PHYSICAL REVIEW VOLUME 150, NUMBER 1 7 OCTOBER 1966

Critical Magnetic Field of Thin Superposed Films

RUDOLF KLEIN AND GASTON FISCHER
RCA Laboratories Ltd., Zürich, Switzerland
(Received 11 May 1966)

Thin superposed films of two superconductors or of a superconductor and a normal metal are studied in a parallel magnetic field near a second-order transition. From the linearized integral equation for the position-dependent order parameter and the boundary conditions for the magnetic-field-dependent kernel at the interface, an implicit equation for the critical magnetic field is derived under the assumption that the order parameter has a different but constant value in each film. Near the transition temperature of the double film T_{cns} and at the absolute zero of temperature, the results are expressed in terms of the usual relevant parameters characterizing the two constituents of the compound film, and are compared with some recent experimental data.

I. INTRODUCTION

VARIOUS theories for the transition temperature of superposed films have been proposed. The formulas found by de Gennes,[1] Werthamer,[2] and Silvert and Cooper[3] can explain the experimental observations quite well. Recently, proximity effect studies in a parallel magnetic field[4] have also been made, and de Gennes and Hurault[5] were able to give a theoretical interpretation for the observed "breakdown" fields above which a normal layer appears at the "normal metal" surface of the sandwich.

In this paper the critical magnetic field is calculated for a sandwich consisting of two adjacent thin films of "dirty" material. The results are discussed in connection with experiments on the transition temperature T_c and critical fields H_c for very thin aluminum and tungsten films. The large values for T_c and H_c found by Kammerer *et al.*[6] have been interpreted as evidence for Ginzburg surface superconductivity.[7] Kammerer *et al.* have interpreted their results for T_c in terms of de Gennes's formula in the thin-film limit. We show that a generalization of the thin-film model to include the magnetic field can also explain a raise of the critical field H_c above the value obtaining for a homogeneous thin film.

II. DERIVATION OF THE CRITICAL MAGNETIC FIELD

We start from the linearized integral equation for the position-dependent order parameter in the vicinity of a second-order phase transition:

$$\Delta(\mathbf{r})=\frac{V(\mathbf{r})}{\beta}\sum_{\omega}\int d^3r'K(\mathbf{r},\mathbf{r}',\omega)\Delta(\mathbf{r}'),\tag{1}$$

where $V(\mathbf{r})$ is the effective interaction potential, $\beta=(kT)^{-1}$, and $\omega=\pi(2n+1)/\beta$, $n=0, \pm1, \pm2,\cdots$. The integral kernel $K(\mathbf{r},\mathbf{r}',\omega)$ is given by the product of two Green's functions for electrons in the normal phase of the metal,

$$K(\mathbf{r},\mathbf{r}',\omega)=G(\mathbf{r},\mathbf{r}',\omega)G(\mathbf{r},\mathbf{r}',-\omega).\tag{2}$$

$G(\mathbf{r},\mathbf{r}',\omega)$ is a Fourier component of the imaginary-time Green's function in the presence of impurities and magnetic field. These functions have been calculated by Abrikosov and Gorkov,[8] who have discussed the impurity-averaging procedure and have shown that to a good approximation the magnetic field enters the expression for the Green's function only via a phase factor. Without magnetic field the impurity averaged function is

$$\bar{G}(\mathbf{r},\mathbf{r}',\omega)=G_0(\mathbf{r},\mathbf{r}',\omega)\exp[-|\mathbf{r}-\mathbf{r}'|/2l],\tag{3}$$

where

$$G_0(\mathbf{r},\mathbf{r}',\omega)=-\frac{m}{2\pi\hbar^2|\mathbf{r}-\mathbf{r}'|}\exp\left[\left(i\frac{\omega k_F}{|\omega|}-\frac{|\omega|}{\hbar v_F}\right)|\mathbf{r}-\mathbf{r}'|\right]\tag{4}$$

is the Green's function for the pure metal, and l is the mean free path of the electrons. If we take a z axis in the direction of the magnetic field H and an x axis perpendicular to the films we can choose a vector potential with components $(0,A(x),0)$ and the appropriate phase factor entering Eq. (3) is $\exp(ieA(y-y')/\hbar c)$.

Since we are interested in dirty films we have to use the impurity averaged Eq. (1). The form of this equation is preserved, but the new kernel $\bar{K}(\mathbf{r},\mathbf{r}',\omega)$ is not the product of two functions $\bar{G}$ as in Eq. (2). Abrikosov and

---

[1] P. G. de Gennes, Rev. Mod. Phys. **36**, 225 (1964); P. G. de Gennes and E. Guyon, Phys. Letters **3**, 168 (1963).

[2] N. R. Werthamer, Phys. Rev. **132**, 2440 (1963).

[3] W. Silvert and L. N. Cooper, Phys. Rev. **141**, 336 (1966).

[4] J. P. Burger, G. Deutscher, E. Guyon, and A. Martinet, Phys. Letters **17**, 180 (1965); G. Fischer, R. Klein, and J. P. McEvoy, *ibid.* **19**, 193 (1965).

[5] P. G. de Gennes and J. P. Hurault, Phys. Letters **17**, 181 (1965).

[6] M. Strongin, A. Paskin, O. F. Kammerer, and M. Garber, Phys. Rev. Letters **14**, 362 (1965); M. Strongin, O. F. Kammerer, and A. Paskin, *ibid.* **14**, 949 (1965); O. F. Kammerer and M. Strongin, Phys. Letters **17**, 224 (1965).

[7] V. L. Ginzburg, Phys. Letters **13**, 101 (1964).

[8] See A. A. Abrikosov, L. P. Gorkov, and I. E. Dzyaloshinski, *Methods of Quantum Field Theory in Statistical Physics* (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1963).

Gorkov have shown that $\bar{K}$ satisfies the integral equation

$$\bar{K}(\mathbf{r},\mathbf{r}',\omega)=K_0(\mathbf{r},\mathbf{r}',\omega)+\frac{\hbar}{2\pi\tau N(0)}\int d^3r_1K_0(\mathbf{r},\mathbf{r}_1,\omega)\bar{K}(\mathbf{r}_1,\mathbf{r}',\omega)\,. \quad (5)$$

Here the impurity potential has been taken as a δ function, τ is a relaxation time, N (0) is the density of states at the Fermi level and

$$K_0(\mathbf{r},\mathbf{r}',\omega)=\bar{G}(\mathbf{r},\mathbf{r}',\omega)\bar{G}(\mathbf{r},\mathbf{r}',-\omega)\,. \quad (6)$$

We want to derive results for sandwiches consisting of very thin N and S films, for which the respective coherence lengths ξ_n and ξ_s are smaller than the thicknesses d_n and d_s. Then, following de Gennes, we treat $\bar{K}(\mathbf{r},\mathbf{r}',\omega)$ as a constant when $\mathbf{r}$ and $\mathbf{r}'$ lie in the same film. The order parameter is also assumed to be constant, having values Δ_n or Δ_s in the N or S film. If V_n, V_s are the interaction constants, we have from Eq. (1)

$$\begin{aligned}\Delta_n&=\frac{V_n\Delta_n}{\beta}\sum_\omega\int_n d^3r'\bar{K}(r,r',\omega)+\frac{V_n\Delta_s}{\beta}\sum_\omega\int_s d^3r'\bar{K}(r,r',\omega)\,,\\ \Delta_s&=\frac{V_s\Delta_n}{\beta}\sum_\omega\int_n d^3r'\bar{K}(r,r',\omega)+\frac{V_s\Delta_s}{\beta}\sum_\omega\int_s d^3r'\bar{K}(r,r',\omega)\,.\end{aligned} \quad (7)$$

For the integrals over $\bar{K}$ we find from (5)

$$\int\bar{K}(r,r',\omega)d^3r'=\int K_0(r,r',\omega)d^3r'+\frac{\hbar}{2\pi\tau N(0)}\int d^3r_1\int d^3r'K_0(r,r_1,\omega)\bar{K}(r_1,r',\omega)\,.$$

In the last double integral $K_0(r,r_1,\omega)$ is only large for $|r-r_1|<l$. Therefore, we expand $\int\bar{K}(r_1,r',\omega)d^3r'$ around r, which gives

$$\int\bar{K}(r,r',\omega)d^3r'=\frac{\left(\int K_0(r,r',\omega)d^3r'\right)}{\left(1-\frac{\hbar}{2\pi\tau N(0)}\int K_0(r,r',\omega)d^3r'\right)}\,. \quad (8)$$

Now the integral over K_0 is easily evaluated from Eq. (6) and the definition of $\bar{G}$:

$$\int K_0(r,r',\omega)d^3r'=\frac{N_r(0)\pi}{(eA/c)v_F}\arctan\frac{(2eA/c)v_F}{2|\omega|(1+\hbar/2|\omega|\tau)}\,, \quad (9)$$

where we take $N_r(0)$ to be the (constant) density of states at the Fermi surface of that particular film in which the point r is located.

Then Eq. (8) gives approximately

$$\int\bar{K}(r,r',\omega)d^3r'=2N_r(0)/(2|\omega|+D_rq^2)\,, \quad (10)$$

where $D_r=\frac{1}{3}(v_Fl)_r$ is the diffusion coefficient in N or S, and $q^2=\hbar(2eA/\hbar c)^2$. If $\bar{K}$ can be treated as one constant in N and another constant in S, Eq. (10) leads to

$$\begin{aligned}\bar{K}(N,N,\omega)d_n+\bar{K}(N,S,\omega)d_s&=2N_n/(2|\omega|+D_nq^2)\,,\\ \bar{K}(S,N,\omega)d_n+\bar{K}(S,S,\omega)d_s&=2N_s/(2|\omega|+D_sq^2)\,.\end{aligned} \quad (11)$$

The boundary conditions at the metallic interfaces are[1]

$$\frac{\bar{K}(N,N,\omega)}{N_n}=\frac{\bar{K}(N,S,\omega)}{N_s};\quad\frac{\bar{K}(S,N,\omega)}{N_n}=\frac{\bar{K}(S,S,\omega)}{N_s}\,. \quad (12)$$

Inserting (11) and (12) into (7) gives a linear homogeneous system of equations for Δ_n and Δ_s, where the coefficients depend on N_n, N_s, D_n, D_s, V_s, V_s, d_n, d_s. Requiring the determinant of these coefficients to vanish yields an implicit equation for H_c:

$$1=\frac{2\pi N_nV_n}{\beta}\left\langle\sum_\omega\frac{1}{2|\omega|+D_nq^2}\right\rangle_n\frac{N_nd_n}{N_nd_n+N_sd_s}+\frac{2\pi N_sV_s}{\beta}\left\langle\sum_\omega\frac{1}{2|\omega|+D_sq^2}\right\rangle_s\frac{N_sd_s}{N_nd_n+N_sd_s}\,. \quad (13)$$

Here the angular brackets mean an average over the N film in the first term, and over the S film in the second term.

With the expression for the transition temperature T_{cns} of the NS sandwich, derived by de Gennes in the Cooper limit

$$\ln\frac{1.14\hbar\tilde{\omega}}{kT_{cns}}=\frac{N_nd_n+N_sd_s}{N_n{}^2V_nd_n+N_s{}^2V_sd_s}\,, \quad (14)$$

where $\tilde{\omega}$ is a BCS cutoff of the interaction V, we find

$$\frac{N_n{}^2V_nd_n+N_s{}^2V_sd_s}{N_nV_nN_sV_s}\ln\frac{T_{cns}}{T}=\frac{N_nd_n}{N_sV_s}\left\langle\psi\left(\frac{1}{2}+\frac{D_nq^2}{4\pi kT}\right)-\psi(\tfrac{1}{2})\right\rangle_n+\frac{N_sd_s}{N_nV_n}\left\langle\psi\left(\frac{1}{2}+\frac{D_sq^2}{4\pi kT}\right)-\psi(\tfrac{1}{2})\right\rangle_s\,, \quad (15)$$

where $\psi(z)=\Gamma'(z)/\Gamma(z)$.

III. DISCUSSION

A. High Temperatures

From the general formula (15) one can easily find the critical magnetic field near the transition temperature T_{cns}, and also at $T=0$, of the NS sandwich. At intermediate values of T, one has to use numerical methods. Just below T_{cns} where H_c is small one can expand the function ψ:

$$\psi(\tfrac{1}{2}+z)-\psi(\tfrac{1}{2})\simeq z\psi'(\tfrac{1}{2})=\tfrac{1}{2}\pi^2 z.$$

The averaging procedure in this case has to be done over x^2, because $z\sim q^2\sim A^2=H^2x^2$. Since the vector potential is antisymmetric with respect to the geometrical center plane of the sandwich, the angular brackets in Eq. (15) are proportional to the integrals

$$\frac{1}{d_n}\int_{-\frac{1}{2}(d_n+d_s)}^{\frac{1}{2}(d_n-d_s)} x^2dx \quad\text{and}\quad \frac{1}{d_s}\int_{\frac{1}{2}(d_n-d_s)}^{\frac{1}{2}(d_n+d_s)} x^2dx, \tag{16}$$

respectively. In this way we find, for $T_{cns}-T\equiv\Delta T\ll T_{cns}$,

$$H_c(T)=\frac{6\hbar c}{e}\left(\frac{2k\Delta T}{\pi\hbar}\right)^{1/2}\left\{\frac{1}{d_n^2}\,\frac{1+(N_s^2V_s/N_n^2V_n)(d_s/d_n)}{(v_Fl)_n[1+3(d_s^2/d_n^2)]+(N_s^2V_s/N_n^2V_n)(v_Fl)_s[3(d_s/d_n)+(d_s^3/d_n^3)]}\right\}^{1/2}. \tag{17}$$

If $(v_Fl)_n=(v_Fl)_s=v_Fl$; $N_s=N_n$; $V_s=V_n$, $d_s+d_n=d$, this formula reduces to the Ginzburg-Landau result[9] for a homogeneous film of thickness d and material with a bulk transition temperature T_c.

Kammerer *et al.*[6] interpreted their measurements of the high transition temperatures of various thin films as evidence for Ginzburg surface superconductivity. A very thin Ginzburg surface layer of thickness d_G, with an enhanced effective interaction V_G due to the presence of an oxide layer, is in contact with the normal film under it. This superimposed layer problem is then analyzed in terms of de Gennes' formula (14) for T_{cns} under the assumption that the densities of states and mean free paths are equal in the surface and normal film regions. Following these assumptions we obtain from Eq. (17)

$$H_c(T)=\frac{6\hbar c}{e}\left(\frac{2k(T_{cns}-T)}{\pi\hbar v_Fl}\right)^{1/2}\left\{\frac{1}{d_n^2}\,\frac{1+(V_s/V_n)(d_s/d_n)}{1+3(d_s/d_n)^2+(V_s/V_n)[3(d_s/d_n)+(d_s^3/d_n^3)]}\right\}^{1/2}, \tag{18}$$

or, setting $d=d_n+d_s$, $d_n=\alpha d$, and $V_s/V_n=\mu$

$$H_c(T)=\frac{6\hbar c}{ed}\left(\frac{2k(T_{cns}-T)}{\pi\hbar v_Fl}\right)^{1/2}f^{1/2}(\alpha,\mu), \tag{19}$$

where

$$f(\alpha,\mu)=\frac{1}{\alpha^2}\,\frac{1+\mu(1-\alpha)/\alpha}{1+3((1-\alpha)/\alpha)^2+\mu[((1-\alpha)/\alpha)^3+3(1-\alpha)/\alpha]}. \tag{20}$$

Equation (19) differs from the Ginzburg-Landau result only through the appearance of T_{cns} instead of T_c and the function $f(\alpha,\mu)$ which is equal to unity for a homogeneous film. To explain the observed T_c, Kammerer *et al.* assume a thickness of $d_s=20$ Å for the Ginzburg surface region, and this then requires a ratio $\mu=1.84$. Since the most dramatic effects are found for films of small total thickness d, we assume the variable α to have a value of $\frac{3}{4}$ or slightly larger. In this range for α and $1<\mu<2$ the function $f(\alpha,\mu)$ is only slightly smaller than unity, which means that near the transition temperature a replacement of T_c by T_{cns} in the Ginzburg-Landau formula is a very good approximation. (For the special case $d_n=d_s=\frac{1}{2}d$ the function $f(\frac{1}{2},\mu)=1$ for all μ, and the presence of a layered structure rather than a homogeneous film is only reflected in $T_{cns}=T_{cns}(\mu)$ replacing T_c.)

B. Low Temperatures

Near the absolute zero of temperature, Eq. (15) can be simplified with an asymptotic expansion:

$$\psi(\tfrac{1}{2}+z)-\psi(\tfrac{1}{2})\rightarrow\ln(4\gamma z),$$

where $\ln\gamma$ is Euler's constant.

Introducing the parameter

$$\mu'=N_s^2V_s/N_n^2V_n=(N_s^2/N_n^2)\mu, \tag{21}$$

Eq. (15) can be written as

$$\left(1+\mu'\frac{1-\alpha}{\alpha}\right)\ln\frac{T_{cns}}{T}=\left\langle\ln\frac{\gamma D_nq^2}{\pi kT}\right\rangle_n+\mu'\frac{1-\alpha}{\alpha}\left\langle\ln\frac{\gamma D_sq^2}{\pi kT}\right\rangle_s. \tag{22}$$

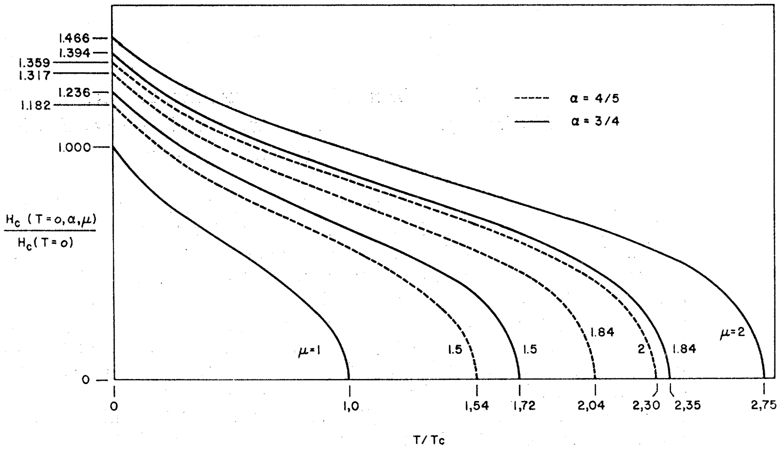

FIG. 1. Sketch of $H_c(T,\alpha,\mu)$ versus T for several sets of the parameters α and μ. The curve for $\mu=1$ is a calculated curve from Rickayzen (Ref. 9), the other curves are sketched by analogy, the only calculated points being on the abscissa and ordinate.

Here, T_{cns} is a function of α, μ, and μ':

$$\ln\frac{1.14\hbar\tilde{\omega}}{kT_{cns}}=\frac{1}{N_nV_n}\,\frac{1+[(1-\alpha)/\alpha](\mu'/\mu)^{1/2}}{1+\mu'(1-\alpha)/\alpha}. \tag{23}$$

Carrying out the integration we obtain

$$H_c(T=0,\alpha,\mu,\mu')=\frac{\hbar c}{ed}(\exp 1)\left(\frac{3\pi kT_{cns}}{\hbar\gamma}\right)^{1/2}\times\left[\frac{|2\alpha-1|^{(2\alpha-1)(\mu'-1)}}{(v_Fl)_n{}^{\alpha}(v_Fl)_s{}^{\mu'(1-\alpha)}}\right]^{1/2(\alpha+\mu'(1-\alpha))}. \tag{24}$$

For a homogeneous film ($\mu'=\mu=1$) we have $T_{cns}=T_c$ and $(v_Fl)_n=(v_Fl)_s=v_Fl$, which leads to Rickayzen's result[9] for the critical magnetic field at $T=0$ of a film of thickness d,

$$H_c(0)=\frac{\hbar c}{ed}(\exp 1)\left(\frac{3\pi kT_c}{\hbar\gamma v_Fl}\right)^{1/2}. \tag{25}$$

Going back to the special case of the Ginzburg surface layer we obtain from (24), (25), and (14)

$$\frac{H_c(T=0,\alpha,\mu)}{H_c(0)}=\left[\left(\frac{1.14\hbar\tilde{\omega}}{kT_c}\right)^{1-\alpha}|2\alpha-1|^{2\alpha-1}\right]^{(\mu-1)/2(\alpha+\mu(1-\alpha))}. \tag{26}$$

Inserting values that pertain to Al for a film[6] of total thickness $d=100$ Å, a surface layer of thickness 20 Å, and $\mu=1.84$, Eq. (26) raises the critical magnetic field at $T=0$ by 30% above the value which one obtains from Eq. (25) for a homogeneous film. This latter value is ≈ 15 kOe, assuming $l=100$ Å, $T_c(\text{Al})=1.18$°K, and $v_F=1.2\times10^8$ cm/sec, so that the critical magnetic field of the layered structure should be 19.5 kOe, which is in reasonable agreement with the experimental result (20–25 kOe at ~ 1°K).

In Fig. 1 we show the expected behavior of $H_c(T)$ in the special case $(v_Fl)_n=(v_Fl)_s$ and $N_n=N_s$, for different values of α and μ.

IV. CONCLUSIONS

We have calculated the critical field $H_c(T)$ of superposed thin films in the Cooper limit. At present the only available experimental data with which our theory can be compared are concerned with superposed films of the same metal, whereby the effective electron-electron interaction in one of the films is assumed to have been enhanced by an adjacent dielectric layer. But our theory also applies to superposed films of two different metals. The interpretation of $H_c(T)$ data for such films is likely to yield more information about the effective electron-electron interaction than a measurement of T_{cns} alone.

---

[9] G. Rickayzen, Phys. Rev. **138**, A73 (1965).