

Weak localization and magnetoconductance in percolative superconducting aluminum films

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In order to investigate the crossover from a homogeneous behavior to an inhomogeneous (percolative) one, the temperature T and magnetic field H dependence of the sheet resistance R_{\square} have been measured for two-dimensional granular aluminum films. Fitting the theory to data on magnetoconductance near T_C with use of the diffusion constant $D(T)$ as a fitting parameter, we have obtained the anomalous T -dependent diffusion constant D . From $D(T)$, the electron diffusion index θ , a certain critical exponent in the percolation theory, has been obtained. In the relation $R_{\square} - \theta$, the value of θ varies abruptly near 1.5 k Ω . This behavior suggests the abovementioned crossover is similar to our previous results determined from the T dependence of the upper critical magnetic field. For percolative films at $H=5$ T, we have found the strong R_{\square} dependence of the prefactor α_T in the expression $\sigma = [\alpha_T e^2 / (2\pi^2 \hbar)] \ln T + \sigma_0$. The relation $\alpha_T \propto 1/R_{\square}$ can be explained qualitatively by a model of scaling law for percolation.

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I. INTRODUCTION

Superconductor-insulator mixtures and granular superconductors have been widely investigated in order to clarify the interplay among percolation, Anderson localization, and superconductivity. Especially, transport properties of two-dimensional (2D) granular superconducting films near the superconducting transition temperature T_C have been very interesting from the viewpoint of the superconductor-insulator transition (SIT). Morphology in the specimens, that is, the granularity on the size of grains, connectedness, and the intergrain spacing depend on the preparation method. When the films are produced by quench condensation onto cold substrates, we may obtain disorder on atomic or mesoscopic scales. In these films near percolation threshold, the SIT has mainly been discussed.¹⁻³ On the other hand, when the films are made by evaporation onto room temperature or heated substrates, we obtain granular films with large clusters of macroscopic scale. For these films, homogeneous-inhomogeneous crossover in granular superconductors has been investigated by using percolation models.⁴⁻⁶

For homogeneous dirty superconductors, the superconducting correlation length ξ_S is simply given by $\xi_S^2 = D\tau_{GL}$, where D and τ_{GL} are the diffusion constant and the Ginzburg-Landau relaxation time $\tau_{GL} = \pi\hbar / \{8k_B|T_C - T|\}$, respectively. However, electron diffusion in percolative superconductors is different from that in homogeneous ones. Here, percolative superconductors can be characterized by the condition $\xi_p > \bar{\xi}_S$, where ξ_p and $\bar{\xi}_S$ are the percolation correlation length and the effective superconducting correlation length, respectively. It is known that the diffusion constant D has an anomalous scale dependence $D(L) \propto L^{-\theta}$, where $\theta \approx 0.9$ is the diffusion index.^{7,8} From the relation $\bar{\xi}_S^2 = D(\bar{\xi}_S)\tau_{GL}$, we obtain $\bar{\xi}_S \propto \tau_{GL}^{1/(2+\theta)} \propto |T_C - T|^{-1/(2+\theta)}$. As a result of anomalous T -dependent D , the value of the upper critical magnetic field H_{C2} near T_C for the case of $\xi_p > \bar{\xi}_S$ is expected to show the anomalous temperature dependence⁹ $H_{C2} \propto 1/\bar{\xi}_S^2 = 1/[D(\bar{\xi}_S)\tau_{GL}] = (1 - T/T_C)^{2/(2+\theta)}$. This power-law relation

between H_{C2} and T at temperatures $T < T_C$ near T_C had been observed in three-dimensional (3D) In-Ge, Al-Ge,¹⁰ and 2D Pb.¹¹

We have investigated $H_{C2}(T)$ in 2D granular Al films in the previous work.⁴ The index θ increases steeply near characteristic sheet resistance $R^* \approx 1.5$ k Ω and approaches ≈ 0.9 with increase of R_{\square} . In films with $R_{\square} > R^*$, the excess conductance σ' due to superconducting fluctuations decreases drastically as compared with that estimated from theories for homogeneous films. These increments and decrements can be explained qualitatively by the model based on the scaling law of percolation. We will extend the idea of the percolation model, in order to examine the effects on the weak localization and the magnetoconductance $\Delta\sigma$ at $T > T_C$. A preliminary result of some $\Delta\sigma$ data was given in Ref. 5.

The formula for $\Delta\sigma$ in homogeneous dirty superconductors is given by the theory including the diffusion constant D . Therefore, the temperature dependence of D can also be directly investigated by the analysis of $\Delta\sigma$ near T_C . We expect that $D(T)$ shows the same anomalous temperature dependence as that determined from the analysis of $H_{C2}(T)$ because of the symmetrical temperature dependence of $\tau_{GL}(T)$ with respect to T_C .

As temperature decreases, the interference of a single electron wave scattered by random potentials is enhanced and brings the precursor effect on $\ln T$ dependence of the sheet conductance as $\sigma = [\alpha_T e^2 / (2\pi^2 \hbar)] \ln T + \sigma_0$. It has been reported that the weak localization effect in percolative films is weaker than that expected from the theory for homogeneous films.^{12,13} In the networks with loops larger than the phase relaxation length L_{in} , the interference effect is suppressed. It is considered that α_T decreases with increasing ξ_p beyond L_{in} .^{14,15} However, disorder dependence of the prefactor α_T has not been studied well.

In this paper, report the experimental data for crossovers from a homogeneous behavior to percolative one in the diffusion constant and the electron localization effect. From the experimental data on $D(T)$ determined by the analysis of the magnetoconductance near T_C , we will discuss the R_{\square} depen-

dence of diffusion index θ . As a reason for good correlation of superconducting properties with R_{\square} , we will discuss the effect of quantum percolation in the present superconducting granular films.

II. THEORETICAL BACKGROUND

First, theories for homogeneous 2D films will be explained. Then we will show theories for percolative films.

According to theories for weak electron localization¹³ and Coulomb interaction effects,¹² temperature dependence of conductance due to quantum corrections in normal metallic films is given by

$$\begin{aligned}\sigma' &= [(1-F) + p] \frac{e^2}{2\pi^2\hbar} \ln T = (\alpha_{T,I} + \alpha_{T,L}) \frac{e^2}{2\pi^2\hbar} \ln T \\ &= \alpha_T \frac{e^2}{2\pi^2\hbar} \ln T.\end{aligned}\quad (1)$$

Here $(1-F)$ is a screening term which goes to zero ($F=1$) in the limit for the short-range interaction and goes to 1 ($F=0$) in the limit for long-range interaction; p is the exponent of the temperature term in the inelastic scattering rate $1/\tau_{in} \propto T^p$. Even in superconducting films, σ' can be expressed by Eq. (1) under high magnetic fields which can suppress superconductivity. In the low-temperature region, inelastic scattering is mainly due to electron-electron (e-e) processes. For the films whose thermal length τ_T is longer than the film thickness ($\tau_T = \sqrt{\hbar D/k_B T} > d$), the inelastic scattering rate due to e-e processes has been calculated by Al'tschuler *et al.*¹⁶ and Fukuyama and Abrahams¹⁷ as follows:

$$1/\tau_{ee} = \frac{k_B T}{\hbar} \frac{e^2}{2\pi^2\hbar} R_{\square} \ln \left(\frac{\pi\hbar}{e^2 R_{\square}} \right).\quad (2)$$

There are two contributions of superconducting fluctuations to the conductance of thin films above T_C . The first is the Aslamazov-Larkin contribution $\sigma'_{AL} = e^2/(16\hbar\eta)$, where $\eta = \ln(T/T_C)$.¹⁸ The second is the Maki-Thomson contribution σ'_{MT} .^{19,20} The improved expression by Reizer²¹ is given by $\sigma'_{MT} = [e^2/(2\pi\hbar b_2^{1/2})][\ln(1+B_2) + 2B_2 \ln(1+1/B_2)]$, where $b_2 = [4e^2/(3\pi\hbar)]R_{\square}^N$, and $B_2 = \pi b_2^{1/2}/(8\eta)$, and R_{\square}^N is the sheet resistance at the normal state. Thus, the temperature dependence of R_{\square} in 2D superconductors is given by

$$\begin{aligned}1/R_{\square} &= 1/R_{\square}^N + \sigma'_{AL}(T, T_C) + \sigma'_{MT}(T, T_C, R_{\square}^N) \\ &\quad + [\alpha_T e^2/(2\pi^2\hbar)] \ln(T/T_0).\end{aligned}\quad (3)$$

Magnetoconductance $\Delta\sigma$ is defined as $\Delta\sigma(T, H) = \sigma(T, H) - \sigma(T, 0)$. The $\Delta\sigma$ in 2D dirty superconductors is given by the sum of superconducting fluctuations (AL and MT) and localization terms $\Delta\sigma(T, H) = \Delta\sigma_{AL} + \Delta\sigma_{ML} + \Delta\sigma_L$. The $\Delta\sigma_{AL}$ was calculated by Abrahams and Tsuneto²² as follows:

$$\Delta\sigma_{AL}(H, T) = -\frac{e^2}{16\hbar\eta}(1 - g_{AL}),\quad (4)$$

where

$$g_{AL} = 2 \left(\frac{2\eta}{\lambda_0 a} \right)^2 \left[\frac{\lambda_0 a}{2\eta} + \Psi \left(\frac{1}{2} + \frac{\eta}{\lambda_0 a} \right) - \Psi \left(1 + \frac{\eta}{\lambda_0 a} \right) \right],$$

Ψ is the digamma function, $\lambda_0 = \pi\hbar/8k_B T$, and $a = 4eDH/c\hbar$.

$\Delta\sigma_{MT}$ was calculated by Lopes dos Santos and Abrahams²³ as follows:

$$\Delta\sigma_{MT} = -\frac{e^2}{2\pi^2\hbar} \beta(\eta, \tau_{in}) \left[Y(\tau_{in} a) - Y \left(\frac{\lambda_0}{\eta} a \right) \right],\quad (5)$$

where $Y(x)$ is given by $Y(x) = \ln(x) + \Psi(x)$. When $\eta \ll 1$, $\beta(\eta, \tau_{in})$ is given by $\beta(\eta, \tau_{in}) = (\pi^2/4)[\eta - \pi\hbar/(8k_B T \tau_{in})]$. The $\Delta\sigma_L$ is shown as follows:²⁴

$$\Delta\sigma_L = \frac{e^2}{2\pi^2\hbar} \left[\frac{3}{2} Y(\tau_1 a) - \frac{1}{2} Y(\tau_{in} a) \right],\quad (6)$$

where $1/\tau_1 = 1/\tau_{in} + 4/(3\tau_{SO})$ and τ_{SO} is the spin-orbit scattering time.

The electron-diffusion problem in percolative specimens is treated as a random walk on the random network.⁷ We consider first a particle which has started on the infinite cluster, at one point 0 , and moves during time t by a distance $r(t)$. In the macroscopic limit, when $r \gg \xi_p$, $\langle r^2(t) \rangle = 2Dt$ is expected, where D is a certain diffusion constant for a particle. On the other hand, if a $a < r(t) < \xi_p$ is satisfied, then self-similarity implies that $\langle r^2(t) \rangle \propto t^{2/(2+\theta)}$, where a is a length of the order of the typical grain size. When a scale dependent diffusion coefficient is defined as $\langle r^2(t) \rangle = D(r)t$, the relation $D(r) \propto r^{-\theta}$ is satisfied at the condition $a < r < \xi_p$. This equation will be the starting point of our following discussion on granular superconductors.

For percolative superconducting films, the value of D in the expressions (4)–(6) for $\Delta\sigma$ due to quantum corrections above T_C is not constant but determined on the length range $\bar{\xi}_S$ as mentioned in the Introduction. Therefore, we can consider that the value of $D(\bar{\xi}_S)$ depends on T in the region $\bar{\xi}_S < \xi_p$ as follows:

$$D(\bar{\xi}_S) \propto \bar{\tau}_{GL}^{-\theta(2+\theta)} \propto |1 - T/T_C|^{\theta(2+\theta)}.\quad (7)$$

A model for weak localization phenomena in 2D percolation networks was proposed by Palevski and Deutscher.¹⁴ Within the framework of their model, the percolation network can be divided into two basic parts, if $\xi_p \gg (L_{in}, w)$ (w indicates the width of the channel). One part, on the scale of $L_{in} \cong [D(L_{in})\tau_{in}]^{1/2}$, consists of many loops of different radii smaller than that of the order of L_{in} . The other is the rest of the network which on this scale does not contain loops smaller than L_{in} . The first part can be regarded as a 2D system with an average sheet resistance $R(L_{in})$ on the scale L_{in} . Since the second term becomes negligible, they obtained following relation:

$$\alpha_{T,L}^{\text{inhomo}} = \frac{R(L_{in})}{R_{\square}} \alpha_{T,L}^{\text{homo}},\quad (8)$$

where $\alpha_{T,L}^{\text{homo}}$ and $\alpha_{T,L}^{\text{inhomo}}$ are the coefficient of $\ln(T)$ term in Eq. (1) due to weak localization in homogeneous and inho-

homogeneous films, respectively. It can be considered that the measured sheet resistance R_{\square} of a high resistive film in the percolative region is determined mainly by the change of a coupling strength between grains and depends strongly on the strength. Therefore, it is considered that the length L_{in} can be regarded as almost constant for high resistance films and we obtain following relation:

$$\frac{R(L_{in})}{R_{\square}} = \begin{cases} 1 & (\xi_p < L_{in}) \\ \propto 1/R_{\square} & (\xi_p \gg L_{in}). \end{cases} \quad (9)$$

According to percolation theories, the resistance of discontinuous films varies as $R \propto (p - p_c)^{-\mu}$, when $p (> p_c) \rightarrow p_c$, where p , p_c , and μ are the film coverage, the critical coverage, and the critical conductance exponent, respectively. Since the percolation correlation length diverges as $\xi_p \propto |p - p_c|^{-\nu}$, the variation of the dc resistance can be expressed in terms of ξ_p as $\xi_p \propto R^{\nu/\mu}$.

III. SAMPLE PREPARATIONS

Aluminum films were made by deposition onto glass substrates patterned by photolithography. We prepared films in wide ranges of d and R_{\square} using two different methods.

(1) The single-layer method, aluminum was evaporated in a pressure $1 \times 10^{-5} - 3 \times 10^{-4}$ mb with a deposition rate $0.5 - 1$ Å/s. Except for one 190-Å-thick film, the range of thickness is from 60 to 100 Å.

(2) The multilayer method [almost the same method as that for Film of Oxidized Metal Particles (FOMP) (Ref. 25)]. After deposition of 30–40 Å Al film in a pressure $1 \times 10^{-5} - 3 \times 10^{-5}$ mb with a deposition rate ~ 2 Å/s, an Al film was oxidized in 2.0×10^{-4} mb for 1 min. We repeated this process four times. The range of total thickness is from 120 to 180 Å. By this method, we can prepare sufficiently thick but high resistive films.

We connected a personal computer with the thickness monitor of a quartz crystal oscillator and the digital voltmeter. We recorded *in situ* the resistance $R(d)$ during deposition. It has been found that the relation $R(d) \propto (d - d_c)^{-\mu}$, $\mu = 1.2 - 1.3$ is satisfied in the range $d - d_c < 30$ Å, which is explained by the percolation model. The value of d_c depends on deposition conditions. We have mentioned details of measurement method and d dependence of R_{\square} .⁴ According to the TEM micrograph of thin Al-Al₂O₃ film taken by Laibowitz *et al.*,²⁶ clusters of Al-Al₂O₃ films have a labyrinthlike structure.

IV. EXPERIMENTAL RESULTS AND DISCUSSIONS

A. Magnetoconductance above T_C

The clusters in percolative films are discontinuous. The only maximum cluster expands from end to end. The maximum cluster has multifractal geometry. Because the dimension of the maximum is an intermediate value between 1 and 2 in the scale ($L < \xi_p$), the electron diffusion is weaker than that of two-dimensional film. As T approaches T_C , extending of $\bar{\xi}_S$ is shorter than that of homogeneous 2D films and $D(\bar{\xi}_S)$ is suppressed. It is considered that $D(\bar{\xi}_S)$ above T_C depends on T similarly to that below T_C .

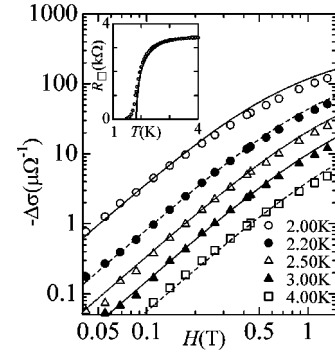


FIG. 1. The H dependence of the magnetoconductance $\Delta\sigma$ for multilayer films with $R_{\square}^N = 3750 \Omega$ at various temperatures. The lines are the best fit of the sum of Eqs. (4)–(6) to the data using $D(T)$ as a fitting parameter at each temperature. The inset shows the temperature dependence of R_{\square} . We obtained the $T_C = 1.81$ K by fitting Eq. (3) (solid line in the inset) to the experimental data.

We measured the resistance of films above T_C in the magnetic field region $H \leq 5$ T. Figure 1 shows the H dependence of magnetoconductance at various temperatures for a typical inhomogeneous film. For analyses of $\Delta\sigma$ with the sum of quantum correction terms due to fluctuation and weak localization effects, we first fit Eq. (3) to data on $R_{\square}(T)$ using α_T and T_C as fitting parameters. The inset in Fig. 1 shows the experimental data on $R_{\square}(T)$, where the line is the theoretical curve (3). Secondly, we analyzed $\Delta\sigma$ using this value of T_C . We fit the sum of Eqs. (4)–(6) to data using D as a fitting parameter, where the inelastic scattering time τ_{in} is calculated from Eq. (2) and τ_{SO} is assumed to be very small compared with τ_{in} at the investigated low temperatures as $T < 5$ K. The lines in Fig. 1 show the theoretical ones obtained from the above procedure. At low magnetic fields, the relation $\Delta\sigma \propto H^2$ is satisfied as expected from theories.

Figure 2 shows temperature dependence of D for films near the crossover region. In the relatively high-temperature regions, the value of D decreases as T decreases. We fit Eq. (7) to data in these regions, regarding θ as a fitting parameter. For the film with the lowest resistance in Fig. 2, the value of D is almost constant in the wide temperature region near T_C .

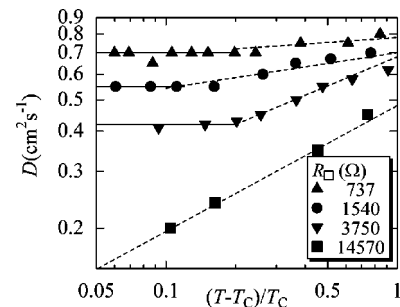


FIG. 2. The $(T - T_C)/T_C$ dependence of the diffusion constant D . The solid lines show the well-known homogeneous behavior $\xi_p < \xi_s$. The broken lines are fitted by Eq. (7) using θ as a fitting parameter.

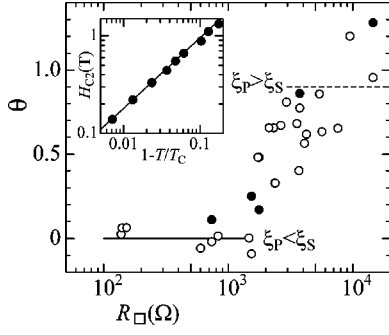


FIG. 3. The R_{\square} dependence of θ . The horizontal axis R_{\square} means R_{\square}^N . The closed and open marks show θ obtained from the analysis of $\Delta\sigma$ above T_C and analysis of $H_{C2}(T)$ below T_C , respectively. The solid and dotted lines show the values of θ for homogeneous and percolative films, respectively. The inset shows the typical data of $H_{C2}-T$. We obtained $\theta=0.77$ fitting the equation $H_{C2}(T)=H_{C2}^0(1-T/T_C)^{2/(2+\theta)}$ to the experimental data.

This behavior is the same as that of homogeneous 2D films. For films with larger value of R_{\square} , it is clear that the crossover occurs from the region with T -independent D to that with T -dependent D . This crossover suggests that the inhomogeneous behavior ($\bar{\xi}_S < \xi_P$) changes to the homogeneous behavior ($\bar{\xi}_S > \xi_P$) as T approaches T_C and the length $\bar{\xi}_S$ increases. Although the temperature corresponding to each crossover does not vary systematically with increases of R_{\square} , such crossover behavior is reasonable because the length ξ_P increases and the temperature region with $\bar{\xi}_S > \xi_P$ decreases as the resistance R_{\square} increases.

On the other hand, the value of D below T_C is obtained from data on H_{C2} using the relation $D=4cK_B/(\pi e)[H_{C2}(T)/(T_C-T)]^{-1}$. In the R - T curve at a constant H , $T_C(H)$ was defined as a temperature at which half of R_{\square}^N was restored.⁴ We had noted that we obtain the similar power law H_{C2} - T relation for percolative films by the use of different criterion $T_C(H)$. Strictly speaking, however, the H_{C2} - T relation, namely, the temperature dependence of H_{C2} near T_C seems to depend on this criterion. This means that there is some ambiguity in the data on $D(T)$ at temperatures very near T_C . Actually, in the H_{C2} - T analysis, we could not observe an evident crossover as shown in Fig. 2. On the contrary, for the analysis of magnetoconductance, we can obtain reliable temperature dependence of D by the use of the only value of T_C at zero magnetic field. Therefore, the $D(T)$ in Fig. 2 indicates the crossover from the inhomogeneous to homogeneous region.

Figure 3 show the R_{\square} dependence of θ . The open and closed marks show the data obtained from analyses of H_{C2} and $\Delta\sigma$, respectively. The inset shows the typical experimental data on H_{C2} - T . We obtained θ from fitting the relation $H_{C2} \propto (1-T/T_C)^{2/(2+\theta)}$ to experimental data. The values of θ are nearly 0 for films with smaller than 1.5 k Ω . This means the scale-independent D for films in the homogeneous region, where the condition $\bar{\xi}_S > \xi_P$ is satisfied. As R_{\square} increases, index θ deviates near 1–2 k Ω from the solid line $\theta=0$ for the homogeneous system. The values of θ in the high R_{\square} region are close to that estimated from the classical

TABLE I. The properties of typical films. N_{layer} is number of layers. The value of T_C was determined from fitting Eq. (3) to the data. θ_1 and θ_2 are obtained from analyses of H_{C2} and $\Delta\sigma$, respectively. α_T is coefficient of $\ln T$ in Fig. 4.

R_{\square}^N (Ω)	N_{layer}	d (\AA)	T_C (K)	θ_1	θ_2	α_T
14570	1	57	1.54	0.95	1.28	0.45
3750	4	155	1.81	0.77	0.86	2.01
1770	4	145	1.99	0.48	0.17	2.15
1540	4	131	1.97	-0.09	0.25	1.92
737	1	75	2.17	-0.02	0.11	1.79

percolation model as shown by the dotted line. The R_{\square} dependence of index θ determined from the analysis of $\Delta\sigma$ is almost the same as that obtained from the analysis of $H_{C2}(T)$ shown by different marks. Some properties of typical films are listed in Table I.

B. Electron weak localization

Figure 4 shows temperature dependence of the sheet conductance σ for various multilayer films at $H=5$ T. Behaviors of single-layer films are discussed in the following section. The conductance of the present films shows the $\ln T$ dependence and superconductivity seems to disappear at measured temperatures. The data for films with the lowest value of conductance show some deviation from the $\ln T$ dependence. However, it cannot be considered that these films are in the strong electron localization regime, because the temperature dependence does not show the relation $R \propto e^{1/k_B T}$ expected for the strong localization. By fitting Eq. (1) to data on σ vs $\ln T$, we determined the coefficient α_T .

Figure 5 shows the R_{\square} dependence of α_T . The value of α_T is almost the constant value=2 for relatively clean films. This value is reasonable, if we take account of the sum of two contributions of weak localization ($p=1$) with inelastic

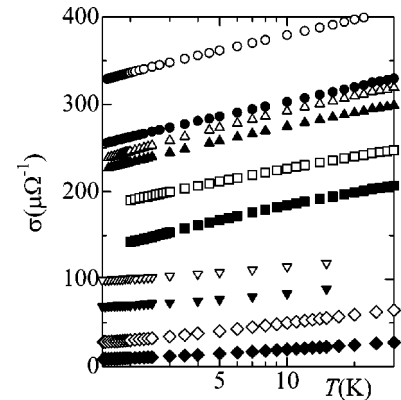


FIG. 4. Temperature dependence of the sheet conductance for various films at $H=5$ T. The values of σ of the present films show the $\ln(T)$ dependence and indication of superconductivity cannot be observed.

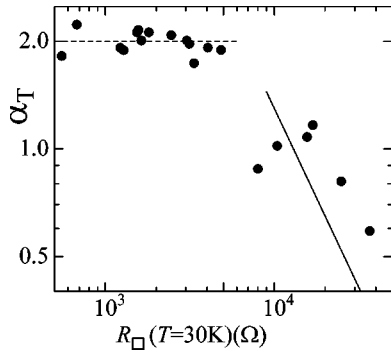


FIG. 5. The R_{\square} dependence of α_T . The value of horizontal axis R_{\square} is defined at $T=30$ K. Values of α_T were determined using Eq. (1) from data in Fig. 4 at temperatures $10 \text{ K} < T < 30 \text{ K}$. The dotted line shows that α_T is independent of R_{\square} for the case $\xi_p < L_{in}$. The solid line shows the relation $\alpha_T \propto 1/R_{\square}$, which is suggested from Eq. (8).

scattering rate due to e-e scattering and Coulomb anomaly ($F=0$) in Eq. (1). The value of α_T decreases when R_{\square} increases beyond 6–8 k Ω . This decrease cannot be explained by the theories of the weak localization and the Coulomb anomaly for the 2D homogeneous system. We consider that this behavior is due to a crossover from homogeneous to inhomogeneous films. For the purpose of the explanation of the α_T – R_{\square} relation, we apply the percolation model¹⁴ for weak localization phenomena in 2D percolation networks to the present data. The fact that the value of α_T is below unity indicates that percolation reduces not only weak localization but also Coulomb anomaly effects. However, there is no theoretical prediction for the percolation effect on the Coulomb anomaly. Assuming that the effect on the coefficient $(1-F)$ in Eq. (1) due to the Coulomb anomaly is similar to weak localization effects, we obtain the relation $\alpha_T \propto 1/R_{\square}(\xi_p \gg L_{in})$ from Eq. (8), which is shown by the solid line in Fig. 5. For clarifying the crossover in the α_T – R_{\square} relation and also the effect of percolation on the Coulomb anomaly, not only theoretical investigations but also detailed experimental studies of temperature and magnetic field dependence of dirty films are necessary.

C. The R_{\square} and d dependence of transport properties

We mentioned that the crossover from homogeneous to inhomogeneous films occurs as R_{\square} increases, where we assumed the following relation $\xi_p \propto R_{\square}^{\nu/\mu}$. We prepared the films whose R_{\square} was almost the same but thickness was different. In Figs. 6(a)–6(d'), we show the d and R_{\square} dependence of θ , H_{C2} , T_C , and α_T , in order to clear the relevant scaling parameter. For data on θ , we showed the result from the analysis of $H_{C2}(T) = H_{C2}^0(1 - T/T_C)^{2/(2+\theta)}$ at temperatures below T_C . The closed and open marks show data for the multilayer films whose thickness is thicker than 100 Å and data for the single-layer films whose thickness is thinner than 100 Å, respectively. By the single-layer method, we obtained exceptionally a thick films whose sheet resistance is relatively high: The data are shown by triangular mark (▲). The mark (■) shows the data for films that consist of ten layers.

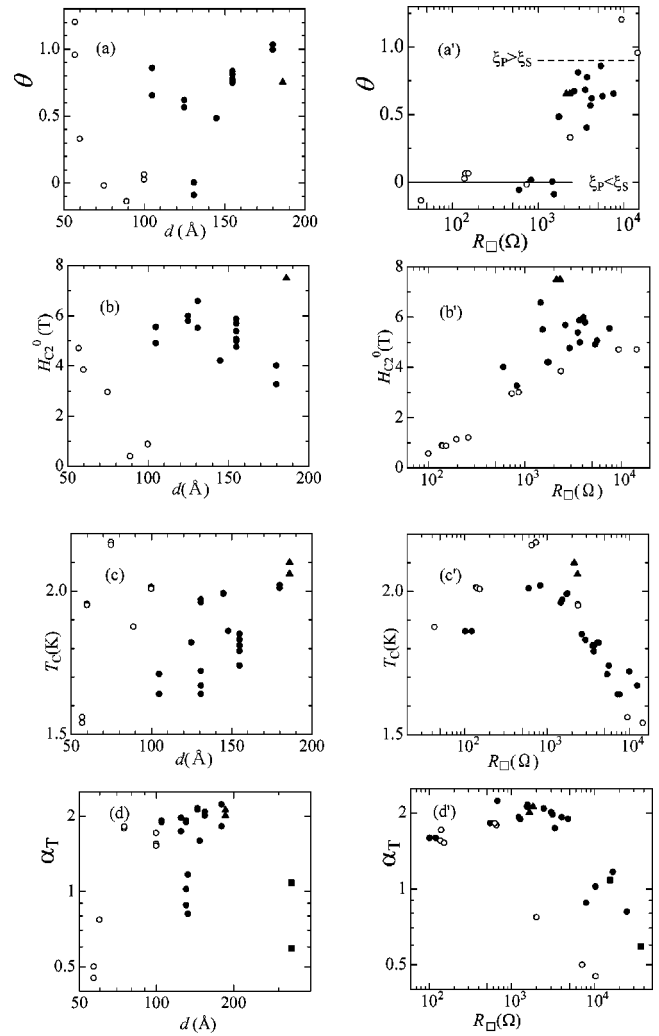


FIG. 6. d dependences of θ , H_{C2}^0 , T_C , and α_T are shown in (a), (b), (c), and (d), respectively. R_{\square} dependences of those properties are shown in (a'), (b'), (c'), and (d'). The horizontal axis R_{\square} in (a'), (b'), (c'), and (d') means R_{\square}^N . In (d') the value of horizontal axis R_{\square} is defined at $T=30$ K. The closed (●) and open (○) marks show the data for the multilayer and single-layer films, respectively. The (▲) and (■) show the data for a thick single-layer and ten-layer films, respectively.

Figures 6(a)–6(d) show d dependence of those properties. Values of θ , H_{C2} , T_C , and α_T for single-layer films seem to correlate with d . If it is assumed that the coverage p is related to d , this behavior is reasonable. However, the data for the thick single-layer film and multilayer films deviate from such behaviors. It means that these values do not depend on d . On the other hand, as known from Fig. 6(a'), it is found that there is no remarkable difference in θ – R_{\square} relation among multilayer films, single-layer films, and even exceptional thick single-layer films. As shown in Figs. 6(b') and 6(c'), the superconducting properties, not only θ but also H_{C2}^0 in the expression of $H_{C2}(T) = H_{C2}^0(1 - T/T_C)^{2/(2+\theta)}$ and T_C , show good correlation with R_{\square} .⁴

As a reason for correlations of superconducting properties with R_{\square} , following quantum percolation model may be considered. We regard the present granular films as random net-

works which consist of grains bound by junctions whose normal tunneling resistance R_N^{micro} varies in place. <> Theories point out that in such a junction with R_N^{micro} , the phase coherence between two constituent superconductors at $T=0$ can establish only when R_N^{micro} is smaller than $h/4e^2$. When such circumstances can be allowed at finite temperatures, it can be considered that there are both Josephson junction and non-Josephson junction in a film. This indicates that the effective percolation coverage, that is, the effective ξ_p in *superconducting* films depends on the ratio of the number of Josephson junctions to that of all. Taking into account that this ratio depends on the macroscopic R_{\square} , it is reasonable that superconducting parameters correlate to the macroscopic R_{\square} . For further investigations of the quantum percolation, not only experimental but also theoretical detailed studies are necessary.

Figure 6(d') shows R_{\square} dependence of α_T . It is found that there is a remarkable difference of α_T between two kinds of films. The value of α_T in single-layer films seems to decrease when the R_{\square} increases beyond 1–2 k Ω . Thick single-layer films show the same behavior as that of multilayer films. It seems that the crossover resistance of single-layer films is smaller than that of multilayer films. It is deduced in the framework of a classical percolation model that α_T is a function of d because ξ_p is longer in thinner films. However, Figs. 6(d) and 6(d') show that α_T is a function of neither thickness d nor sheet resistance R_{\square} . In order to examine whether α_T depends on the number of layers, we made the films which consist of ten layers. The data are shown by closed square marks in Figs. 6(d) and 6(d'). It is shown that α_T is not determined by the number of layers. At the present stage, we have no explanation for this difference of α_T between single-layer and multilayer films.

V. CONCLUSION

For clarifying the crossover from homogeneous to percolative behaviors, we have investigated the superconducting and electron localization properties of evaporated granular aluminum films. In order to change the coupling strength between grains and the area coverage, we have adopted two different kinds of film preparation. One of them is a single-layer method and the other is a multilayer method. We have analyzed the data on $R_{\square}(T, H)$ as follows. (1) $\Delta\sigma = \Delta\sigma_{\text{AL}} + \Delta\sigma_{\text{MT}} + \Delta\sigma_{\text{L}}$ for the magnetoconductance and (2) $\sigma = \alpha_T(e^2/2\pi^2\hbar)\ln T$ for the conductance at a high magnetic field $H=5$ T.

It has been found that the superconducting properties θ , T_C , and H_{C2}^0 correlate much better with the sheet resistance R_{\square} than the film thickness d . This result indicates that the percolation length ξ_p is determined by R_{\square} , but not by d . For this reason, we discussed the quantum percolation model.

In order to confirm the T dependence of diffusion constant $D(T)$ above T_C , we fit the theories to the data on $\Delta\sigma$ with use of $D(T)$ as a fitting parameter. The value of $D(T)$ decreases as T approaches T_C . This result is consistent with the behavior expected from the percolation model. Because $D(T)$ slightly depends on T_C , we cannot refer the quantitative comparison of the strength of $D(T)$ between results from the $\Delta\sigma$ analysis above T_C and H_{C2} analysis below T_C .

Although the value of coefficient α_T is almost constant ≈ 2 in the homogeneous region for both single-layer and multilayer films, the α_T for multilayer films decreases steeply with increase of R_{\square} and shows $\alpha_T \propto 1/R_{\square}$ in the region beyond $R_{\square} \cong 5-8$ k Ω . For the R_{\square} dependence of α_T , however, there is large difference of α_T between thin single-layer films and thick multilayer films. At present, we have no exact explanation for this difference.

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¹A. M. Goldman, *Physica E (Amsterdam)* **18**, 1 (2003); B. G. Orr, H. M. Jaeger, and A. M. Goldman, *Phys. Rev. B* **32**, 7586 (1985).

²A. Frydman, O. Naaman, and R. C. Dynes, *Phys. Rev. B* **66**, 052509 (2002); L. Merchant, J. Ostrick, R. P. Barber, Jr., and R. C. Dynes, *ibid.* **63**, 134508 (2001).

³N. Mason and A. Kapitulnik, *Phys. Rev. B* **65**, 220505 (2002).

⁴K. Yamada, H. Fujiki, B. Shinozaki, and T. Kawaguti, *Physica C* **355**, 147 (2001).

⁵K. Yamada, B. Shinozaki, and T. Kawaguti, *Physica E (Amsterdam)* **18**, 286 (2003).

⁶A. D. Kent, A. Kapitulnik, and T. H. Geballe, *Phys. Rev. B* **36**, 8827 (1987).

⁷P. G. de Gennes, in *Proceedings of the NATO Advanced Study Institute on Percolation, Localization and Superconductivity*, edited by A. M. Goldman and S. A. Wolf, Vol. 109 of *NATO Advanced Studies Institute, Series B: Physics* (Plenum, New York, 1984), p. 83.

⁸D. Stauffer and A. Aharony, *Introduction to Percolation Theory*, 2nd ed. (Taylor and Francis, London, 1994).

⁹S. Alexander and E. Halvy, *J. Phys. (France)* **44**, 805 (1983).

¹⁰A. Gerber and G. Deutscher, *Phys. Rev. B* **35**, 3214 (1987).

¹¹A. Gerber and G. Deutscher, *Phys. Rev. Lett.* **63**, 1184 (1989).

¹²B. L. Altshuler, A. G. Aronov, and P. A. Lee, *Phys. Rev. Lett.* **44**, 1288 (1980).

¹³E. Abrahams, P. W. Anderson, D. C. Licciardello, and T. V. Ramakrishnan, *Phys. Rev. Lett.* **42**, 673 (1979).

¹⁴A. Palevski and G. Deutscher, *Phys. Rev. B* **34**, 431 (1986).

¹⁵M. Aprili, J. Leseur, L. Dumoulin, and P. Nedellec, *Solid State Commun.* **102**, 41 (1997).

¹⁶B. L. Altshuler and A. G. Aronov, *Solid State Commun.* **30**, 115 (1979).

¹⁷H. Fukuyama and E. Abrahams, *Phys. Rev. B* **27**, 5967 (1983).

¹⁸L. G. Aslamazov and A. I. Larkin, *Phys. Lett.* **26A**, 238 (1968).

¹⁹K. Maki, *Prog. Theor. Phys.* **39**, 897 (1968).

²⁰R. S. Thompson, *Phys. Rev. B* **1**, 327 (1970).

²¹M. Yu. Reizer, *Phys. Rev. B* **45**, 12949 (1992).

²²E. Abrahams and T. Tsuneto, *Phys. Rev.* **152**, 416 (1966).

- ²³J. M. B. Lopes dos Santos and E. Abrahams, Phys. Rev. B **31**, 172 (1985).
- ²⁴S. Hikami, A. I. Larkin, and Y. Nagaoka, Prog. Theor. Phys. **63**, 707 (1980).
- ²⁵S. Kobayashi, A. Nakamura, and F. Komori, J. Phys. Soc. Jpn. **59**, 4219 (1990).
- ²⁶R. B. Laibowitz, E. I. Alessandrini, and G. Deutscher, Phys. Rev. B **25**, 2965 (1982).