## Electron-Electron Interaction Parameter in Gold Films from the Proximity Effect\*

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McMillan's tunneling theory of the proximity effect has been applied to results on goldaluminum-oxide-aluminum thin-film sandwiches. Measurements of the transition temperature and minimum energy gap give consistent agreement with the theory and indicate that the interaction parameter in gold is  $0.072 \times 0.004$ .

We have used the McMillan tunneling theory of the proximity effect<sup>1</sup> to analyze the results of measurements on superimposed normal and superconducting films separated by a thin barrier. The system reported here is gold(1800 Å)-alumina(4-12 Å)-aluminum(1200 Å). The alumina provides a weak barrier which constrains the electrons to remain in one film a comparatively long time before crossing over into the other film. This localization leads to unequal amplitudes of the electronic wave functions in the two films, with the difference depending on electron energy and barrier penetration probability. If, in addition, the films are thinner than their coherence lengths, the BCS potentials are constant across each. The above model simplifies the theory; McMillan was able to calculate an explicit relationship between the minimum energy gap  $\Omega_N$  and the transition temperature  $T_c$  of the sandwich. From this relationship, we can use measured values of  $\Omega_N$  and  $T_c$  to determine N(0)V in the normal film.

McMillan introduced the parameters  $\Gamma_s$  and  $\Gamma_N$ , which are related to the rate at which a single conduction electron leaves its respective film, and are therefore proportional to the transmission coefficient at the barrier. In the steady state we must have

$$N_N(0)d_N\Gamma_N = N_S(0)d_S\Gamma_S , \qquad (1)$$

where  $d_N(d_S)$  is the thickness and  $N_N(0) [N_S(0)]$ is the electron density of states per unit volume in the normal (superconducting) film. We choose  $d_N$  and  $d_S$  such that  $\Gamma_N \approx \Gamma_S$ ; and in the limit of small  $\Gamma_N$  and  $\Gamma_S$ , McMillan has obtained

$$\Omega_N = \frac{\Gamma_N}{1 + \Gamma_N / \Delta_S} \left[ 1 + \frac{\ln(2\Delta_S / \Omega_N)}{\ln(\Omega_N / \Delta_N)} \right], \tag{2}$$

where  $\Delta_N$  ( $\Delta_S$ ) refers to the energy gap in the normal (superconducting) film when it is not in proximity to another film. To obtain  $\Gamma_N$ , McMillan has solved for the transition temperature of the sandwich. For N(0)V = 0 in the normal metal and small  $\Gamma_s$  (corresponding to  $\Gamma_N' \leq 0.15$  in our experiments), he obtained  $\Gamma_s = 1.3(T_{cs} - T_c)$ , where  $T_{cs}$  refers to the superconducting film alone. Therefore a measurement of  $T_c$  and  $T_{cs}$  gives us  $\Gamma_s$  directly and we obtain  $\Gamma_N$  through Eq. (1). It has been shown<sup>2</sup> in aluminum-lead proximity specimens without a barrier that a relatively large N(0)V in the normal film does not affect  $T_c$  if  $\Gamma_s$  is small. We assume this relation is also true for our specimens with a barrier, and use it to determine  $\Gamma_N$ .

The films were vacuum deposited from an electron beam gun using 99.999%-pure aluminum and gold. It is not clear to what extent the alumina inhibits interdiffusion, but specimens were cooled to 77 K within a few hours. We measured  $d_N$  and  $d_S$  by standard quartz-crystal microbalance techniques; and since the positions of sources, crystal, and substrate were unchanged for each evaporation, we obtained a good measurement of their ratio. We used published<sup>3</sup> values of N(0) which reflect recent band calculations.<sup>4</sup>

A tunnel junction was formed between a superconducting aluminum film and the normal side of the proximity sandwich, as shown in the inset of Fig. 1. After four aluminum strips were evaporated, oxidized, and masked, three junctions were made with the same gold evaporation. Different thicknesses of alumina were evaporated on each, and the final aluminum film extended to the fourth junction so that  $\Delta_s$  and  $T_{cs}$  were measured on the same film used in the sandwiches. The dependence of  $\Gamma_{\it N}'$  (=  $\Gamma_{\it N}/\Delta_{\it S}$ ) on alumina thickness (assuming bulk density<sup>5</sup>) is also shown in Fig. 1 with the value corresponding to a transmission coefficient of 0.001 marked. All four edges of each junction were masked with a 100-Å alumina film to decrease edge effects-especially for the superimposed films. For the same reason we chose for  $T_c$  and  $T_{cS}$  the point at which the dV/dIcurves of the junctions no longer show energygap structure. Resistive transitions are usually broad and give slightly higher  $T_c$ , indicating



FIG. 1. Average thickness of the evaporated alumina barrier plotted against  $\Gamma_{N'}$ , which is proportional to the electron transmission coefficient at the barrier. Insets show film geometry. Masking leaves a  $\frac{1}{2}$ -mm  $\times \frac{1}{2}$ -mm active junction area.

some enhanced conductivity in the edges. The temperatures are determined from the vapor pressure of helium-3, and since only the difference  $T_{cS} - T_c$  is important, the absolute accuracy does not enter.

The measurement of the energy gap is not so straightforward. The best method, tunneling into a normal metal to obtain the density of states, is not possible because of thermal smearing. Our smallest  $\Omega_{N'}$  (=  $\Omega_{N}/\Delta_{s}$ ) would correspond to a 15- $\mu V$  gap if lead were used in place of aluminum in the sandwich, and we would require temperatures well below 40 mK for the necessary accuracy.<sup>2</sup> We have instead used a superconducting aluminum film and measured dV/dI at 0.35 K. These curves have sharp features at the energy gap but the interpretation is not only direct. Figure 2 demonstrates the procedure we have adopted. Tunneling theory predicts structure at  $\Delta_1 \pm \Omega_N$ , where  $\Delta_1$  is the energy gap in the aluminum cross strips. We select for  $\Delta_1$  the position midway between the peaks, and take  $\Delta_1 + \Omega_N$  to be at the intersection of the experimental curve and the curve one would obtain for a normal metal. For much larger gaps this procedure must be modified, but then the details are relatively less important and those results are not used in our estimate of N(0)V.

In Fig. 3 we show our results together with some theoretical curves from Eq. (2). The agreement is good for  $\Gamma_N' \leq 0.1$ , but for larger values we are not satisfying the conditions under which Eq. (2) was derived. When the barrier is too thin, the theory is inapplicable and the values approach those found with no barrier at all. The inset table summarizes the results for  $\Gamma_{N}' < 0.05$ , and gives N(0)V which is calculated from Eq. (2) and



FIG. 2. Procedure used to determine  $\Omega_N$  from the differential resistance curves of the tunnel junction.



FIG. 3. Measured values of  $\Omega_{N'}$  plotted against  $\Gamma_{N'}$ , with theoretical curves generated from Eq. (2) shown for comparison. Inset table gives data for  $\Gamma_{N'} < 0.05$  with the calculated values of N(0)V.

the usual BCS relation

$$\Delta_N = 2\omega_c \exp\left[-\frac{1}{N(0)V}\right].$$

We find N(0)V for an 1800-Å gold film to be 0.72 ± 0.004.

Recent measurements<sup>6</sup> of N(0)V for copper and silver indicate similar results, and we plan further measurements on these metals. It would also be of interest to study the thickness dependence and extrapolate N(0)V to the bulk metal.

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<sup>1</sup>W. L. McMillan, Phys. Rev. <u>175</u>, 537 (1968).

<sup>2</sup>J. T. Hauser and H. C. Theuerer, Phys. Rev. Lett. 14, 270 (1965).

<sup>3</sup>W. L. McMillan, Phys. Rev. <u>167</u>, 331 (1968).

<sup>4</sup>N. E. Christensen, Solid State Commun. <u>9</u>, 749 (1971); N. W. Ashcroft and J. W. Wilkins, Phys. Rev.

Lett. <u>14</u>, 285 (1965).

<sup>5</sup>D. H. Prothero, S. M. Freake, and C. J. Adkins, Physica (Utrecht) 55, 744 (1971).

<sup>6</sup>C. Vallette, Solid State Commun. <u>9</u>, 895 (1971); E. Krätzig, Solid State Commun. <u>9</u>, 1205 (1971).

## Metallic State of the Electron-Hole Liquid, Particularly in Germanium

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We have calculated the ground-state energy of an electron-hole liquid. The kinetic and exchange energies are included exactly, and the correlation energy is estimated using Hubbard's modification of the random-phase approximation. In an isotropic electron-hole liquid, the metallic state is not bound relative to free excitons. In Ge the anisotropic band structure leads to a substantial binding of the metallic state. Application of a large  $\langle 111 \rangle$  strain to Ge reduces the situation to one resembling the isotropic case.

Recently there has been much interest in the properties of Ge in which a high concentration of excitons have been optically excited.<sup>1</sup> Since the lowest exciton states in Ge are indirect, they are relatively long-lived and many experiments can be performed under equilibrium conditions. Keldysh<sup>2</sup> has proposed that the striking changes that occur with increasing exciton density in the recombination radiation, far infrared absorption, and the electrical conductivity, are due to the formation of metallic droplets of the electron-hole liquid. In this Letter we wish to report microscopic calculations of the ground-state energy of a metallic electron-hole liquid. We consider three cases: (a) an ideal electronhole liquid of equal-mass particles, (b) Ge (unstrained), and (c) Ge with a large  $\langle 111 \rangle$  strain.

In the weak-binding limit in which the exciton binding energy  $E_B \ll E_d$ , the lowest direct energy gap, and the total number of excited carriers is very small compared to the number of atoms, we may write the Hamiltonian as

$$H = \sum_{\vec{k},\sigma,i=1}^{I} \epsilon_{i}^{a}(\vec{k}) a_{\vec{k},i,\sigma}^{\dagger} a_{\vec{k},i,\sigma} + \sum_{\vec{k},\sigma,j=1}^{J} \epsilon_{j}^{b}(\vec{k}) b_{\vec{k},j,\sigma}^{\dagger} b_{\vec{k},j,\sigma} + \sum_{\vec{q}} \frac{4\pi e^{2}}{\kappa q^{2}} \rho(\vec{q}) \rho(-\vec{q}), \qquad (1)$$

where  $a_{\vec{k},i,\sigma}^{\dagger}$  and  $b_{\vec{k},i,\sigma}^{\dagger}$  are creation operators for electrons and holes with spin  $\sigma$  in I and J sub-bands, respectively, and the wave vector  $\vec{k}$  is measured from the extremum of each sub-band. The Coulomb