **23**, 10 (1966).

<sup>2</sup>J. J. Hauser, H. C. Theuerer, and N. R. Werthamer, *Phys. Rev.* **136**, A637 (1964).

<sup>3</sup>J. Rosenblatt, J. J. Hauser, C. Valette, and J. P. Burger, in *Proceedings of the Eleventh International Conference on Low Temperature Physics, St. Andrews, Scotland, 1968*, edited by J. F. Allen, D. M. Findlayson, and D. M. McCall (St. Andrews University, St. Andrews, Scotland, 1968).

<sup>4</sup>J. Rosenblatt, J. J. Hauser, and J. P. Burger, to be published.

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<sup>6</sup>J. J. Hauser, H. C. Theuerer, and N. R. Werthamer, Phys. Rev. 142, 118 (1966).

## EFFECT OF FLUCTUATIONS IN THE SUPERCONDUCTING ORDER PARAMETER ON THE TUNNELING DENSITY OF STATES

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Fluctuations in the superconducting order parameter above the transition temperature result in structure in the tunneling density of states. From the shape of this structure a value is derived for the decay rate of the fluctuations.

Fluctuations in the superconducting order parameter above the transition temperature are predicted to affect a variety of experimental quantities, e.g., electrical conductivity,<sup>1</sup> specific heat,<sup>2</sup> and diamagnetic susceptibility.<sup>3</sup> Fluctuation effects have so far been reported only in the electrical conductivity.<sup>4,5</sup> We have observed the effect of fluctuations on the electronic density of states (EDS), derived from tunneling measurements.

The measurements were carried out on  $M_1$ -I- $M_2$  junctions in which  $M_1$  is a metal film in the fluctuation state above the transition temperature  $T_{c1}$ ,  $M_2$  is a film in the superconducting state, and I is a thin insulating layer. We find that the EDS in the fluctuation regime of  $M_1$  exhibits a depression from the normal-state value over a small energy range  $E_c$  centered about the Fermi level. As the temperature is raised the EDS approaches that of the normal metal. As the temperature is lowered the depression in the EDS deepens and for  $T < T_{c1}$  develops into an energy gap in the quasiparticle spectrum. From the observed structure in the EDS the decay rate of fluctuations in the Cooper-pair density is deduced.

Metal  $M_1$  was granular Al, a material which

has been used previously by several workers to study fluctuation phenomena.<sup>5-7</sup> The granular Al was deposited onto glass slides by using two different techniques: evaporation in an oxygen atmosphere<sup>8</sup> or cosputtering<sup>9</sup> Al and  $\text{SiO}_2$ . The insulating layer I was  $\text{Al}_x\text{O}_y$  which was grown by exposing the granular Al to laboratory air for several minutes. The film  $M_2$  was In, Pb, or the alloy<sup>10</sup>  $\text{Pb}_{0.7}\text{Bi}_{0.3}$ , approximately 3000 Å thick, evaporated on top of the insulating layer. The parameters of the three junctions reported in this Letter are given in Table I. Here,  $R_j$  is the junction resistance and  $\Delta_j(0)$  is the energy gap of metal  $M_j$  at  $T=0^\circ\text{K}$ . The quantity  $\rho_0$  is the normal resistivity and  $d$  is the thickness of the granular Al film. The length  $(\xi_0 l)^{1/2}$  ( $\xi_0$  is the Pippard coherence length and  $l$  is the effective mean free path<sup>11</sup>) appears in the expression for the temperature-dependent Landau-Ginzburg coherence length<sup>12</sup>  $\xi(T) = 0.85(\xi_0 l)^{1/2} \epsilon^{-1/2}(T)$ , where  $\epsilon(T) = (T - T_{c1})/T_{c1}$ .

The first derivative  $dI/dV$  and the second derivative  $d^2I/dV^2$  of the junction  $I$ - $V$  characteristics were determined by the conventional technique of superposing a small audio-frequency voltage  $v$  on the dc bias  $V$  and measuring the resulting ac cur-

Table I. Properties of junctions.

Junction	$R_j$ ( $\Omega$ )	$M_2$	$\Delta_2$ (mV)	$M_1$	$T_{c1}$ ( $^\circ\text{K}$ )	$2\Delta_1(0)/k_B T_{c1}$	$\rho_0$ ( $10^{-6} \Omega \text{ cm}$ )	$d$ ( $\text{\AA}$ )	$(\xi_0 l)^{1/2}$ ( $\text{\AA}$ ) <sup>a</sup>
I	990	$\text{Pb}_{0.7}\text{Bi}_{0.3}$	1.794	Sput.	2.59	3.45	840	2460	120
II	14.4	$\text{Pb}_{0.7}\text{Bi}_{0.3}$	1.766	Evap.	2.36	3.54	72	550	420
III	14.7	Pb	1.280	Evap.	2.12	3.47	38	800	580

<sup>a</sup>The Pippard coherence lengths  $\xi_0$  were computed by multiplying the value  $\xi_0 = 16000 \text{\AA}$  for ordinary Al ( $T_c = 1.18^\circ\text{K}$ ) by ratio of the energy gap  $\Delta_1(0)$  of ordinary Al to that of the granular film. The effective mean free paths  $l$  were calculated from the formula  $\rho_0 l = 1.6 \times 10^{-11} \Omega \text{ cm}^2$  [J. L. Olson, Electron Transport in Metals (Interscience Publishers, Inc., New York, 1962), p. 84].