RESISTIVE VOLTAGES IN SUPERCONDUCTING Pb-In ALLOY FILMS (III)

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The thin films of Pb-50In alloy with thickness d ranging from 700 Å to 6000 Å have been prepared by a vacuum evaporation technique to study the resistive voltages at 4.2 K in the mixed state of this material. The critical current density J_c determined from the V-I characteristics varies with an external magnetic field following an existing theoretical prediction. The superconducting-to-normal transition is found to occur at two stages, or at the two upper critical fields H_{c2} and H'_{c2}, in the thinner films with d<1500 Å, as seen in the voltage-field curve. It is also found that a mosaic structure due probably to a shrinkage of evaporated film exists in the thinner films, which causes a sharp increase in the residual resistivity but does not affect appreciably the superconducting properties of the alloy films, with only increasing a pinning force.

1. INTRODUCTION

Since the original work of type II superconductivity developed by Abrikosov in 1956, it is nowadays known that in addition to the complete expulsion characteristic of the Meissner state, a type II superconductor can also exhibit a mixed state, in which the magnetic flux penetrates the sample in the form of quantized flux lines --- frequently referred to as vortices or fluxoids. The superconducting properties are characterized by various parameters such as, critical temperature T_c, thermodynamic critical field H_c, lower and upper critical fields H_{c1} and H_{c2}, penetration depth λ, coherence length ξ, and Ginzburg-Landau parameter $\kappa(=\lambda/\xi)$.[1)] These quantities depend on the kind of a material, as well as the size or the form of the sample. For example, when one of the dimensions of a superconducting specimen becomes comparable to the penetration depth, its critical field becomes much higher than that of a bulk sample of the same material at the same temperature, as described later.

Using dc and pulsed currents, we have so far been interested in the resistive voltages due to the flux motion in Pb-In alloy films. In

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previous works, we measured the effect of a small oscillatory magnetic field on the dc voltage in $Pb_{0.4}In_{0.6}$ alloy films[2] and the transient behavior of the flux motion in $Pb_{0.9}In_{0.1}$ alloy films.[3] However,there still remain experimental difficulties such as the difference in the alloy composition between the evaporated film and the source ingot and the presence of crystalline imperfections in the grown films. In the present work, the starting alloy composition was set to a 1:1 ratio, $Pb_{0.5}In_{0.5}$, or Pb-50In according to the notation by Farrell et al.[4] Here the dc characteristics of the superconducting alloy films and its film surface were examined as before.

2. EXPERIMENTAL

The sample preparation was proceeded with the same manner as the previous one: The two elements were melted in an evacuated quartz ampul at 400°C for 24 hours. The synthesized alloy was then evaporated from a heated tantalum boat onto a glass substrate kept at room temperature in a high vacuum of around 10^{-7} Torr. The shape of the specimen was of a four-probe type; 1.8 mm in width and 3.5 mm in length for the potential difference measurement.

The film thickness d, an important factor in thin film studies, was estimated by a weight method using the expression of

$$d = m/\pi\delta h^2, \qquad (1)$$

where m is the amount of the evaporated alloy, h the vertical height between the substrate and the boat, and δ the alloy density. Here the alloy density was estimated by a linear interpolation procedure as

$$\delta = \delta_{Pb}(1 - x) + \delta_{In}x. \qquad (2)$$

δ_{Pb} and δ_{In} are the densities of Pb and In, respectively, and x is the alloy ratio, now equal to 1/2; thus δ=9.31 g/cm^3 with δ_{Pb}=11.34 g/cm^3 and δ_{In}=7.28 g/cm^3.

The electrical measurements were made by a dc potentiometric technique in a perpendicular magnetic field with respect to the film surface. Both the specimen and a home-made superconducting solenoid were immersed in liquid helium bath. Some of the bulk superconducting parameters for Pb-50In alloy are known as below;[4]

T_c=6.39 K, H_{c1}=152 G, H_{c2}=3.50 kG,
and residual resistivity at 4.2 K =1.78 x 10^{-5} ohm-cm.

3. RESULTS AND DISCUSSION

3.1 Electrical measurements

Figure 1 shows the resistivities ρ at 300, 77 and 4.2 K as a function of thickness for several samples of Pb-50In alloy films. In this alloy composition a marked increase in ρ with decreasing thickness below 1000 Å is to be noted, compared with that of other alloy composition.[3] Such a sharp increase in ρ is not simply due to the decrease in the mean free path of conduction electrons as observed in a number of metals, but rather it can be attributed to the crystalline imperfections of deposited films. As shown later, the thinner films of Pb-50In alloy involve many defects or islands, probably due to a thermal shrinkage of the film after deposition. A decrease in the mean free path of thin films and small wires has been known as a classical size effect, in which electrons are scattered elastically or inelastically in part at the boundaries. Now an excellent review on these effects exists,[5] and we have compared the observed resistivity ρ at 4.2 K with a simple expression of the form

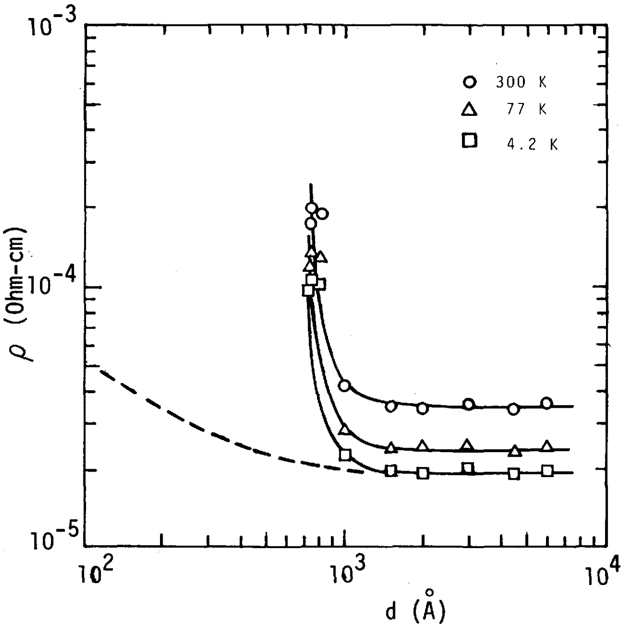

Fig. 1. The thickness dependence of the resistivity at three fixed temperatures for Pb-50In alloy films. The dashed curve is a plot of eq.(3) with the bulk resistivity[4] ρ_0=1.78 x 10^{-5} Ohm-cm at 4.2 K and ℓ=500 Å.

$$\rho = \rho_0(1 + 0.4\ell/d), \tag{3}$$

where ρ_0 is the bulk resistivity and ℓ is the bulk mean free path of electrons. The dashed curve in Fig. 1 is a plot of eq. (3) with the best-fit values of ρ_0=1.78 x 10^{-5} Ohm-cm and ℓ=500 Å. Thus this evaluation shows that the resistivity of uniform films would decrease much more slowly, at least down to 500 Å, than the observed one; the discrepancy is due to the imperfections of actual films, as mentioned above.

The temperature dependence of the resistivity was measured only for one sample to estimate the critical or transition temperature T_c of thin film; T_c=6.46 K for a film with d=3000 Å. This value is slightly large compared with the bulk value (=6.39 K).[4] Because of the lack of available data on the systematic variation of T_c with thickness, it is not certain whether the difference is due to the size effect. Probably it may stem from the deviation of alloy composition of thin film from that of the starting bulk ingot.

The behavior of the critical current of type II superconductors is well established. As with type I superconductors, there is an upper limit to the transport current density which a type II superconductor will carry without losing its property of zero resistance. This critical current is a function of the external field and the temperature. In contrast with type I superconductors, usual type II superconductors are alloys or compounds and contain an appreciable defect density and for these materials the resistance transition (superconducting-to-normal state) is spread over a finite current range. Usually the magnetic field is applied and the current is increased until the first measurable voltage appears across the terminals of a four-probe technique. This current is taken to be the critical value. It depends on the sensitivity of the measuring apparatus, usually of the order of $10^{-7}\sim10^{-6}$ volts and is a measure of the first detectable pinning force or viscous flux-flow in the material. Such a voltage-current characteristic at different fields is shown in Fig. 2 for a sample with d=3000 Å. The initial nonlinear portion arises from a flux-pinning and then the resistive voltage V_s increases linearly with increasing the sample current I_s---- an ohmic portion or flux-flow state. The critical current density J_c versus transverse field H_o characteristic of the present samples will be shown later.

Although the upper critical field H_{c2} of a material is determined accurately by magnetization measurements, the resistance transition is often measured as well, in which the resistive voltage is measured when the applied magnetic field is varied. Typically the field value where the curve is vertical (measured using a very small transport current so that the self-field of the sample is negligible) is identified as H_{c2}. Ideally H_{c1} and H_{c2} are obtainable from the resistance transition as the field at which a voltage first appears (H_{c1}) and the field of the abrupt rise (H_{c2}). In practice, however, the transition is not sharp.

Fig. 2. Typical voltage-current characteristic at 4.2 K for a sample with d=3000 Å, the transverse magnetic field being fixed at constant.

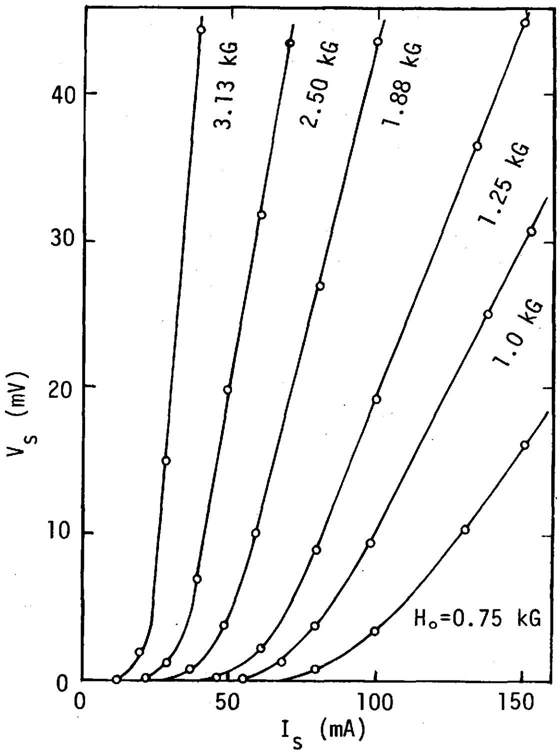

Figure 3 illustrates some of our results for the alloy films with different thickness, measured at different sample current I_s, as indicated. It is to be noted that for thinner films with $d \leq 1500$ Å the superconducting-to-normal transition is gradual and occurs at two stages; we here take H'_{c2} as the initial stage and H_{c2} as the final stage ----- the field at which the sample enters completely the normal state. For thicker films the transition takes place at one stage, though gradual. Since both transitions are gradual, an extrapolation procedure in the observed curves is employed to estimate the values of H'_{c2} and H_{c2}.

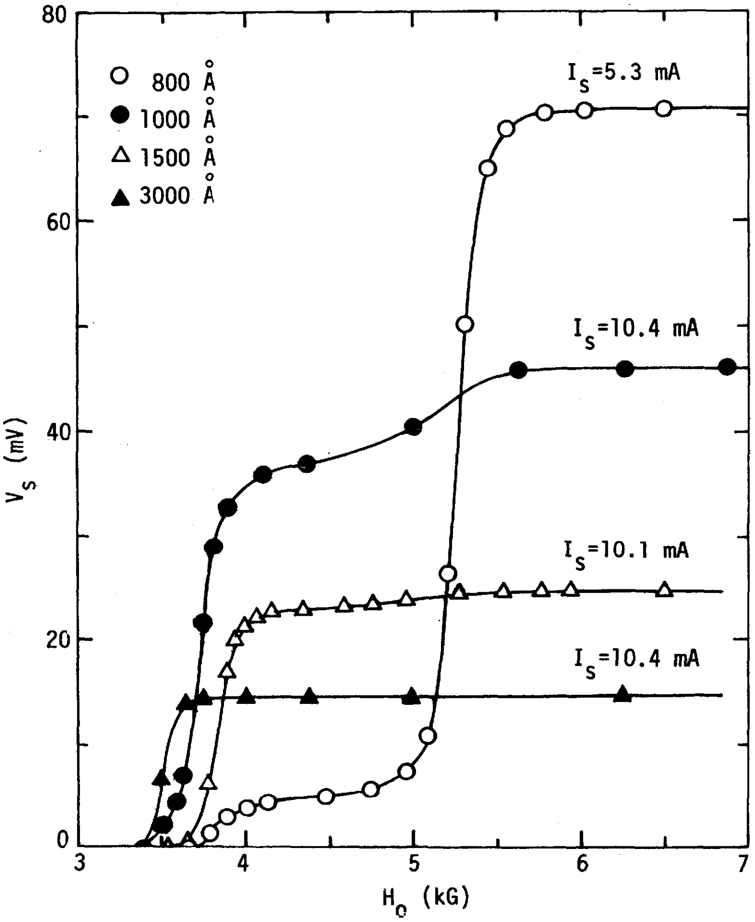

Fig. 3. Resistive voltage V_s versus transverse magnetic field H_0 for some samples with different thickness, under a constant sample current I_s as indicated.

The upper critical fields H_{c2} and H'_{c2} thus determined are plotted in Fig. 4 against the film thickness for Pb-50In alloy films, where H_{c2} is indicated by open circles and H'_{c2} by open squares. It is evident that the critical field increases with decreasing thickness for thinner films with $d < 2000$ Å, but for thicker films with $d > 3000$ Å it tends to increase with thickness rather than approaching to the bulk value.

According to a discussion given by Lynton,$^{6)}$ the size effect of the critical field H_s, after his notation, is written by

$$H_s/H_c = 1 + \lambda/d, \text{ for } d \gg \lambda, \tag{4}$$

where λ is the penetration depth and d the thickness of a sample.

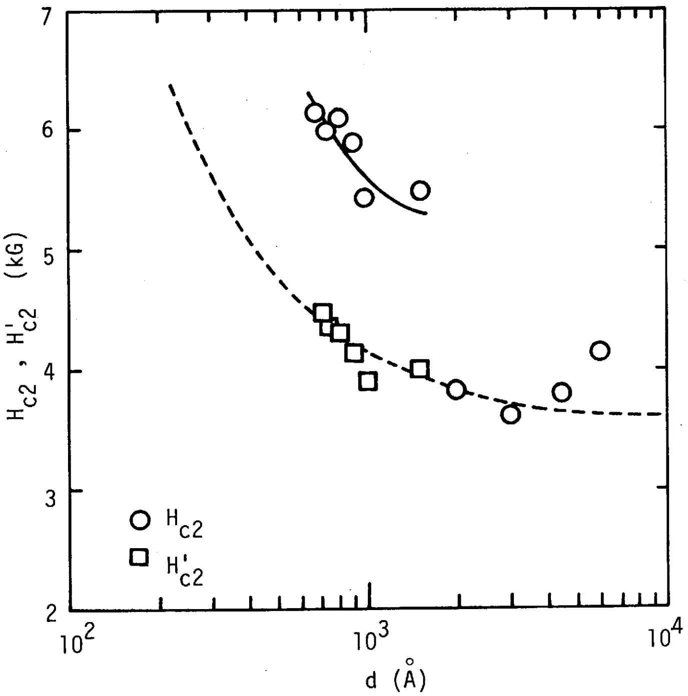

Fig. 4. Thickness dependence of the upper critical fields H_{c2} and H'_{c2} (see text)for Pb-50In alloy films; the bulk value H_{c2}=3.5 kG.[4)] The dashed curve is a plot of eq. (6) with the best-fit value of λ_o=180 Å.

This expression is derived from the basic Gorter-Casimir thermodynamic description for an ideal type I superconductor, which in an external field H acquires an effective magnetization M(H) and becomes normal when

$$\int_0^H M(H)dH = H_c^2/8\pi. \qquad (5)$$

The integral is the area under the magnetization curve. It is also assumed that this curve remains linear to H up to a critical field H_s. Similarly, for the present alloy films we write the observed thickness dependence of upper critical fields H_{c2} and H'_{c2} in the form

$$H_{c2}/H_{c2b} = 1 + \lambda_o/d, \qquad (6)$$

where λ_o is only a parameter resembling to the penetration depth in eq. (4) and H_{c2b} is the bulk value (=3.5 kG) of Pb-50In alloy obtained from the magnetization curve.[4)] The experimental points are fitted well by eq. (6) with the best-fit value λ_o=180 Å. Here we do not take account of the demagnetization factor of the sample and only we refer to the functional form of eq. (4) for comparison; hence at present it is uncertain whether λ_o has any physical meaning.

Finally, Fig. 5 shows the critical current density J_c at 4.2 K plotted against the transverse magnetic field H_o for various alloy films. The critical current was dtermined from the V-I characteristic for each sample such as shown in Fig. 2. It is known that the high critical currents in some type II (hard) superconductors are due to the presence of defects in the material and the current density remains large up to H_{c2} in the J_c-H_o curve. The presence of a large defect density is also shown by the irreversible magnetization curve. Such a J_c-H_o characteristic is also seen in our alloy films; the absolute value of J_c increases with decreasing the film thickness, indicating the increased crystalline imperfections in the thinner films. For quantitative discussion

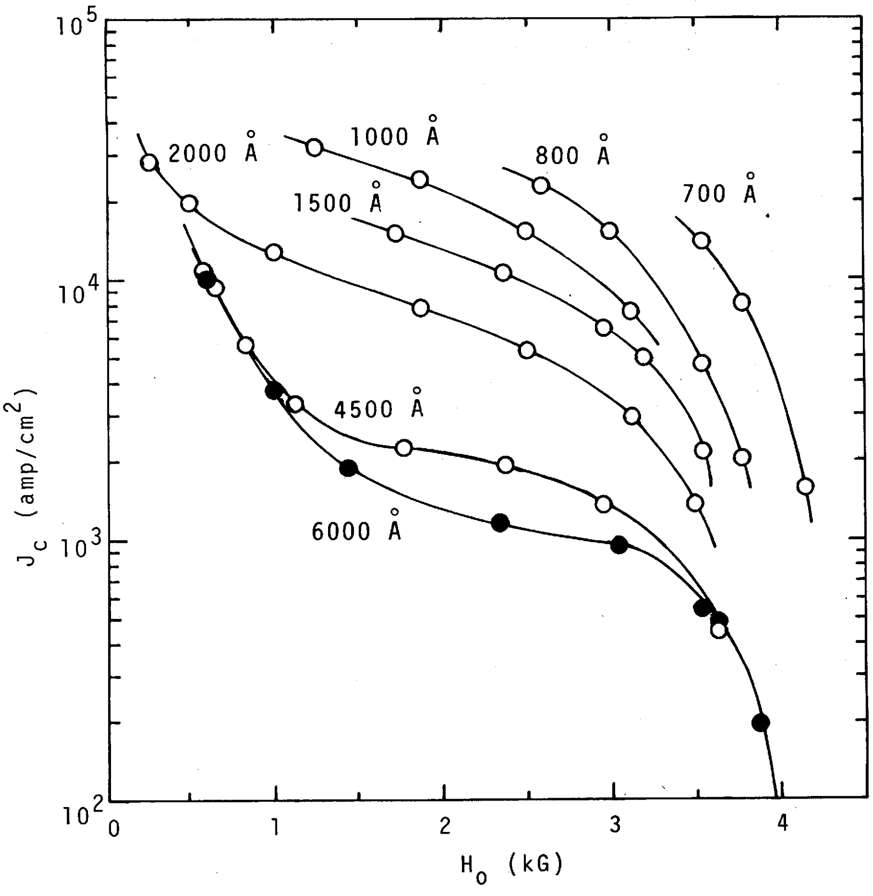

Fig. 5. Critical current density J_c versus transverse magnetic field H_o of the alloy films with various thickness at 4.2 K.

various models have been proposed to evaluate the critical current in the mixed state as a function of transverse field. For example, by assuming that the current is carried in a layered series of thin laminae, El Bindari and Litvak[7)] derived a current variation of the form

$$J_c(H)/J_c(0) = 1 - (H/H_{c2})^{1/3}, \qquad (7)$$

where $J_c(0)$ is the critical current density at zero field. The equation is reported to agree well with experiments on a number of high κ materials. Though not shown in Fig. 5, we have also found that our data follow approximately eq. (7) with suitable values of $J_c(0)$ and H_{c2} obtained for each film over a wide range of magnetic field.

3.2 Film surface

The film surface of some of evaporated Pb-50In alloy films was examined by a scanning electron microphotograph (SEM) using Hitachi HSM-2A type. The results are shown in Fig. 6. Similar to the previous SEM pictures of Pb-60In and Pb-10In alloy films,[2,3)] the thinner films of Pb-50In alloy also consist of a mosaic structure with a number of voids but seem to be continuous. Such a structure is probably due to a shrinkage of the deposited layer. And it is likely that the shrinkage is slightly large in this alloy composition compared with other alloy composition. As shown in Fig. 1, it may also be responsible for a sharp increase in residual resistivity of the thinner films with thickness d < 1000 Å. However, it is interesting to note that the

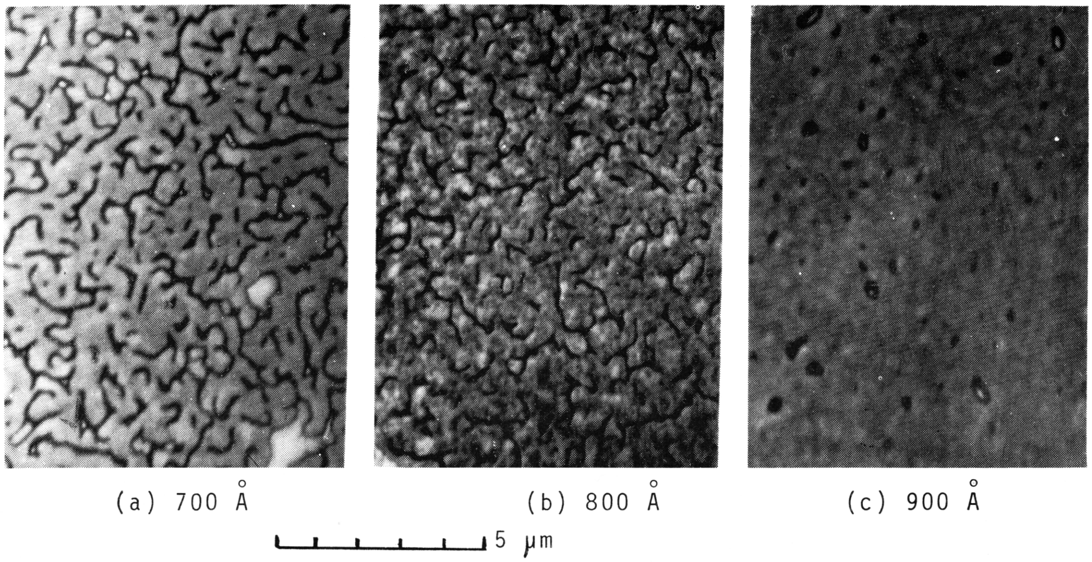
(a) 700 Å (b) 800 Å (c) 900 Å

Fig. 6. The SEM pictures of Pb-50In alloy films with different thickness.

superconducting properties, e.g., H_{c2} (Fig. 4), vary monotonically as the thickness is decreased.

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