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THE PROXIMITY EFFECTS OF SUPERCONDUCTING MULTILAYER FILM

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#### Abstract

This paper considers the proximity effects of superconducting multilayer films composed of different metals. Using the Silvert and Cooper method, the relationship between the critical temperature of a superconducting multilayer film with strong heterogeneity and its geometric structure is given under the Cooper limit.

By using modern material preparation techniques such as sputtering, evaporation, epitaxy etc., based on some peoples' designs of atomic scales, we synthesized various previously non-existent new types of materials and studied their structures and properties. This has been an important trend in solid physics in recent years. For example, besides semiconductor materials, there have also been reports recently of making new materials with "multilayered ultrathin common lattice structures" (LUCS) from niobium and copper possessing different properties [1].

When compared with large pieces of homogeneous specimens, multilayered thin film systems have two new factors which should be considered. One is the appearance of many hetergeneous interfaces. See Reference [2] for an analytical discussion of them. The second factor is the proximity effects caused by the many hetergeneous layers being periodically adjacent to eachother and the superconducting electron pairs possessing nonlocalization. This paper makes a preliminary investigation of this problem. We began from a simplified model under hetergeneous interface conditions, used the GLAG theory and gave the general relationship between the multilayered film's critical temperature and geometric factors.

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We conceived that the multilayered film was composed from two types of pure superconducting materials,  $S_1$  and  $S_2$ , and their thicknesses were separately  $d_1$  and  $d_2$ . In order to simplify the problem and outstanding focal points, we assumed that the property difference concentrations of the two types of materials were reflected on their electro-acoustic coupling constant (NV), separately (NV)<sub>1</sub> and (NV)<sub>2</sub>. The other properties were the same.

We used the Silvert and Cooper method to calculate the proximity effects [3].

The virtual time, Fourier transformation, Green's function and abnormal Green's function of the superconductor satisfy the following formulas in the  $T_c$  area:

$$G_{\omega}(\mathbf{r}, \mathbf{r}') = G_{\omega}^{(0)}(\mathbf{r} - \mathbf{r}') - \int d\mathbf{r}_{1} d\mathbf{r}_{2} G_{\omega}^{(0)}(\mathbf{r}, \mathbf{r}_{1}) \Delta(\mathbf{r}_{1}) G_{-\omega}^{(0)}(\mathbf{r}_{2}, \mathbf{r}_{1}) \Delta^{*}(\mathbf{r}_{2}) G_{\omega}(\mathbf{r}_{2}, \mathbf{r}'), \qquad (1) F_{\omega}(\mathbf{r}, \mathbf{r}') = \int d\mathbf{r}_{1} G_{\omega}(\mathbf{r}, \mathbf{r}_{1}) \Delta(\mathbf{r}_{1}) G_{-\omega}^{(0)}(\mathbf{r}_{1}, \mathbf{r}'), \qquad (2)$$

In the formula,  $G_{\omega}$  and  $F_{\omega}$  are separately the completely normal and abnormal functions,  $G_{\omega}^{(0)}$  is the zero order Green function and  $\Delta$  (r) is the energy gap function which changes with the space. They should satisfy the following self-consistent conditions:

$$\Delta(\mathbf{r}) = V(\mathbf{r}) \cdot F(\mathbf{r}, \mathbf{r}), \qquad (3)$$

In the formula  $F(r,r) = k_{B}T \sum_{r} F_{r}(r,r)$  and  $k_{B}$  is the Boltzmann

constant. In the T<sub>c</sub> vicinity,  $\Delta$ (r) is a small quantity and G<sub> $\omega$ </sub> and F<sub> $\omega$ </sub> rely on the expansion in powers of  $\Delta$ :

$$G_{\bullet}(\mathbf{r},\mathbf{r}') = \sum_{n=0}^{\bullet} G_{\bullet}^{(n)}(\mathbf{r},\mathbf{r}'), \qquad (4)$$

$$G_{\bullet}^{(m)}(\mathbf{r},\mathbf{r}') = -\int d\mathbf{r}_1 d\mathbf{r}_2 G_{\bullet}^{(0)}(\mathbf{r},\mathbf{r}_1) \Delta(\mathbf{r}_1) G_{\bullet\bullet}^{(0)}(\mathbf{r}_2,\mathbf{r}_1) \cdot \Delta(\mathbf{r}_2) G_{\bullet}^{(m-1)}(\mathbf{r}_2,\mathbf{r}'), \quad (5)$$

$$Y_{\bullet}(\boldsymbol{r},\boldsymbol{r}') = \sum_{m=1}^{\bullet} F_{\bullet}^{(m)}(\boldsymbol{r},\boldsymbol{r}'), \qquad (6)$$

$$F_{\sigma}^{(m)}(\mathbf{r}, \mathbf{r}') = \int d\mathbf{r}_1 G_{\sigma}^{(m-1)}(\mathbf{r}_1, \mathbf{r}) \Delta(\mathbf{r}_1) G_{-\sigma}^{(0)}(\mathbf{r}', \mathbf{r}).$$
(7)

By separately substituting formulas (5) and (7) into formulas (4) and (6), we can test and verify this.

We can see from the above expansion formulas that  $F_{\omega}^{(m)}$  is the 2m-1 times homogeneous function of  $\Delta$ . In later calculations, we only keep the first order terms of  $\Delta$  and in this way formula (3) changes to

$$\Delta(\mathbf{r}) = V(\mathbf{r}) k_{\rm B} T \sum_{\alpha} \int d\mathbf{r}_1 G_{\alpha}^{(0)}(\mathbf{r} - \mathbf{r}_1) \Delta(\mathbf{r}_1) G_{-\alpha}^{(0)}(\mathbf{r} - \mathbf{r}_1), \qquad (8)$$

In the formula

$$u = (2n + 1)\pi k_B T$$
  $n = 0, \pm 1, \pm 2, \cdots$ 

The zero order Green function is known

$$G_{-}^{\omega} = \left(-\frac{m}{2\pi|\mathbf{r}-\mathbf{r}'|}\right) \exp\left[ik|\mathbf{r}-\mathbf{r}'|\operatorname{sgn}\omega - \frac{|\omega'||\mathbf{r}-\mathbf{r}'|}{v_1}\right].$$
(9)

We substitute formula (9) into formula (8)

$$\Delta(\mathbf{r}) = V(\mathbf{r}) \int d\mathbf{r}_1 \Gamma^{(0)}(\mathbf{r} - \mathbf{r}_1) \Delta(\mathbf{r}_1), \qquad (10)$$

In the formula

$$\Gamma^{(0)} = k_0 T \sum_{\alpha} \left( \frac{m}{2\pi |\mathbf{r} - \mathbf{r}'|} \right)^2 \exp\left[ -\frac{2|\omega| \cdot |\mathbf{r} - \mathbf{r}'|}{\nu} \right]. \tag{11}$$

By assuming each layer of the multilayered film is parallel to the xy plane, we can first carry out metric integration of formula (1) and obtain

$$\Delta(z) = V(z)N_z \frac{1}{\nu_f} k_b T \sum_{\mu} \int \Delta(z') E_1 \left( \frac{2\omega(z-z')}{\nu_f} \right) dz', \qquad (12)$$

In the formula,  $N_z$  is the fixed region density of the z area and E, is the improper index integral, defined as

$$E_{m}(u) = \int_{1}^{u} \frac{dt}{t^{n}} \exp[-ut].$$
(13)

For convenience of calculations, we make the following stipulations: each layer's coordinates of  $S_1$  are kd  $\langle z \langle (k+a)d;$ each layer's coordinates of  $S_2$  are  $(k+a)d \langle z \langle (k+1)d, k=0, \pm 1, \pm 2...; d=d_1+d_2; a=d_1/d; \mu = (VN)_2/(VN)_1$ .

Each layer of superconducting multilayer films is relatively thin. When we consider the so-called Cooper limit, that is, the thickness of each layer is smaller than the coherent length of a corresponding material, at this time, the energy gap function of each level is approximately a constant, separately taken as  $\Delta_1$  and  $\Delta_2$ . Further, disregarding the macroscopic boundary of the superconductor, we consider the layer number of multilayer film to be infinite.

By letting  $z_1$  and  $z_2$  separately be the two points in the  $S_1$  and  $S_2$  layers and using formula (12) for the two points, we can obtain two linear unrelated equations:

$$\Delta_{1} = (NV)_{1}f_{11}\Delta_{1} + (NV)_{1}f_{12}\Delta_{2}, \qquad (14)$$

$$\Delta_2 = \mu(NV) f_{\rm R} \Delta_1 + \mu(NV) f_{\rm R} \Delta_2, \tag{15}$$

In the formula,  $f_{ij}$  is determined by the precise positions of  $z_1$  and  $z_2$ . By taking  $z_1$  and  $z_2$  in each period k, we can obtain

these two equations. However, because of the translational symmetry of the system, if there is only the equations between  $z_1$  and  $z_2$ , then they are linear and unrelated and do not lose generality. Taking  $z_1$  and  $z_2$  in the period of k=0,  $f_{ij}$  can be written in the following form:

$$f_{11} = \frac{\pi k_{\rm B}T}{\nu_{\rm f}} \sum_{\alpha} \sum_{k} \int_{kd}^{(k+\alpha)d} E_1 \left(\frac{|z_1 - z'|}{l_{\alpha}}\right) dz',$$

$$f_{12} = \frac{\pi k_{\rm B}T}{\nu_{\rm f}} \sum_{\alpha} \sum_{k} \int_{(k+\alpha)d}^{(k+1)d} E_1 \left(\frac{|z_1 - z'|}{l_{\alpha}}\right) dz',$$

$$f_{21} = \frac{\pi k_{\rm B}T}{\nu_{\rm f}} \sum_{\alpha} \sum_{k} \int_{kd}^{(k+\alpha)d} E_1 \left(\frac{|z_2 - z'|}{l_{\alpha}}\right) dz',$$

$$f_{22} = \frac{\pi k_{\rm B}T}{\nu_{\rm f}} \sum_{\alpha} \sum_{k} \int_{(k+\alpha)d}^{(k+1)d} E_1 \left(\frac{|z_2 - z'|}{l_{\alpha}}\right) dz',$$

$$l_{\alpha} = \nu_{\rm f}/2 |\omega|.$$
(16)

The set of equations (14) and (15) have non-zero solution conditions and the coefficient determinate is zero:

$$[(VN)_{f_{11}} - 1] \cdot [\mu(VN)_{f_{22}} - 1] = \mu(VN)_{f_{11}} \cdot f_{12}.$$
(17)

We can find the relationship of  $T_c$  and d, a,  $\mu$  and  $(VN)_1$  from formula (17). Under common conditions, we can only use the numerical solution method to solve formula (17) yet under strong heterogeneous conditions wherein  $\mu \rightarrow 0$ , we can obtain an approximate analytical result. At this time, formula (17) changes to

$$u = \frac{1}{(NV)_1}.$$
 (18)

After completing the integration of z'

$$f_{11} = (\pi k_B T) \sum_{\alpha} \frac{1}{2|\alpha|} \left\{ \sum_{k=-1}^{\infty} \int_{1}^{\infty} \frac{dt}{t^2} \exp\left[-\frac{x_1 t}{l_{\alpha}}\right] \times \left[ \exp\left(\frac{k+\alpha}{l_{\alpha}} dt\right) - \exp\left(\frac{k}{l_{\alpha}}\right) \right] + \sum_{k=1}^{\infty} \int_{1}^{\infty} \frac{dt}{t^2} \exp\left[\frac{x_1 t}{l_{\alpha}}\right] \times \left[ \exp\left(-\frac{dk t}{l_{\alpha}}\right) - \exp\left(-\frac{k+\alpha}{l_{\alpha}} dt\right) \right] + \int_{1}^{\infty} \frac{dt}{t^2} \left[ 1 - \exp\left(-\frac{\alpha d - x_1}{l_{\alpha}} t\right) \right] \right\}. (19)$$

Under the sign of integration, the summation of k is

$$f_{11} = (\mathbf{x} t_B T) \sum_{\sigma} \frac{1}{|\sigma|} \left\{ 1 + \frac{1}{2} \int_{1}^{\sigma} \frac{dt}{t^2} \frac{1 - \exp\left(-\frac{\sigma dt}{l_{\sigma}}\right)}{1 - \exp\left(-\frac{dt}{l_{\sigma}}\right)} \right\}$$
$$\times \exp\left(-\frac{dt}{l_{\sigma}}\right) \left[ \exp\left(\frac{\mathbf{x}_1 t}{l_{\sigma}}\right) + \exp\left(\frac{\sigma d - \mathbf{x}_1}{l_{\sigma}}t\right) \right]$$
$$- \frac{1}{2} \int_{1}^{\sigma} \frac{dt}{t^2} \left[ \exp\left(-\frac{\mathbf{x}_1 t}{l_{\sigma}}\right) + \exp\left(-\frac{\sigma d - \mathbf{x}_1}{l_{\sigma}}t\right) \right], \quad (20)$$

We then find the mean of  $z_1$  in [0,ad]

$$\langle f_{11} \rangle = (\pi t_B T) \sum_{\omega} \frac{1}{|\omega|} \left\{ 1 - \frac{l_{\omega}}{\sigma d} \int_{1}^{\omega} \frac{dt}{t^3} \times \frac{\left[ 1 - \exp\left(-\frac{\sigma dt}{l_{\omega}}\right) \right] \left[ 1 - \exp\left(-\frac{1 - \sigma}{l_{\omega}} dt\right) \right]}{1 - \exp\left(-\frac{dt}{l_{\omega}}\right)} \right\}.$$
 (21)

We will first analyze several types of limit situations below.

When a=1, that is, a homogeneous superconductor wherein the entire multilayer film is  $S_1$  material, the second term in the curly brackets in formula (21) is zero and formula (21) changes to

$$\langle f_{11} \rangle = (\pi k_B T) \sum_{\alpha} \frac{1}{|\alpha|}.$$
 (22)

Based on de Gennes' transformation [4]:

$$(\pi k_{\rm B} T) \sum_{\sigma} \frac{1}{|\omega|} \rightarrow \ln \frac{1.14 \,\omega_{\rm D}}{T_{\rm c}}.$$
 (23)

Formula (18) then changes into the  $T_{c}$  formula of the BCS theory

$$T_c = 1.14\omega_{\rm D} \exp\left(-\frac{1}{NV}\right),\tag{24}$$

In the formula,  $\omega_{\rm D}$  is the Debye frequency.

When a=0, the entire multilayered film is normal metal  $S_2$ , the term within the curly brackets of formula (21) is zero and from formulas (14) and (15),  $\Delta_1 = \Delta_2 = 0$  which is the expected result.

When each layer is very thin and  $d \rightarrow 0$ , the system approaches being a homogeneous system, the factor in the curly brackets of formula (21)  $\rightarrow$  a and the T<sub>c</sub> formula changes to

$$T_c = 1.14\omega_{\rm D} \exp\left(-\frac{1}{\alpha NV}\right). \tag{25}$$

This shows that the system is equivalent to a homogeneous superconductor with an equivalent coupling constant of (aNV).

Under common conditions, by introducing a parameter  $l \omega_0$ , formula (21) changes to

$$(\mathbf{x}_{\mathsf{h}}T)\sum_{\boldsymbol{\nu}}\frac{1}{|\boldsymbol{\omega}|}\{1-\boldsymbol{\gamma}\}=\langle f_{\mathsf{H}}\rangle, \qquad (26)$$

$$r = \frac{l_{a0}}{ad} \int_{1}^{a} \frac{dt}{t^{3}} \frac{\left[1 - \exp\left(-\frac{adt}{l_{a0}}\right)\right] \left[1 - \exp\left(-\frac{1 - a}{l_{a0}}dt\right)\right]}{1 - \exp\left(-\frac{dt}{l_{a0}}\right)}, \quad (27)$$

In the formulas, the selection of  $1_{\omega 0}$  should cause formula (26) to be equal to formula (21). In this way, the  $T_{c}$  formula can be written as

$$T_{e} = 1.14\omega_{\rm D} \exp\left[-\frac{1}{(NV)_{\rm b}(1-\gamma)}\right].$$
 (25)

By dividing formula (28) with  $T_0 = 1.14 \omega_D \exp \left[-\frac{1}{(NV)}\right]$ , we obtain

$$\frac{T_{f}}{T_{0}} = \exp\left[-\frac{1}{(NV)_{1}} \cdot \frac{\gamma}{1-\gamma}\right]. \quad (29)$$

Using copper and niobium multilayered film as examples, Fig. 1 shows the relationship of  $T_c/T_o \sim a$  and  $x(x=d/l_{\omega 0})$  as well as the (NV)  $\approx 0.3$  of the niobium.

When the curves in Fig. 1 are in  $x \rightarrow \infty$ , they all tend to approach 1. The curve corresponding to a=0.2 crosses with the longitudinal axis at 0.04 but this is not drawn in the figure.

Fig. 1

In the above calculations we did not consider the special effects of the heterogeneous interfaces and therefore the results are basically the same as those of the common and simple S-N proximity effects. The  $T_c$  value of the entire multilayer film system is between the large material  $T_c$  values of the two types of metal. Therefore, if we find in the tests that the  $T_c$  value of the superconducting multilayer film has certain differences, for example, higher or lower than the large material  $T_c$  of each constituent element, then this is direct evidence of the influence of heterogeneous interfaces on the structures of the electrons.

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