Granular Effect on Electron Conduction in Discontinuous Metal Films

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The seminal work by Dolan and Osheroff [Phys. Rev. Lett. 43, 721 (1979)], which reported anomalous low-temperature conduction of high-resistivity thin-film metal strips is reanalyzed. It is argued that the observed logarithmic increase of resistance with decreasing temperature in their 3 nm thick Au–Pd strips can be ascribed to the granularity effect on electron conduction in discontinuous metal films. This reanalysis is further supported by the measurements on conducting Pb₄(SiO₂)₁₋ₓ nanogranular films, where x is the volume fraction of Pb.

1. Introduction

The scaling theory of localization by Abrahams et al.¹¹ is one of the cornerstone advances in the field of quantum electron transport in disordered media over the past half century. It states that no metallic states exist in 2D as temperature $T \rightarrow 0$. In particular, for a 2D system in the diffusive regime ($1 \ll k_\text{F}l_e \ll \infty$, with $k_\text{F}$ being the Fermi wavenumber, and $l_e$ the elastic mean free path), the scaling theory predicts a small logarithmic increase of the sheet resistance ($R_\text{C}$) with decreasing $T$, which is known as the weak-localization (WL) effect. In practice, a $\ln T$ increase of $R_\text{C}$ can also arise from the electron–electron interaction (EEI) effect predicted by Altshuler and coworkers,² which usually dominates over the WL effect in homogeneous metal films even in zero magnetic fields.

The first experiment reporting a logarithmic dependence and taken to support the scaling theory was the Dolan–Osheroff’s measurement on ultrathin, discontinuous Au–Pd films, which were prepared by the evaporation method.³ In the literature, it has fervently been presented that the experiment³ worked hand in hand with theory,¹ thus fostering a good understanding of electron wave scattering in random media.⁴–⁹ Notably, Dolan and Osheroff reported a relatively large $\ln T$ increase in $R_\text{C}$ by about 8% as $T$ decreased in a comparatively narrow range from 2.2 to 0.32 K in a ≈3 nm-thick film (denoted film C in the seminal article³). This particular film had a $R_\text{C} = 4600 \text{ }\Omega$ (corresponding to an anomalously large resistivity $\rho \approx 1400 \mu\Omega\text{cm}$) which was more than one order of magnitude higher than that of evaporated continuous Au–Pd films.¹⁰–¹² Dolan and Osheroff noticed the granular structure of the film and attributed the large $R_\text{C}$ value to the tunneling resistance between metallic islands, while they and Anderson et al.¹³ still ascribed the $\ln T$ increase to the WL effect. At the time, the Coulomb effect on electronic conduction in granular metals in the metallic regime was yet to be explored. In this work we aim to clarify the situation and point out that the low-$T$ resistance increase reported in Ref. [3] is primarily due to a many-body effect in inhomogeneous granular metals¹⁴–¹⁶ rather than WL nor Altshuler–Aronov EEI effect.

2. Results and Discussion

As far as inhomogeneous granular effect is concerned, Beloborodov and coworkers¹⁴–¹⁶ have recently studied the Coulomb interaction effect on the conductance of granular metal films above the metal–insulator transition, i.e., $\delta_T = G_T/(2e^2/h) > g_T^\delta$, where $(g_T^\delta) G_T$ is the (dimensionless) tunneling conductance between neighboring grains, $e$ is the electronic charge, and $2\pi h$ is the Planck constant. Here the critical conductance is defined by $g_T^\delta = (1/2\pi d) \ln(E_c/\delta)$, where $d$ is the array dimensionality of the granular metal film, $E_c$ is the charging energy, and $\delta$ is the average level spacing in the constituent metal grains. The theory of Beloborodov and coworkers predicts that in the temperature range $g_T^\delta/k_B \leq T < E_c/k_B$ (which is pertinent to film C, where $k_B$ is the Boltzmann constant), the change in conductivity is logarithmic in $T$ for all array dimensions ($d = 1, 2, \text{ and } 3$), and given by

$$ \Delta \sigma(T) = \sigma(T) - \sigma(T_0) = \frac{\sigma_0}{2\pi d g_T^\delta} \ln \left( \frac{T}{T_0} \right) $$ (1)
where $T_0$ is the reference temperature, and $\sigma_0$ is the conductivity without the Coulomb interaction. Note that this lnT dependence is the consequence of and specific to the granularity effect. This robust logarithmic behavior has not been addressed in Refs. [1,2], which focus on homogeneous disordered systems.

We have carefully reanalyzed and found that the low-T sheet conductance of film C in Ref. [3] can be well described by Equation (1) with a single fitted parameter $g_T \approx 2.3$, see Figure 1. Assuming the Au–Pd grains in film C have a disc shape with $\approx 3$ nm height and $\approx 30$ nm diameter, we estimate the characteristic temperatures to be $T^* \approx \frac{g_T}{\sigma_0} \approx k_B T \approx 0.1 K$ and $E_c/\epsilon_0 \approx 100 K$. Thus, Equation (1) can be applied to describe the data in Figure 1. We further remark that below the characteristic temperature $T^*$, the granularity effect would play a progressively diminishing role, and the Coulomb correction to the conductance should recover that predicted by the EEI effect in Ref. [2] (see also Ref. [17]). Conceptually, this occurs when coherent electron motion on scales larger than the grain size dominates the transport process. That is, the crossover is expected to take place at temperatures below $T^*$, where the thermal diffusion length $l_T^* = \sqrt{D^* h/k_B T}$ becomes longer than the grain size $a$, with $D^*$ being the effective electron diffusion constant of the granular metal.\cite{26}

We estimate $D^* \approx 0.1 \mathrm{cm}^2 \mathrm{s}^{-1}$ in film C. For comparison, we point out that a crossover from the inhomogeneous granularity effect, Equation (1), to the homogeneous Altshuler–Aronov EEI effect has explicitly been observed in $\approx 9$ nm thick Ag$_{0.73}$(SnO$_2$)$_{1-x}$ granular films, where $T^* \approx 15 K$.\cite{27}

The reasons why the WL effect can be ignored in film C are as follows. In 2D homogeneous disordered systems, the WL effect causes a sheet conductance increase given by $\Delta \sigma = \alpha e^2/(2\pi^2 h) \ln(T/T_0)$, which is governed by an electron dephasing time $\tau_\phi \propto T^{-p}$ with $p$ being a temperature exponent. The prefactor $\alpha = 1/(4\pi)$ for weak (strong) spin-orbit interaction in Au–Pd films, $p \to 0$ due to the ”saturation of electron dephasing time” at low temperatures.\cite{22,23} In the presence of the granularity effect, Beloborodov and coworkers\cite{24–26} have shown that the WL effect will be suppressed above a characteristic temperature $g_T^* \delta/\epsilon_0$, which is $0.25 K$ in film C. Thus, the Dolan–Osheroff results\cite{3} cannot be ascribed to the homogeneous nor inhomogeneous WL effect. Besides, if the resistance increase in film C were due to the WL effect, one would expect the film E’ in Ref. [3], which had a $\rho_0$ value about a factor 2 higher than that of film C, also reveal an lnT dependence instead of the observed exponential temperature dependence (see the Table II in Ref. [3]).

We have measured three 0.73 $\mu$m thick co-sputtered granular Pb$_x$(SiO$_2$)$_{1-x}$ films, where $x$ is the volume fraction of Pb, to further test the validity of the prediction of Equation (1). The average grain size of Pb is 5.3 nm, and the granular array is effectively 3D.\cite{29} The main panels of Figure 2a,b show that a logarithmic dependence of conductivity is clearly observed in the two films with $x = 0.57$ and 0.60, respectively. (The lowest measurement temperature is 12 K to keep Pb in the normal state to avoid the superconducting fluctuations.) The straight lines are the theoretical predictions of Equation (1). The fitted values are $g_T \approx 2.0$ and 0.92, compared with $g_T^* \approx 0.40$ calculated for these two films. This conductance decreases with decreasing $T$ cannot be ascribed to the Altshuler–Aronov EEI effect, which would take place below $T^* \approx 0.34$ and $0.74 K$ for the $x = 0.57$ and 0.60 films, respectively. The inset of Figure 2a shows the $T$ dependence of normalized conductivities for three films with $x = 0.51, 0.57,$ and 0.60, as indicated. While the two films with $x = 0.57$ and 0.60 fall close to the metal–insulator transition, the $T$-dependent behavior of the film with $x = 0.51$ exhibits an insulating feature, suggesting that Pb islands are well separated and thus granular hopping conduction\cite{14,23} has to occur at low temperatures. This conjecture is confirmed by the plot shown in the inset of Figure 2b, where the sample resistivity reveals a $\rho \propto \exp(\sqrt{T_0/T})$ dependence below about 40 K, as expected.

Figure 1. Variation of sheet conductance (blue symbols) with temperature for the film C of Ref. [3]. The straight line is the theoretical prediction of Equation (1) for granular metals in the metallic regime.

Figure 2. a,b) Conductivity change as a function of temperature for Pb$_x$(SiO$_2$)$_{1-x}$ granular films, with Pb volume fraction $x$ as indicated. The red straight lines are the theoretical predictions of Equation (1). Inset of (a) shows the normalized conductivities as a function of temperature for three films. The conductivities of the $x = 0.60, 0.57,$ and 0.51 films at 273 K are 1.34, 0.48, and $2.13 \times 10^{-3} \Omega^{-1} \mathrm{cm}^{-1}$, respectively. Inset of (b) shows the stretched-exponential temperature behavior of the insulating film ($x = 0.51$), indicating granular hopping conduction.
3. Conclusion

In summary, we reanalyze and suggest that the $\ln T$ increase of $R$ in Dolan–Osheroff’s discontinuous Au–Pd films originates from the Coulomb interaction effect in granular metals rather than being a manifestation of the scaling theory of localization in the weakly disordered regime. Our measurements on $\text{Pb}_x(\text{SiO}_2)_{1-x}$ films further support the theoretical prediction of the granularity effect on electron conduction in discontinuous metal films.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available on request from the corresponding author. The data are not publicly available due to privacy or ethical restrictions.

Keywords

electrical transport, electron–electron interaction, granular metals, metal–insulator transition, quantum correction, weak-localization effect

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