Superconductor-insulator transition in ultrathin Pb films: Localization and superconducting coherence

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We have measured temperature-dependent resistivities of a set of Pb ultrathin films by repeating *in situ* deposition at liquid-He temperature. Observed conductance in the fermion region can be described by a single-parameter scaling function (β function) with a self-consistent theory for localization in two dimensions. Based on estimated values of electrical localization length ($\xi_{\rm loc}$) and superconducting coherence length ($\xi_{\rm super}$), the superconductor-insulator transition is confirmed to occur at the point where $\xi_{\rm loc}\sim 2\xi_{\rm super}$. Moreover, only samples exhibiting ln$T$ dependence in the fermion region become global coherent superconductors at low temperature.

Recently, interplay between disorder and superconductivity in two dimensions (2D) has gathered interest as a field of study.[1–10] In particular, superconductor-insulator (SI) transition in 2D has attracted considerable interest in connection with the quantum Hall liquid-insulator transition.[11] Scaling behavior for the SI transition has been the subject of numerous experiments[4–6,12] in which a value of the critical sheet resistance has been obtained near $h/4e^2$. Fisher and coworkers argued the SI transition in a disordered superconducting thin film by scaling formulation.[2,3] In this argument, they did not directly address localization length of electrons (quasiparticles). When one considers the formation of Cooper pairs, the relationship between electrical localization length $\xi_{\rm loc}$ and superconducting coherence length $\xi_{\rm super}$ seems to be important. One can speculate that the SI transition occurs at the point where $\xi_{\rm loc}=\xi_{\rm super}$. If $\xi_{\rm loc}$ is smaller than $\xi_{\rm super}$, the system cannot form a Cooper pair but if $\xi_{\rm loc}$ is longer than $\xi_{\rm super}$, the system can form a Cooper pair. However, there is no experimental evidence to support this theory regarding the SI transition.

In this paper, we discuss experiments wherein we measured the temperature dependence of resistivities of ultrathin Pb films by sequential repeating *in situ* deposition at low temperature. We indicate that all conductance data in the fermion region, where we can treat the carriers as fermion, fit a single universal curve (β function) of a self-consistent theory for localization in 2D.[13] We estimated $\xi_{\rm loc}$ and $\xi_{\rm super}$ from observed data, and show that the SI transition occurred at a point where $\xi_{\rm loc}\sim 2\xi_{\rm super}$. Furthermore, we discovered that films exhibiting ln$T$ dependence in the fermion region show global coherent superconducting behavior at low temperature; and that films exhibiting $\exp[-(T_0/T)^\alpha]$ dependence in the fermion region show insulator (Bose glass) behavior at low temperature.

The Pb films are grown by vapor deposition of the metal onto liquid-He-cooled glass substrates. A sequence of films can be fabricated and measured *in situ*. The film is 2.5 mm $\times$4.0 mm in area. Temperature dependence of resistance is measured using a Keithley model 182 nanovoltmeter and a Keithley model 6512 electrometer in the range of 2$\sim$10 K.

Figure 1 shows $R_\square(T)$ curves of 39 sequential Pb films. Film thickness is controlled by evaporation time and varies from 13.8 to 42.8 Å. The thickness of the thickest film is calculated as the average Pb grain height [Fig. 2 shows the atomic force microscope (AFM) topography of the thickest film]. The thickness of other films is evaluated from evaporation time. The film changes from an insulator to a superconductor with increasing film thickness. The sheet resistance of the thinnest film changes from 10^7 to 10^{13} Ω while temperature decreases from 10 to 2 K. In the thickest film, superconducting transition takes place at 3.5 K and a residual sheet resistance of about 20 Ω remains after the transition occurs. This residual resistance is observed in samples which show superconducting transition. Residual resistances decrease with temperature and reach a minimum near 2.3 K in some samples. A critical resistance which distinguishes insulators (R increases as $T\rightarrow 0$) from superconductors (R decreases as $T\rightarrow 0$) lies between 10^4 and 10^5 Ω. Considering the fermion region and the boson region, where we can treat carriers as bosons, we analyze the data below and above 4 K, separately. Our analyses include estimation of the critical sheet resistance from the data observed below 4 K and esti-

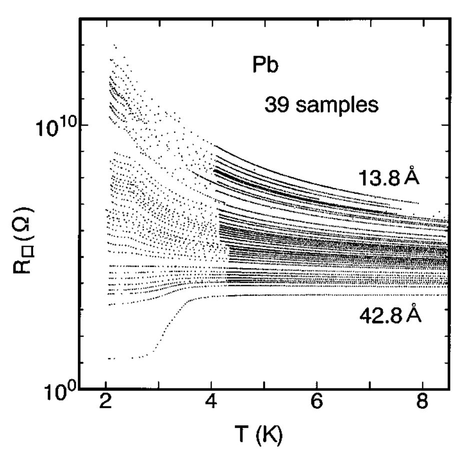

FIG. 1. Resistance of the samples vs temperature. The film is grown sequentially *in situ* by evaporation of Pb. The film changes to superconductor from insulator as film thickness increases. Film thickness changes from 13.8 to 42.8 Å. The superconducting transition takes place at 3.5 K in the thickest film.

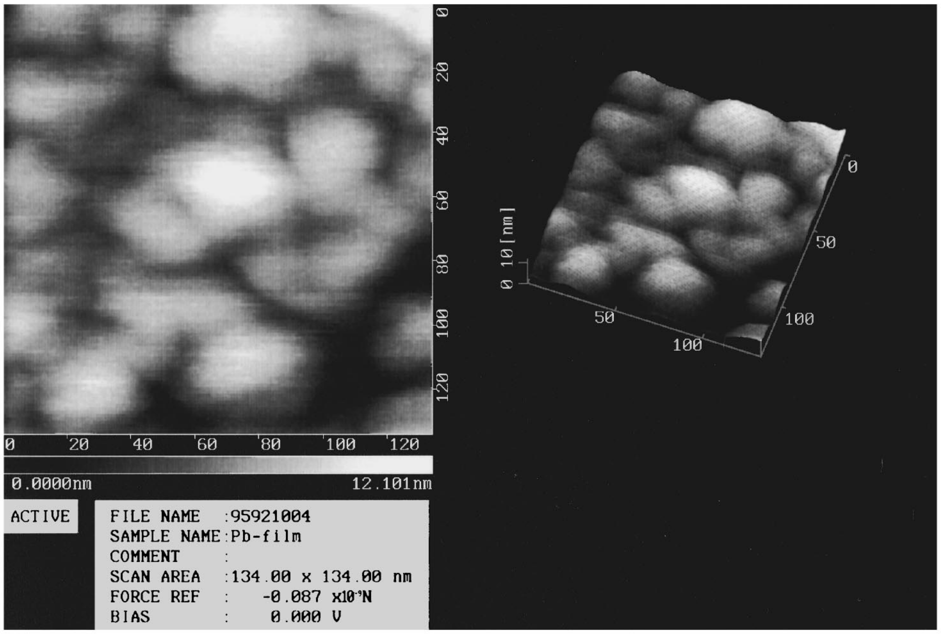

FIG. 2. AFM topography of the thickest (42.8 Å) film. This value represents the average Pb grain height. The thicknesses of other films is calculated from the evaporation time.

mation of the electrical localization length $\xi_{\rm loc}$ of each film from the data observed above 4 K. The validity of the analyses will be discussed later in this paper.

In Fig. 3, we have plotted the temperature coefficient of resistance (TCR) below 2.3 K versus sheet resistance at 2.3 K for films near the SI transition. Films with positive values of TCR become superconductors at low temperature and films with negative values of TCR become insulators at low temperature. The critical sheet resistance which distinguishes insulators from superconductors is regarded as the point where TCR is zero. From Fig. 3 the critical resistance is found to be between 20∼23 kΩ. This value is close to h/e^2.

In a high-temperature region above 4 K, we measure characteristics of electrical transport from a strongly disordered regime to a weakly disordered regime in a sequence of films.

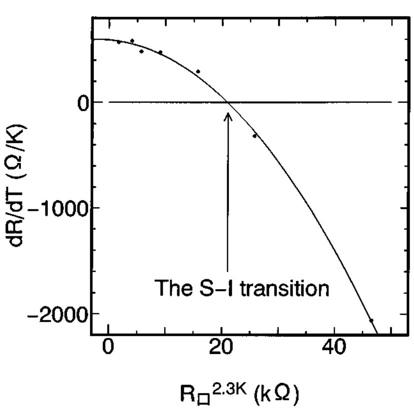

FIG. 3. Temperature coefficient of resistance (TCR) vs sheet resistance at 2.3 K. The sheet resistance is a parameter of the degree of disorder in the films. A positive TCR indicates that a film becomes a superconductor ($T\rightarrow 0$) and a negative TCR indicates that a film becomes an insulator ($T\rightarrow 0$). The SI transition takes place at the point where TCR is zero.

The temperature dependence of resistivities of strongly disordered films takes the form $R_{\Box}(T)=R_0\exp[(T_0/T)^{\alpha}]$, where T_0 is general activation energy. In hopping electrical transport, the value of α is unity and in variable range hopping (VRH) transport, the value of α is 1/3 in 2D.[14] The conductance of films in a weak-localized regime shows logarithmic temperature dependence. In localization theory,[15] the conductance of a weak-localized regime is given by

$$\sigma=\sigma_0+\frac{e^2p}{2\pi^2\hbar}\ln T, \tag{1}$$

where p is a constant determined by the scattering mechanism of electrons. The observed coefficient of $\ln T$ is 2.59×10^5 and the value of p is 2.1, which is consistent with the theory. Next, we analyze the observed data described so far in terms of the conventional β function,[16] $\beta(g)\equiv d(\ln g)/d(\ln L)$, where L is the sample size and g is the dimensionless conductance $[g=(\hbar/e^2)\sigma]$. The sample size L is regarded as a cutoff length due to inelastic scattering: $L^2=DT^{-p}$, where D is a diffusion constant of electrons. The β function is expressed as

$$\beta(g)=-\frac{2}{p}\frac{d(\ln g)}{d(\ln T)}. \tag{2}$$

Using this form, we have plotted all the conductance data above 4 K in Fig. 4. We can see that all the data above 4 K fit a single universal curve. We have also tried to plot the β function of all the conductance data below 4 K, but these data cannot be fitted to a single universal curve. Thus, from Fig. 4 we prove that all the films are in the disorder fermion region above 4 K but are not in the disorder fermion region below 4 K. The result that all the data are scaled by the β function above 4 K confirms our analyses.

Furthermore, to verify the criterion that the SI transition occurs at the point where $\xi_{\rm loc}=\xi_{\rm super}$, we estimate the elec-

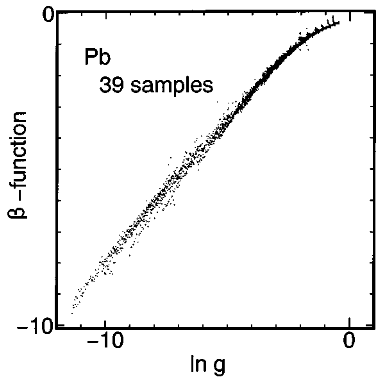

FIG. 4. The β function vs dimensionless conductance for all films above 4 K. This figure shows that the conductance of all films studied fit a single universal curve. This confirms that all films are in the fermion region above 4 K. Below 4 K, data cannot be fitted to a single universal curve.

trical localized length ξ_{loc} from the high-temperature data. Wölfe and Volhardt have produced a self-consistent theory[13] of the transition from weak to strong localization in a thin film, neglecting interaction. They found that

$$\frac{\hbar}{e^2 R_{\square}} = \frac{1}{2\pi^2}\,[\ln(1+y^2)](1+y)\exp(-y), \tag{3}$$

where $y = L/\xi_{loc}$, ξ_{loc} is an electrical localization length related to the elastic mean free path l by $\xi_{loc} = l\ \exp(\pi k_F l/2)$, and k_F is the Fermi wave number. At a finite temperature, sample size L is related to T by $L \sim T^{-p/2}$. The temperature dependence of the sheet resistance above 4 K can be written in the form of Eq. (3) because all data are fitted to the β function above 4 K. By fitting the data to Eq. (3), we estimate the electrical localization length ξ_{loc} of each film. We then estimate the superconducting coherence length (ξ_{super}) of each film. ξ_{super} is related to the elastic mean free path l by

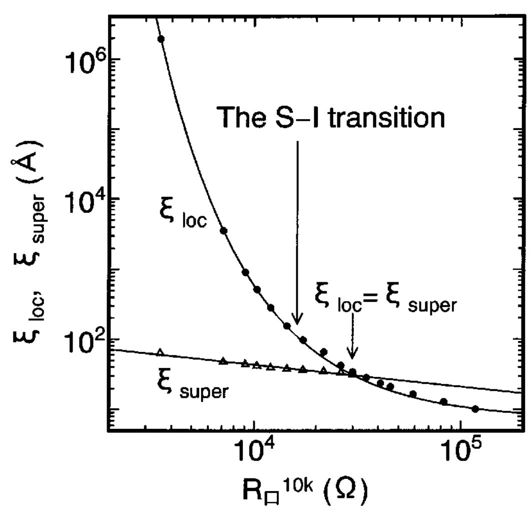

FIG. 5. The superconducting coherence length (ξ_{super}) and the electrical localized length (ξ_{loc}) vs the sheet resistance at 10 K. The sheet resistance at 10 K is a parameter of the degree of disorder in the films. The SI transition takes place at a point where $\xi_{loc} \simeq 2\xi_{super}$, and the criterion that the SI transition takes place at the point where $\xi_{loc} = \xi_{super}$ is not observed.

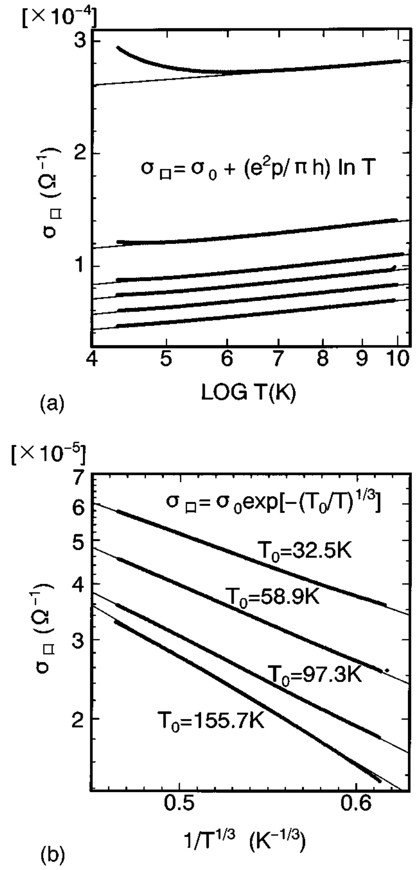

FIG. 6. (a) Conductance vs temperature for superconducting films. The conductance shows logarithmic temperature dependence. This indicates that the films are weak-localized regimes. Observed coefficient of $\ln T$ is 2.59×10^5 and the value of p is 2.1. (b) Conductance vs temperature for insulator films near the SI transition. The conductance shows VHR transport $1/\sigma_{\alpha} \propto \exp[-(T_0/T)^{1/3}]$. T_0 is the general activation energy.

$\xi^2_{super} = \xi_{BCS} l$,[17] where ξ_{BCS} is a superconducting coherence length in a clean superconductor.

We have plotted ξ_{super} and ξ_{loc} vs the sheet resistance at 10 K in Fig. 5. ξ_{loc} and ξ_{super} of the thickest film is about 2×10^6 and 70 Å, respectively. If the SI transition is induced by breaking the Cooper pair, the SI transition takes place when ξ_{loc} becomes the same length as ξ_{super}. In other words, if ξ_{loc} is larger than ξ_{super} the film behaves as a superconductor, and if ξ_{loc} is smaller than ξ_{super} the film behaves as an insulator. But in practice, the transition takes place at a point where $\xi_{loc} \sim 2\xi_{super}$. From the above result, it is found that relationship of ξ_{super} and ξ_{loc} does not determine the SI transition immediately. Therefore, the SI transition is not induced by the pair-breaking mechanism.

Furthermore, we discovered a difference between the insulator and the superconducting films in terms of the temperature dependence of resistivities in the fermion region. The conductance of superconducting films exhibits logarithmic temperature dependence above 4 K. On the other hand, the conductance of the insulator films above 4 K near the SI transition exhibits $\exp[-(T_0/T)^{1/3}]$ dependence which indicates VRH transport. In Fig. 6 we have plotted conductance vs temperature near the SI transition for the superconducting films and the insulator films that show VRH transport. We found that the films that exhibit VRH transport in the Fermi region become insulators and that the logarithmic temperature-dependent films become global coherent superconductors at low temperature.

We believe that our observed SI transition is a granular one mediated by competition between grain charging and Josephson coupling. Granular systems are susceptible to charging due to the fact that placing an electron on an isolated grain of capacitance C costs an electrostatic energy E_C. Thus, the SI transition is considered as competition between E_C and Josephson-coupling energy E_J, i.e., the system becomes an insulator when $E_C > E_J$ and the system becomes a superconductor when $E_C < E_J$.[18] We roughly calculated E_C and E_J of the films near the SI transition. First, we calculated E_J using the Ambegaokar-Baratoff equation, $E_J = \pi\hbar\Delta/4e^2R_N$,[19] where Δ is the BCS gap energy for Pb and R_N is the normal resistance of the system. E_J is about $\sim 5.6\times10^{-23}$ J ($\sim$4 K) using $\Delta/k_B = 12$ K, $R_N = 10$ kΩ. Next, we calculated E_C using $E_C = q^2/4\pi\epsilon_0\kappa d$ [where $\kappa = \epsilon(1+d/2s)$ and d, s, and ϵ are the size of grains], the spacing of neighboring grains, and the dielectric constant, respectively.[20,21] Our calculated value of E_C is about $\sim 1.2\times10^{-21}$ J ($\sim$80 K), where $q=2e$, $\epsilon=10$ for the glass substrate, $d=30$ nm, and $s=10$ nm using the value measured by AFM. The value of E_C ($\sim$20 K) for $q=e$ is close to the value of generalized activation energy (T_0) from 2D VRH conduction $\exp[-(T_0/T)^{1/3}]$. We claim that the films exhibiting VRH conduction have a gap structure on the order of E_C, that phase fluctuation occurs at low temperature; and that the films exhibiting ln$T$ dependence have no gap structure and become global coherent superconductors at low temperature.

We conclude that the relationship between ξ_{loc} and ξ_{super} does not determine the SI transition, but that the relationship between E_C and E_J determines the SI transition. We suggest that films in the region of $\xi_{\mathrm{super}} < \xi_{\mathrm{loc}} < \sim 2\xi_{\mathrm{super}}$ become Bose glass phase[2] at low temperature since the system can form a Cooper pair in the region $\xi_{\mathrm{super}} < \xi_{\mathrm{loc}}$ and these films have a gap structure which induces VRH conduction in the fermion region.

In our next experiment, we will observe the SI transition at lower temperatures than we did during the present experiment and we will investigate the scaling feature of the Bose glass-vortex glass transition.

In summary, we induced the SI transition by tuned disorder and we estimated the critical resistance which distinguishes superconductors from insulators (20$\sim$23 kΩ). We proved that the relationship between electrical localized length ξ_{loc} and superconducting coherence length ξ_{super} does not determine the SI transition. The SI transition takes place at the point where $\xi_{\mathrm{loc}} \simeq 2\xi_{\mathrm{super}}$. We observed characteristics of electrical transport from a strongly disordered regime to a weakly disordered regime in the fermion region ($T \geqslant 4$ K), and found that all the conductance data above 4 K fit a single universal curve (β function) of a self-consistent theory. Below 4 K, all the conductance data do not fit a single universal curve. An important feature of our findings is the difference of temperature dependence of conductance in the fermion region between insulating and superconducting films. We claim that films in the region of $\xi_{\mathrm{super}} < \xi_{\mathrm{loc}} < \sim 2\xi_{\mathrm{super}}$ become bose glass phase at low temperature; and that these films have a gap structure which induces VRH conduction in the fermion region. We also claim that films exhibiting ln$T$ dependence have no gap structure and become global coherent superconductors at low temperature.

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