

## Field penetration into proximity-coupled superconducting multilayers

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The penetration depth  $\lambda_{\text{eff}}$  of a magnetic field into multilayered superconductors is studied for the V-Ag proximity system. It is shown that  $\lambda_{\text{eff}}$  first decreases with increasing multilayer period, but then increases. This peculiar behavior is explained as the crossover of the system from a single superconductor to a composite one. With regard to the proximity effect, the nonmonotonic variation in  $\lambda_{\text{eff}}$  reveals a novel feature of thin normal films. This feature is understood by considering the fact that the normal-layer thickness restricts the electron mean free path.

Alternate deposition of thin superconducting and normal-metal layers produces a superconductor coupled by the proximity effect. This new class of materials arouses interest due to unusual characteristics different from those of current superconductors.<sup>1</sup> Besides, the fundamental study of artificial multilayers is of particular importance for revealing superconducting properties which cannot be studied otherwise.

Interest to date concerning the magnetic properties of superconducting multilayers has been focused mainly on high-field properties.<sup>1</sup> To the authors' knowledge, low-field properties have not yet been uncovered. In the present paper, field penetration into a proximity-coupled superconducting multilayer system V-Ag is presented for what we believe to be the first time.

Multilayered V-Ag films were prepared by ultrahigh-vacuum electron-beam evaporation. Total thicknesses range from 3100–6400 Å. Since field penetration is sensitive to the surface state, both sides of each sample end with Ag layers, which are stable in air and have a lustrous surface. The artificial periodicity was examined by x-ray diffraction, and the difference between the designed and the observed periods was found to be less than 5%. The superconducting transition measured by the inductive change in a field perpendicular to the films gives  $\Delta T < 20$  mK for the 10–90% width. Details of sample preparation as well as the structural quality are reported elsewhere.<sup>2</sup>

The penetration depth was determined by means of the ac susceptibility  $\chi_{\parallel}^{\prime}$ , which was measured with the Hartshorn-type mutual inductance bridge. The sample ( $8 \times 20$  mm<sup>2</sup>) was set parallel to the ac magnetic field, where the demagnetizing effect becomes negligible. For all samples, measurements were carried out under a 1-Oe ac field of 132 Hz. We confirmed that no appreciable difference appeared for fields of 0.2–2 Oe and up to 500 Hz. In Fig. 1 we show typical results for  $\chi_{\parallel}^{\prime}$  measured as a function of temperature, where  $-4\pi\chi_{\parallel}^{\prime}$  grows gradually, reflecting the temperature-dependent penetration depth.

Considering the dimension of the samples, one can say that the penetration in a direction perpendicular to the layer is responsible for  $\chi_{\parallel}^{\prime}$  (see inset of Fig. 1). We therefore assign  $\lambda_{\text{eff}}$  as the penetration depth in this direction.

Supposing penetration is exponential on the macroscopic scale, the magnetic induction in a multilayer is given by  $B \cosh(z/\lambda_{\text{eff}})/\cosh(D/2\lambda_{\text{eff}})$ , where  $B$  is the applied in-

duction,  $D$  is the total thickness of multilayer, and  $z=0$  denotes its center. The exponential form is reasonable because the analysis falls into the regime of the dirty local limit (see below). Thus the relation between  $\lambda_{\text{eff}}$  and  $\chi_{\parallel}^{\prime}$  is expressed as

$$-4\pi\chi_{\parallel}^{\prime} = 1 - \frac{2\lambda_{\text{eff}}}{D} \tanh\left(\frac{D}{2\lambda_{\text{eff}}}\right). \quad (1)$$

From Eq. (1), we evaluate  $\lambda_{\text{eff}}$  for four samples with the same thickness ratio of V and Ag (1:2). In Fig. 2 we show  $\lambda_{\text{eff}}$  as a function of multilayer period  $d$  ( $=d_{\text{V}}+d_{\text{Ag}}$ , the V- and Ag-layer thicknesses, respectively) for several reduced temperatures  $t$ . Curves of  $\lambda_{\text{eff}}$  vs  $d$  show peculiar characteristics, being similar for all  $t$  measured: As  $d$  increases,  $\lambda_{\text{eff}}$  first decreases and then turns up above  $d \approx 600$  Å. In the following, we attempt to explain this particular feature.

From the resistivity of a thick single V film and also of the multilayers, the mean free path in the V layer was estimated to be 13–23 Å. This indicates that the V layers in the present samples belong to the dirty limit.

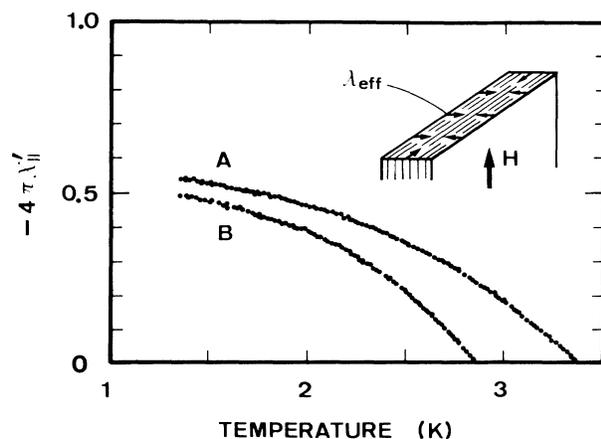


FIG. 1. Typical results of ac susceptibility as a function of temperature. (A) V(240 Å)/Ag(480 Å) with total thickness of 5520 Å, (B) V(160 Å)/Ag(320 Å) with total thickness of 5120 Å. Inset shows the sample geometry against applied field  $H$ .  $\lambda_{\text{eff}}$  is field penetration perpendicular to the layers.

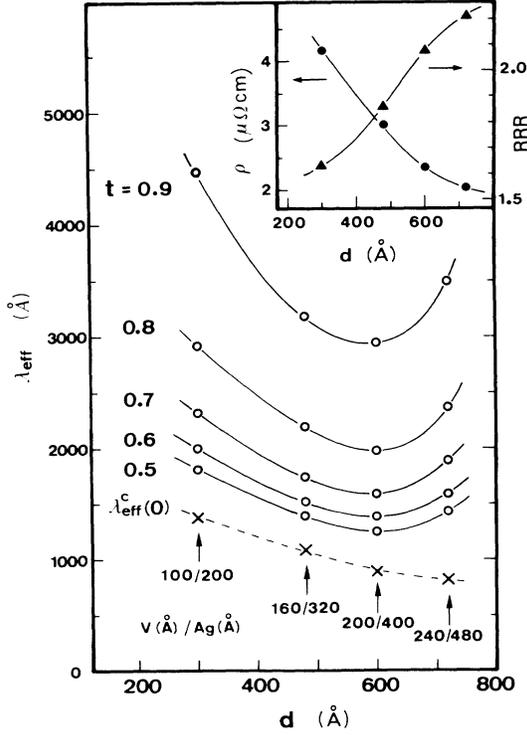


FIG. 2. Penetration depth  $\lambda_{\text{eff}}$  vs multilayer period  $d$  at several reduced temperatures  $t$  for samples with the same thickness ratio of V and Ag (1:2). Calculated zero-temperature penetration depth  $\lambda_{\text{eff}}^c(0)$  is also shown. Inset gives their resistivity  $\rho$  and residual resistivity ratio RRR vs  $d$ . Solid and dashed lines are guides for the eye.

The inset of Fig. 2 shows the resistivity  $\rho$ , measured parallel to the layers just above  $T_c$ , and the residual resistivity ratio RRR of the multilayers, where  $\rho$  decreases and RRR increases with increasing  $d$ . Since these quantities are mainly governed by Ag layers, their variations with  $d$  inform us that scattering at the V-Ag boundary restricts substantially the mean free path of electrons in Ag. This permits us to replace it by  $d_{\text{Ag}}$ , and thus the coherence length  $\xi_{\text{Ag}}(T)$  is expressed as

$$\xi_{\text{Ag}}(T) = (d_{\text{Ag}}/T_c)^{1/2} (\hbar v_F / 6\pi k_B t)^{1/2};$$

here  $v_F$  is the Fermi velocity of Ag,  $t$  is the reduced temperature, and  $T_c$  is the transition temperature of the V-Ag multilayer system. Estimated values of  $\xi_{\text{Ag}}(T_c)$  are tabulated in Table I, where for reference the values for  $d_V/d_{\text{Ag}} = 100 \text{ \AA}/100 \text{ \AA}$  and  $d_V/d_{\text{Ag}} = 100 \text{ \AA}/50 \text{ \AA}$  are also given. As seen there,  $\xi_{\text{Ag}}(T_c) > d_{\text{Ag}}$  for all samples, implying that the Ag layer can also be considered as dirty in the superconductivity sense.

Unfortunately, there exists no theoretical treatment for a multilayer system. As an approximation, therefore, we first discuss the field penetration into the layers in the framework of the conventional theory for a homogeneous superconductor in the dirty local limit. The penetration depth  $\lambda_{\text{eff}}(T)$  is given as<sup>3</sup>

$$\lambda_{\text{eff}}(T) = \lambda_L(T) J^{-1/2}(0, T) (\xi_0/l)^{1/2}, \quad (2)$$

where  $\lambda_L(T)$  is the London penetration depth,  $\xi_0$  is the Bardeen-Cooper-Schrieffer (BCS) coherence length,  $l$  is the mean free path, and  $J(R, T)$  is the integral kernel defined in the BCS theory.<sup>4</sup>  $\lambda_L(0)$ ,  $\xi_0$ , and  $l$  in Eq. (2) are, respectively, given by<sup>5</sup>

$$\lambda_L(0) = 1.33 \times 10^8 \gamma^{1/2} (n^{2/3} S/S_F)^{-1} \text{ cm}, \quad (3)$$

$$\xi_0 = 7.95 \times 10^{-17} (n^{2/3} S/S_F) (\gamma T_c)^{-1} \text{ cm}, \quad (4)$$

$$l = 1.27 \times 10^4 (n^{2/3} \rho S/S_F)^{-1} \text{ cm}, \quad (5)$$

where  $n$  (in  $\text{cm}^{-3}$ ) is the electron density,  $\gamma$  (in  $\text{erg cm}^{-3} \text{K}^{-2}$ ) is electronic coefficient of the specific heat,  $\rho$  (in  $\Omega \text{ cm}$ ) is the normal-state resistivity, and  $S/S_F$  is a ratio of the actual Fermi-surface area to that of free electrons. Note that in our case the quantities in Eqs. (2)–(5) are effective ones for the multilayer system used. Substituting Eqs. (3)–(5) into (2), we get

$$\lambda_{\text{eff}}^c(T) = 1.05 \times 10^{-2} J^{-1/2}(0, T) \frac{\lambda_L(T)}{\lambda_L(0)} \left( \frac{\rho}{T_c} \right)^{1/2} \text{ cm} \quad (6)$$

As the simplest case, we put  $T = 0$ . Since  $J(0, 0) = 1$ , one gets

$$\lambda_{\text{eff}}^c(0) = 1.05 \times 10^{-2} \left( \frac{\rho}{T_c} \right)^{1/2} \text{ cm}. \quad (7)$$

Equation (7) includes only experimentally determined quantities so that we can discuss the penetration depth without any fitting parameter. In Fig. 2, we also show  $\lambda_{\text{eff}}^c(0)$  thus obtained, which monotonically decreases with increasing  $d$ . Below  $d \approx 600 \text{ \AA}$ , it reproduces reasonably well the experimental results for  $\lambda_{\text{eff}}$  vs  $d$ , suggesting that the V-Ag multilayer system with small  $d$  behaves like a single superconductor, i.e., field penetration is strongly connected to the mean free path determining  $\rho$  and to  $T_c$  in the BCS fashion.

For  $d > 600 \text{ \AA}$ , however,  $\lambda_{\text{eff}}$  deviates considerably from that of the single-superconductor approximation given by Eq. (7). This deviation is probably caused by the composite nature of a multilayered system.<sup>6</sup> Comparison with the previously studied upper critical field  $H_{c2\parallel}(T)$  (parallel to the layer) for the same samples gives some insight to this behavior.<sup>7</sup> When  $d$  is small enough,  $H_{c2\parallel}(T)$  behaves like that of a single, three-dimensional superconductor. But when  $d$  exceeds a certain threshold,  $H_{c2\parallel}(T)$  shows dimensional crossover and this crossover takes place at higher re-

TABLE I. Sample parameters of V-Ag multilayer.

| $d_V/d_{\text{Ag}}$<br>( $\text{\AA}/\text{\AA}$ ) | $T_c$<br>(K) | $\rho$<br>( $\mu\Omega \text{ cm}$ ) | $\xi_{\text{Ag}}(T_c)$<br>( $\text{\AA}$ ) | $\xi_{\text{Ag}}(T_c)/d_{\text{Ag}}$ |
|--|--------------|--------------------------------------|--|--------------------------------------|
| 240/480  | 3.38         | 2.03                                 | 895  | 1.9                                  |
| 200/400  | 3.34         | 2.36                                 | 822  | 2.1                                  |
| 160/320  | 2.86         | 3.01                                 | 795  | 2.5                                  |
| 100/200  | 2.44         | 4.15                                 | 680  | 3.4                                  |
| 100/100  | 3.33         | 8.37                                 | 412  | 4.1                                  |
| 100/50   | 3.64         | 14.24                                | 278  | 5.6                                  |

duced temperature for greater  $d$ , implying that the superconducting coupling between V layers becomes weaker. In terms of the field penetration, weaker coupling results in longer penetration depth, so that  $\lambda_{\text{eff}}$  is expected to increase with  $d$ .

Although quantitative discussion cannot be made at the present stage, we believe that the appearance of a minimum  $\lambda_{\text{eff}}$  at  $d \approx 600 \text{ \AA}$  results from the competition of two factors, the electron mean free path in the Ag layers and the coupling strength between the V layers. In other words, this peculiar behavior is considered as a crossover from a single superconductor to a composite one.

In the meantime, the above-mentioned nonmonotonic variation of  $\lambda_{\text{eff}}$  with  $d$  is also interesting from the viewpoint of the proximity effect in the Ag layer. In the following, we evolve our study from this point of view.

Experimental studies intended to see the effect of normal-metal thickness on the penetration depth have so far been made with bilayer samples,<sup>8</sup> where the normal-layer thickness is on a scale of *several thousands* of angstroms. The characteristic so far revealed on this subject is that the penetration depth of the normal-metal layer increases with its thickness, at least at a finite temperature.<sup>8</sup> However, in our case the observed  $\lambda_{\text{eff}}$  below  $d \approx 600 \text{ \AA}$  evidently decreases with increasing  $d$ , suggesting that the variation of  $\lambda_{\text{Ag}}$  (penetration depth of Ag layer) with  $d_{\text{Ag}}$  in a thin region is opposite to that in a thick region. To confirm this novel implication, the effect of variation of the V-layer thickness must be eliminated.

For this purpose, we carried out further measurement of  $\lambda_{\text{eff}}$  as a function of  $d_{\text{Ag}}$ , where the V-layer thickness is fixed at  $100 \text{ \AA}$ . In Fig. 3  $\lambda_{\text{eff}}$  is shown at several reduced

temperatures  $t$  for these samples. When  $d_{\text{Ag}}$  increases, the order parameter in V should deteriorate because of its decreased volume fraction, so that one may expect longer field penetration. Contrary to this expectation, the observed  $\lambda_{\text{eff}}$  decreases with increasing  $d_{\text{Ag}}$ , demonstrating unambiguously that  $\lambda_{\text{Ag}}$  decreases with increasing  $d_{\text{Ag}}$  at least below  $200 \text{ \AA}$ .

Why is the behavior of  $\lambda_{\text{Ag}}$  vs  $d_{\text{Ag}}$  in a thin region different from that in a thick region? A qualitative understanding is attainable by taking into consideration the boundary scattering of electrons in the theory of proximity effect.

If the electron-electron interaction in Ag is negligible, one can get a position-dependent penetration depth in the dirty limit as<sup>9</sup>

$$\lambda_{\text{Ag}} = \frac{\hbar c}{2F(x)} N_{\text{Ag}} \left( \frac{k_B T \rho_{\text{Ag}}}{\hbar} \right)^{1/2} \propto \frac{1}{F(x)} \left( \frac{1}{l_{\text{Ag}}} \right)^{1/2}, \quad (8)$$

where  $F(x)$  denotes the pair field amplitude as a function of distance  $x$  from the interface,  $N_{\text{Ag}}$  is the density of states in Ag, and  $\rho_{\text{Ag}}$  is its resistivity. In Eq. (8), the mean free path  $l_{\text{Ag}}$  can be approximated by  $d_{\text{Ag}}$  as long as the Ag layer is thin enough. The profile of  $F(x)$  is qualitatively obtained by estimating  $\xi_{\text{Ag}}/d_{\text{Ag}}$  because  $\xi_{\text{Ag}}$  is considered to be a damping constant of hyperbolic cosine function  $F(x)$ . As listed in Table I,  $\xi_{\text{Ag}}(T_c)/d_{\text{Ag}}$  is several times greater than unity for small multilayer period, meaning  $F(x)$  is nearly constant. On the assumption that the absolute value of  $F(0)$  does not appreciably change with  $d_{\text{Ag}}$ , one finds that the dominant factor determining the  $d_{\text{Ag}}$  dependence of  $\lambda_{\text{Ag}}$  is  $(1/d_{\text{Ag}})^{1/2}$ . So, for  $d_{\text{Ag}}$  below a certain limit,  $\lambda_{\text{Ag}}$  decreases with increasing  $d_{\text{Ag}}$ , qualitatively reproducing the experimental observations.

When  $d_{\text{Ag}}$  becomes greater, however, the simple proportionality of  $\lambda_{\text{Ag}}$  to  $(1/d_{\text{Ag}})^{1/2}$  is by no means relevant. As listed in Table I, in this regime  $\xi_{\text{Ag}}(T_c)/d_{\text{Ag}}$  approaches unity and hence  $F(x)$  is no longer constant, but spatially varies. That is,  $F(x)$  deteriorates at the center of the Ag layer. This effect eventually elongates  $\lambda_{\text{Ag}}$  through the factor  $1/F(x)$  in Eq. (8). On the contrary, the factor  $(1/l_{\text{Ag}})^{1/2}$  becomes less dominant, because  $l_{\text{Ag}}$  approaches the intrinsic mean free path instead of  $d_{\text{Ag}}$ .<sup>10</sup> This tendency seems to appear as the increase in  $\lambda_{\text{eff}}$  measured in the region of  $d > 600 \text{ \AA}$  in Fig. 2.

To see the relation between the two different treatments, the single-superconductor approximation and consideration from the viewpoint of the proximity effect, the work of Simon and Chaikin<sup>8</sup> is suggestive. According to them, a thin proximity-effect superconductor with a spatially constant order parameter acts like an ordinary superconductor, while thick films with spatially varying order parameters depart from it.

In summary, we have presented the penetration depth  $\lambda_{\text{eff}}$  of a V-Ag multilayer system as a function of multilayer period  $d$  (thickness ratio, 1:2). For a short period ( $d < 600 \text{ \AA}$ ),  $\lambda_{\text{eff}}$  can well be explained by a single superconductor, while for longer period ( $d > 600 \text{ \AA}$ ), the weak coupling nature between V layers becomes significant. Competition between these two behaviors results in a minimum  $\lambda_{\text{eff}}$  at  $d \approx 600 \text{ \AA}$ . Viewed from the proximity

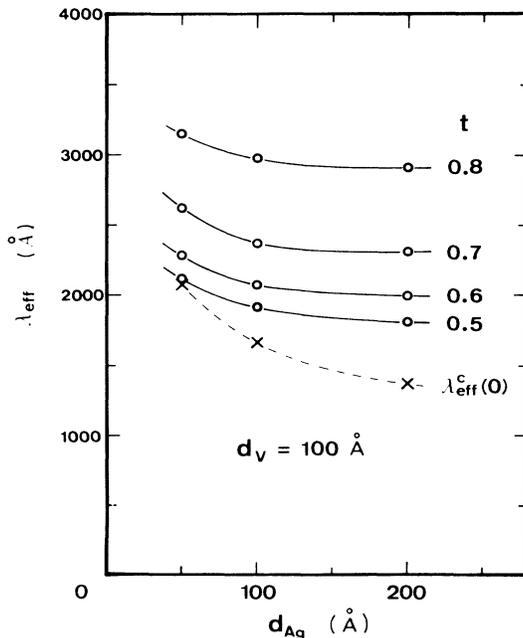


FIG. 3. Penetration depth  $\lambda_{\text{eff}}$  at several reduced temperatures  $t$  for samples with different Ag-layer thickness  $d_{\text{Ag}}$ . Calculated zero-temperature penetration depth  $\lambda_{\text{eff}}^{\text{S}}(0)$  is also shown. Solid and dashed lines are guides for the eye.

effect, the nonmonotonic variation of  $\lambda_{\text{eff}}$  with  $d$  exhibits a novel feature of thin normal films, i.e., for normal-metal thickness below a certain limit, the field penetration into the normal layer decreases as the thickness increases. This feature can be understood by taking into consideration the fact that electron scattering at the interface becomes a dominant factor in thin layers.

Finally, we emphasize that it is with multilayered samples that this remarkable feature of a very thin normal metal in the proximity system could be revealed, where the

magnetic field penetrates normal-metal films many times before it decays out, and thus  $\lambda_{\text{Ag}}$  can well be reflected in  $\lambda_{\text{eff}}$ . It would be extremely difficult to find this feature with a bilayer sample.

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<sup>1</sup>For a review, see, e.g., S. T. Ruggiero and M. R. Beasley, in *Synthetic Modulated Structures*, edited by L. Chang and B. C. Giessen (Academic, New York, 1985), p. 365.

<sup>2</sup>N. Hosoito, T. Yamada, K. Kanoda, H. Mazaki, and T. Shinjo, *Bull. Inst. Chem. Res. Kyoto Univ.* **64**, 230 (1986).

<sup>3</sup>M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1975), p. 67.

<sup>4</sup>J. Bardeen, L. N. Cooper, and J. R. Schrieffer, *Phys. Rev.* **108**, 1175 (1957).

<sup>5</sup>T. P. Orlando, E. J. McNiff, Jr., S. Foner, and M. R. Beasley, *Phys. Rev. B* **19**, 4545 (1979).

<sup>6</sup>Surface Ag layers are not responsible for this, because similar

behavior of  $\lambda_{\text{eff}}$  vs  $d$  holds even up to high reduced temperature ( $t=0.9$ ), where  $\lambda_{\text{eff}}$  is far greater than  $d_{\text{Ag}}$ .

<sup>7</sup>K. Kanoda, H. Mazaki, T. Yamada, N. Hosoito, and T. Shinjo, *Phys. Rev. B* **33**, 2052 (1986).

<sup>8</sup>See, for example, R. W. Simon and P. M. Chaikin, *Phys. Rev. B* **23**, 4463 (1981); **30**, 3750 (1984).

<sup>9</sup>Orsay Group on Superconductivity, in *Quantum Fluids, Proceedings of the Sussex University Symposium*, edited by D. F. Brewer (North-Holland, Amsterdam, 1966), p. 26.

<sup>10</sup>As seen in inset of Fig. 2,  $\rho$  and RRR tend to saturate above  $d \approx 600 \text{ \AA}$ .