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Transition Temperature and Tunneling Characteristics of Ferrimagnetic-Insulator–Superconductor Sandwiches

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The depression of the transition temperature of In films (thinner than the effective coherence length ξ_{eff}) on a single ferrite film grown in a parallel magnetic field is inversely proportional to the In film thickness. This result agrees with that already reported for an indium film sandwiched between two ferrites magnetized in parallel field; the transition temperature for the single-ferrite case can be obtained from the double-ferrite case by multiplying the ratio ξ_{eff}/d by 2 (which, except for a small mean-free-path correction, means dividing the indium film thickness d by 2). The same results are obtained if indium is replaced by tin. Tunneling junctions (Al-Al_2O_4-In-Fe_3O_4) were investigated as a function of temperature and In film thickness. The In films were found to display gapless or quasigapless superconductivity. For a fixed reduced transition temperature, the degree of gapless superconductivity is the same as that obtained by the proximity effect of a conducting ferromagnet.

I. INTRODUCTION

In agreement with a theoretical prediction of de Gennes, the transition temperature of In films sandwiched between two Fe_3O_4 films was reported[1,2] to depend on the relative orientation of the magnetizations of the two Fe_3O_4 films. The maximum depression of T_c occurred when the ferrites were deposited in parallel magnetic fields, and the minimum depression occurred when the second ferrite film was deposited in a field antiparallel to the first. Furthermore, the depression of T_c of In films (thinner than two effective coherence lengths ξ_{eff}) sandwiched between two ferrites magnetized in parallel fields was inversely proportional to the In film thickness. Further studies of the effect of an insulating ferrite on a superconductor have been performed by tunneling. As the tunneling junctions will be of the type Al-Al_2O_3-In-Fe_3O_4, it is necessary to study the effect of a single ferrite on a superconducting film and compare this effect with the one previously reported on the superconducting film sandwiched between two ferrites. Finally, the observed dependence of T_c on ξ_{eff}/d (where d is the indium film thickness) will be shown to be independent of the superconductor by replacing indium by tin.

II. EXPERIMENTAL PROCEDURE

As most of the experimental procedure has already been described,[1,2] we confine ourselves here to the few experimental points not mentioned before. The tin films were deposited similarly to the indium films either by evaporation or by getter-sputtering at 77°K. The tunnel junctions were prepared by depositing a 1000 Å thick aluminum strip, followed by thermal oxidation and the deposition of the sandwich as a cross strip. The cross strip sandwich was deposited by sputtering an indium film of known thickness at 77°K and then a 2000 Å Fe_3O_4 film was sputtered over at about 125°K. Some of the sandwiches were made and kept at 77°K until measured in order to avoid spurious diffusion effects. These junctions were then warmed up to room temperature and remeasured. As the junctions remained essentially unchanged, most tunnel junctions were taken out at room temperature before being measured. All the Fe_3O_4 films used in this study were sputtered in a 400-G magnetic field applied parallel to the plane of the film.

III. EXPERIMENTAL RESULTS AND DISCUSSION

As already pointed out,[1,2] it is of the utmost importance in a ferrite-superconductor proximity effect to avoid magnetic field effects which can originate from three sources: closure field of magnetic domains, scratches in a ferrite with the magnetization lying in the plane of the ferrite, and a ferrite with a normal component of the magnetization. This last effect can be demonstrated by evaporating an 18 000 Å indium film on the basal plane of a $BaFe_8Al_4O_{19}$ ferrite.[3] The magnetization of this ferrite is perpendicular to the basal plane, and thus to the film, and as a result, the T_c of this indium film was below 1°K. The same indium film evaporated on an Fe_3O_4 film deposited on a smooth microscope slide has almost the bulk transition temperature of indium (3.4°K). A quantitative estimate of the field effect can be obtained by depositing simultaneously two 1150 Å Pb films, one on a glass substrate and one on the basal plane of $BaFe_8Al_4O_{19}$. The T_c of the Pb film on the ferrite was 6.35°K and as a transverse field of 1 kG was necessary to depress the T_c of the lead film on the glass substrate down to 6.35°K, one can conclude that the Ba ferrite produces a transverse magnetic field of the order of 1 kG.[4] The $BaFe_8Al_4O_{19}$

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[1] J. J. Hauser, Phys. Rev. Letters **23**, 374 (1969).

[2] J. J. Hauser, in International Conference on the Science of Superconductivity, Stanford University, Aug. 26–29, 1969 (unpublished).

[3] I am indebted to L. G. Van Uitert for making this crystal available.

[4] R. C. Sherwood with susceptibility measurements obtained a value of $4\pi M_s = 4$ kG for this Ba ferrite. The lower value found by the proximity effect with lead is due to the fact that the surface of the Ba ferrite is filled with closure domains which reduce the surface transverse magnetic field.

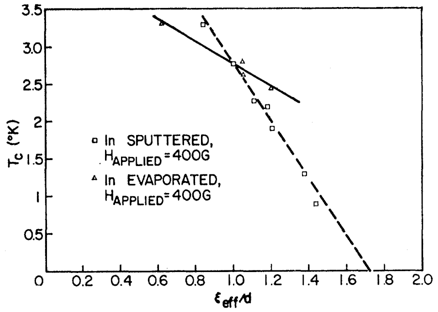

FIG. 1. Transition temperatures of Fe_3O_4-In sandwiches as a function of ξ_{eff}/d, where $\xi_{eff}=(\xi_0/l)^{1/2}$ with $\xi_0=2700$ Å and $l=(84\text{ Å})\times\rho(\text{RT})/\rho(4.2°\text{K})$.

ferrite was now cut and polished parallel to the c axis, and a 10 000 Å indium film was deposited on this new face which now contains the magnetization. The T_c was now 2.67°K and the transition width 0.3°K. This large reduction in T_c is due to the polishing scratches in which the In film senses the magnetization of the ferrite. Thus to avoid such field effects the superconducting film should be deposited on a ferrite film which has been deposited on a glass microslide or some other very smooth surface. The absence of closure field effects from magnetic domains can be expected with Fe_3O_4 films as Fe_3O_4 has a very low magnetocrystalline anisotropy and the very high shape anisotropy of the film will ensure that the magnetization lies in the plane of the film. The absence of any field effect was already demonstrated[1,2] by depositing a 500 Å Al_2O_3 layer between a thin In film and the Fe_3O_4 films which resulted in an In film with the bulk T_c of 3.4°K.

While previous experiments dealt with the coupling of two Fe_3O_4 films through a superconducting layer,[1,2] we will concentrate here on the effect of a single Fe_3O_4 film (deposited in a 400-G field parallel to the film) on a superconducting film. In the case of the two ferrites, the exchange field acting on the electrons of the superconductor was maximum when the magnetizations of the two ferrites were parallel, and minimum when the magnetizations were antiparallel. The exchange field from a single ferrite comes from the magnetizations of its many domains, and if the ferrite is deposited in a parallel magnetic field these magnetizations will be mostly parallel, and the exchange field effect will be maximum. The results for such an experiment is shown in Fig. 1. To prove again that most of the effect reported here is due to the exchange field of the ferrite (and not to the impurity effect caused by the slight diffusion of some impurity spins into the superconductor) one can deposit the ferrite without magnetic field. Although the magnetizations of the domains will still lie in the plane of the ferrite film, some of them will be antiparallel, thereby reducing the net exchange field. Two sandwiches were prepared in this manner with two evaporated indium films yielding the following results: $d=2720$ Å, $T_c=3.36$°K, $\xi_{eff}/d=0.93$; $d=1510$ Å, $T_c=3$°K, $\xi_{eff}/d=1.46$ and it can be seen from Fig. 1 that, as expected, these points are appreciably above the solid line. The data for the sputtered films shown in Fig. 1 are fitted fairly well by the dashed line. This dashed line can be obtained from the line averaging the data for the two ferrite case (see dashed line in Fig. 2 of Ref. 1) by multiplying ξ_{eff}/d by 2. This means that, except for a small mean free path correction, the data for one ferrite can be obtained from the two ferrite case by dividing the indium film thickness by 2. In other words, the exchange field produced by one ferrite deposited in a parallel field is approximately half the exchange field produced by two such ferrites. The linear behavior of T_c versus ξ_{eff}/d is not in complete agreement with de Gennes's paper.[5] In that paper he states that the exchange field should be inversely proportional to d and he suggests[6] that his expression for the exchange field should be inserted in Sarma's[7] relationship for T_c; this procedure will not yield the observed linear behavior of T_c versus $1/d$. This lack of agreement is most probably due to the fact that Sarma's theory was derived for a uniform exchange field throughout the superconductor, which of course leads to a constant order parameter ψ. In fact, there is only a contact interaction between a conduction electron of the superconductor and the spin of the ferrimagnet in the interface plane. Most probably, this contact interaction leads to a function ψ which is minimum at the interface and increases away from the interface. The average ψ might still be considered to be the result of an effective exchange field which is an increasing function of $1/d$, but quantitative agreement with the de Gennes–Sarma theory would not be expected. This point will be further discussed in connection with the tunneling data. The data for the evaporated In films is scant as films thinner than 1500 Å have a tendency to ball-up. The small amount of data for the evaporated films shown in Fig. 1 can again be obtained from the curve for two ferrites (see lower solid line[8] of Fig. 2 of Ref. 1) by multiplying ξ_{eff}/d by 2.

In order to show the generality of this exchange effect, some of the experiments have been repeated with In replaced by Sn. The results are shown in Fig. 2 where the reduced transition temperature ($t_c=T_c/T_{c0}$, where T_{c0} is the transition temperature of a film deposited on glass at 77°K: 3.4°K for In and 3.86°K for Sn) of various sandwiches is plotted as a function of ξ_{eff}/d. The data of Ref. 1 for Fe_3O_4-In-Fe_3O_4 sandwiches are shown by the solid line of Fig. 2. The Fe_3O_4-Sn-Fe_3O_4 sandwiches are represented by squares for the evaporated Sn films and by triangles for the sputtered ones.

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[5] P. G. de Gennes, Phys. Letters 23, 10 (1966).

[6] I am indebted to G. Deutscher for pointing this out to me.

[7] G. Sarma, J. Phys. Chem. Solids 24, 1029 (1963).

[8] As the films for the single ferrite case always satisfy $l\ll\xi_0$ we do not obtain the upper solid line of Fig. 2 of Ref. 1 which correspond to $l\gg\xi_0$.

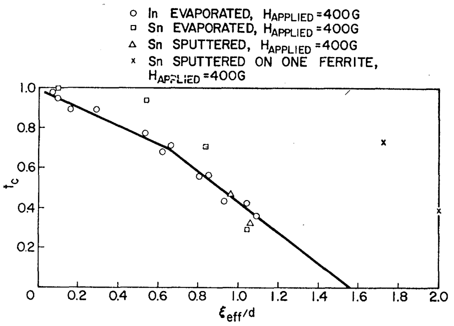

FIG. 2. Reduced transition temperatures for Fe_3O_4-In-Fe_3O_4 and Fe_3O_4-Sn-Fe_3O_4 sandwiches and Fe_3O_4-Sn sandwiches as a function of ξ_{eff}/d, where $\xi_{\mathrm{eff}}=(\xi_0/l)^{1/2}$ with $\xi_0=2700$ Å and $l=(84\ \text{Å})\times\rho(\mathrm{RT})/\rho(4.2°\mathrm{K})$ for In and $\xi_0=2500$ Å and $l=(136\ \text{Å})\times\rho(\mathrm{RT})/\rho(4.2°\mathrm{K})$ for Sn.

Contrary to the In experiments, there is almost no difference between evaporated and sputtered Sn films. This can be explained by the fact that the difference in the mean free path between thin evaporated and sputtered tin films is smaller than the one between evaporated and sputtered indium films. The transition temperature of such sandwiches depends on the actual exchange field which is sensitive to the nature of the superconductor-ferrite interface, which in turn, depends on the superconductor and the way it is deposited (sputtering or evaporation and temperature of deposition). Considering all the variables which enter in such experiments the agreement between the Sn data and In data shown in Fig. 2 can be considered as good. Furthermore, the two data points shown in Fig. 2 obtained for

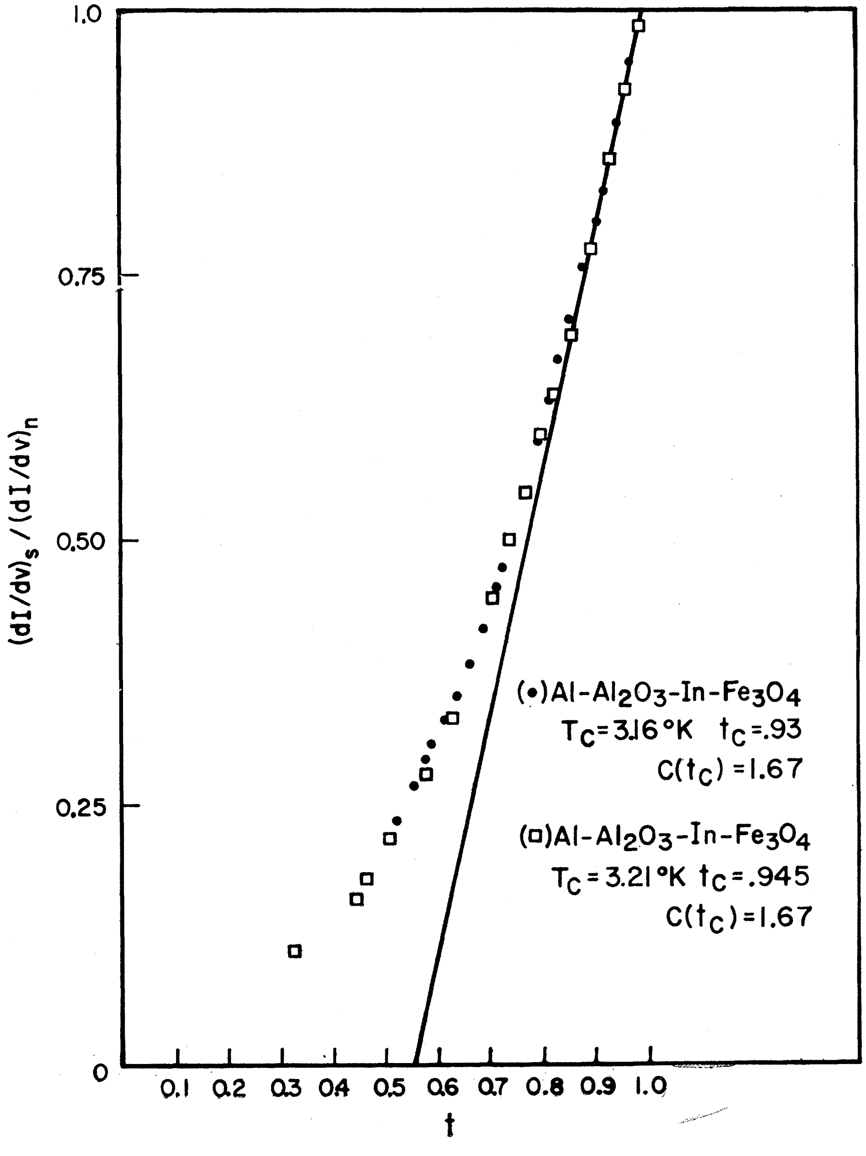

FIG. 3. Relative conductance at zero bias, $(dI/dV)_s/(dI/dV)_n$, as a function of $t=T/T_c$ for two Al-Al_2O_3-In-Fe_3O_4 junctions.

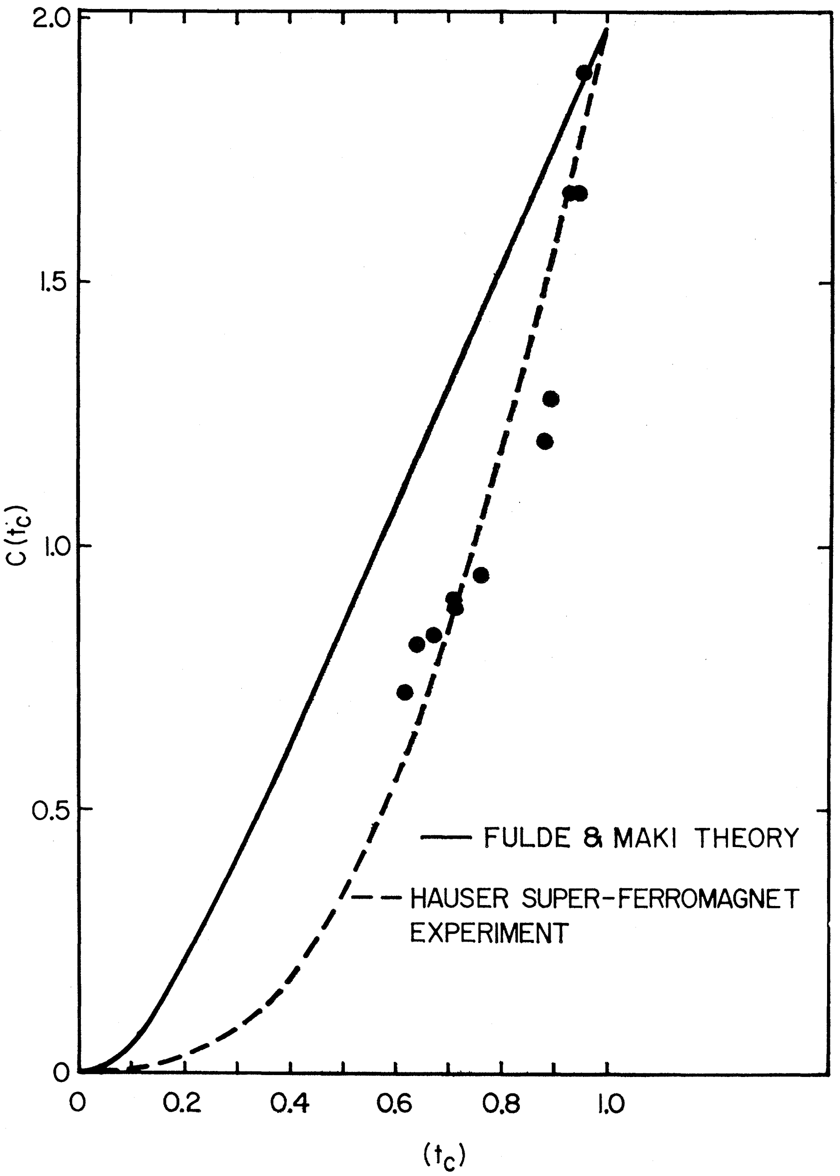

FIG. 4. Plot of $C(t_c)=0.75\{(d/dt)\times[(dI/dV)_s/(dI/dV)_n]\}_{t=1}$ as a function of $t_c=T_c/T_{c0}$, where T_{c0} is the bulk transition temperature of In (3.4°K), for In-Fe_3O_4 sandwiches. The dashed curve in this figure averages the data (Ref. 12, Fig. 5) obtained on Pb-Ni, Pb-Fe, and Pb-Pt sandwiches.

Sn films on a single ferrite, can again be deduced from the double ferrite case by multiplying ξ_{eff}/d by 2.

We now turn our attention to the tunneling experiments on In-Fe_3O_4 sandwiches. The transition temperature of the sandwiches is the temperature at which the dV/dI characteristic is flat; this temperature is in very good agreement with the resistive transition temperature quoted above. Data could not be obtained on In films thinner than 700 Å (which corresponds to $t_c=T_c/T_{c0}=0.6$) because junctions with such thin indium films resulted in shorts. For each In film thickness, dV/dI curves were obtained as a function of temperature from T_c down to the superconducting transition temperature of Al. The normalized conductance at zero bias $(dI/dV)_s/(dI/dV)_n$ is obtained by measuring the ratio $(dV/dI)_{v=4mv}/(dV/dI)_{v=0}$. This reduced conductance is shown as a function of temperature for two similar junctions in Fig. 3. Close to the transition temperature, the data can be fitted very well by a straight line. If one makes the same plot for an Al-Al_2O_3-In junction, the data points show a continuous curvature up to T_{c0}; the limiting slope at T_{c0} (which except for a slight strong coupling correction corresponds to the value obtained from the BCS theory) is 2.66, i.e., a line which intercepts the t axis at 0.625. Furthermore, at any reduced temperature t, the normalized conductance shown in Fig. 3 is larger than the one measured on the In junction. These facts suggest that the ferrite induces gapless or quasigapless superconductivity into the adjoining superconductor.

Fulde and Maki[9,10] have developed a theory of gapless superconductivity induced by the proximity effect of magnetic as well as nonmagnetic films. Among other quantities, they derive the ratio of the conductance at zero bias in the normal and the superconducting state. de Gennes and Mauro[11] pointed out that gapless superconductivity can only occur by proximity with a magnetic film, but it was shown experimentally,[12] that proximity with a normal film leads to "quasigapless" superconductivity which can be fitted by the Fulde-Maki theory. Although this theory does not apply to the present case (primarily because of the insulating nature of Fe_3O_4), we shall nevertheless compare its predictions with the present data. With the boundary condition that the order parameter ψ in the superconducting film be zero at the superconductor-magnetic film interface, Fulde and Maki[10,12] derived the following relation for the normalized conductance at zero bias:

$$1-(dI/dV)_s/(dI/dV)_n=\tfrac{4}{3}(1-t)C(t_c)\,, \qquad (1)$$

where $C(t_c)$ is a universal function of t_c and has been plotted as the solid curve of Fig. 4. Relation (1) predicts that the relative conductance approaches unity linearly as t approaches 1; this fact is verified by the data shown in Fig. 3 and by all other sandwiches. It can be seen from relation (1) that $C(t_c)$ can also be determined as $\frac{3}{4}$ of the experimentally determined slope of the relative conductance versus t at $t=1$. A plot of $C(t_c)$ determined in that manner is shown by the data points of Fig. 4.[13] Although the data points show a greater degree of gapless superconductivity than predicted by the Fulde-Maki theory, they are in excellent agreement with the gapless data obtained on Pb-Ni sandwiches and the quasigapless data obtained on Pb-Pt sandwiches.[12] In the present experiments, the extra conductance cannot arise from such spurious effects as pinholes in the superconducting film[12] because of the insulating nature of Fe_3O_4. Consequently, the data shown in Fig. 4 indicate that for a sandwich with a given reduced transition temperature t_c, the tunneling density of states is the same whether the depairing mechanism is the present exchange field or the normal or magnetic proximity effects. This gives further evidence that the uniform exchange field theory of Sarma[7] does not apply here. His theory was derived within the BCS framework, while as shown previously, the present tunneling density of states is very different from BCS. In the superconductor-ferrite experiments, we do not expect ψ to be determined by an effective boundary condition $\psi=0$ as used by Fulde and Maki to simulate the presence of the ferromagnetic metal. In fact, we expect $\partial\psi/\partial x=0$ (where x is normal to the interface; the boundary condition $\partial\psi/\partial x=0$ as discussed in the Ginzburg-Landau theory is the proper one to use at a superconductor-vacuum or at a superconductor-insulator interface). However, we do expect the contact exchange interaction to depress ψ some finite amount below its bulk value at $x=0$, and as a result lead to a spatially dependent order parameter in the superconducting film. This may be precisely the reason why the Fulke-Maki theory which was derived for a spatially dependent order parameter works so well for the present experiments.

ACKNOWLEDGMENTS

I would like to thank D. R. Hamann for many helpful discussions. I am also thankful to W. H. Haemmerle and T. Lewandowski for technical assistance.

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[9] P. Fulde and K. Maki, Phys. Rev. Letters **15**, 675 (1965).

[10] P. Fulde and K. Maki, Phys. Condensed Matter **5**, 380 (1966).

[11] P. G. de Gennes and S. Mauro, Solid State Commun. **4**, 385 (1966).

[12] J. J. Hauser, Phys. Rev. **164**, 558 (1967).

[13] The value of $C(t_c)=2$ corresponds to $\frac{3}{4}$ of the experimentally determined slope of the relative conductance versus t at T_{c0} for the pure In film.