

行政院國家科學委員會補助專題研究計畫  成果報告  
 期中進度報告

(計畫名稱)

介觀及奈米尺寸結構中電子動力學及相互作用之研究 (II) (2/2)  
Electron Dynamics and Interactions in Mesoscopic and Nanoscale Structures (II) (2/2)

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這篇論文探討金屬鉻顆粒的低溫電子傳輸特性，和穿隧電子態密度 (tunneling density of states)，以實驗結果驗證了最新的理論預測。實驗與理論辦定量的符合，但是更精確的比較仍需要進一步的理論計算。已發表之論文附錄如後：Y. C. Sun, S. S. Yeh, and J. J. Lin, Phys. Rev. B **82**, 054203 (2010)。這是一篇非常詳細的實驗論文。

附件三

## Conductivity and tunneling density of states in granular Cr films

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We have measured the tunneling differential conductances,  $G(V)$ , of four Al/AIO<sub>x</sub>/Cr planar tunnel junctions as well as the conductivities,  $\sigma(T)$ , of the Cr electrodes at liquid-helium temperatures. The Cr electrodes were made to be granular with dimensionless intergrain tunneling conductance spanning from  $g \approx 1$  to  $g \gg 1$ , and the dimensionality of the granular array  $d=3$ . For the samples with  $g \gg 1$ , we found that the measured  $G(V)$  curves display large zero-bias singularities which obey a  $\ln V$  law at low bias voltages ( $\lesssim$  a few millielectron volt) while crossing over to a  $\sqrt{V}$  law at high bias voltages. Simultaneously, the conductivities of the Cr electrodes reveal  $\ln T$  dependence below a characteristic temperature. These results are explained in terms of the recent theory of granular metals. In a sample with  $g \approx 1$ , in addition to the conductivity dependence  $\sigma \propto \ln T$ , we observed a universal scaling behavior of the normalized differential conductance  $[G(V,T) - G(0,T)]/\sqrt{T}$  with the combined parameter  $\sqrt{e|V|}/k_B T$  in a wide temperature interval of 2.5–32 K. This result is not yet understood.

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### I. INTRODUCTION

Granular conductors, which are composite materials of metallic granules and dielectric components, have recently attracted much renewed theoretical attention as tunable systems for addressing mesoscopic physics problems.<sup>1,2</sup> In contrast to disordered “homogeneous systems,” the electronic-transport properties of granular conductors are largely governed by the strength of the intergrain tunneling.<sup>1</sup> Theoretically, a granular conductor is characterized by a number of physical quantities: the mean energy-level spacing in a single granule,  $\delta$ , the dimensionless tunneling conductance between neighboring granules,  $g$  (i.e., the average tunneling conductance between neighboring grains expressed in units of  $2e^2/h$ ), and the single-grain Coulomb charging energy,  $E_c$ . For strong intergrain coupling ( $g \gg 1$ ) and in the not-too-low temperature interval  $g\delta \ll k_B T \ll E_c$  (where  $k_B$  is the Boltzmann constant), charging effects are important yet the quantum-interference weak-localization (WL) effects are suppressed. This unique regime provides a tempting opportunity to probe the electronic conduction properties due to the many-body Coulomb interaction effects in the presence of granularity. The electrical conductivity  $\sigma$  is predicted to obey the law,<sup>2,3</sup>

$$\sigma(T) = \sigma_0 \left[ 1 - \frac{1}{2\pi g d} \ln \left( \frac{g E_c}{k_B T} \right) \right], \quad (1)$$

where  $d$  is the dimensionality of the granular array and  $\sigma_0 = 2(e^2/h)ga^{2-d}$  is the classical conductivity without the Coulomb interaction (i.e., the system conductivity at temperatures  $k_B T \gg E_c$ ), and  $a$  is the radius of the (spherical) grain. It is important to note that, unlike that due to the WL and electron-electron interaction (EEI) effects in weakly disordered homogeneous systems,<sup>4</sup> this  $\sigma \propto \ln T$  law in Eq. (1) is predicted to hold for *all* dimensions since the dimensionality  $d$  only enters the prefactor of the logarithmic correction term. On contrary, the functional form of the tunneling electronic density of states (DOS) is predicted to depend critically on

sample dimensionality. For  $d=3$  (which is pertinent to the present experiment),<sup>2,3</sup>

$$\nu_3(\epsilon) = \nu_0 \left[ 1 - \frac{A}{4\pi g} \ln \left( \frac{g E_c}{\max(k_B T, \epsilon)} \right) \right], \quad (2)$$

where  $\epsilon$  is the tunneling electron energy measured from the Fermi level  $E_F$ ,  $\nu_0$  is the DOS in the absence of Coulomb interaction, and  $A$  is a numerical prefactor. The underlying physics which leads to the conductivity and DOS corrections given in Eqs. (1) and (2), respectively, is due to the presence of local voltage fluctuations between neighboring granules.

Experimentally, it is intriguing that electrical-transport measurements on systems with strong intergrain coupling often revealed  $\ln T$  dependence of *resistivity*,<sup>5,6</sup> rather than of conductivity.<sup>7,8</sup> Therefore, the prediction of Eq. (1) still awaits convincing experimental test. Moreover, there is still no experimental observation in three-dimensional (3D) granular films concerning the electronic DOS predicted by Eq. (2). In the opposite limit of weak intergrain coupling ( $g \ll 1$ ), the conductivity at low temperatures is theoretically and experimentally established to possess the Efros-Shklovskii-type dependence, i.e.,  $\sigma(T) \propto \exp(-\sqrt{T_0/T})$ , where  $T_0$  is a characteristic temperature.<sup>5,7,9–11</sup>

In this work, we have studied four Al/AIO<sub>x</sub>/Cr planar tunnel junctions comprising of granular Cr electrodes with intergrain tunneling conductance spanning from  $g \approx 1$  to  $g \gg 1$ . Moreover, the dimensionality of the granular arrays (the Cr electrodes) was made to have  $d=3$ , insofar as the electronic-transport properties are concerned. Our measured conductivities of the Cr electrodes obey the  $\sigma \propto \ln T$  law, as predicted by Eq. (1). Furthermore, our measured tunneling DOS reveals the  $\ln \epsilon$  law, as predicted by Eq. (2). These results not only verified the recent theory of granular metals<sup>1,2,12</sup> but also clarified the nonmagnetic nature of the “giant” zero-bias conductance dips previously found in many Cr composed tunnel junctions.<sup>13,14</sup> We mention in passing that, previously, the Efros-Shklovskii-type temperature dependence had been found in our nanocontacts formed with

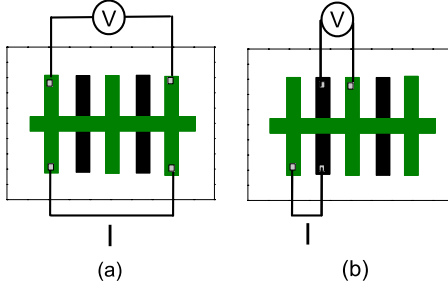


FIG. 1. (Color online) Diagrams depicting (a) four-probe resistance measurements of Cr electrodes and (b) differential conductance measurements of Al/AIO<sub>x</sub>/Cr tunnel junctions. Black strips stand for Al films, and green (gray) strips for Cr electrodes.

granular Cr films, where the  $\sigma \propto \exp(-\sqrt{T_0}/T)$  behavior was observed in the broad temperature interval of 1–100 K.<sup>15</sup> In that case, we experimentally realized the regime  $g \ll 1$ .<sup>16</sup>

This paper is organized as follows. In Sec. II, we discuss our experimental consideration and method for sample fabrication of granular Cr electrodes and Al/AIO<sub>x</sub>/Cr tunnel junctions as well as our measurement procedure. Section III contains our experimental results and discussion. We interpret our measured conductivity and tunneling DOS in terms of the theory of granular metals, and rule out the WL and EEI effects developed for weakly disordered homogeneous conductors to be the origins of our observations. Our conclusion is presented in Sec. IV

## II. EXPERIMENTAL METHOD

The reason for selecting Cr as our electrode material is because Cr films deposited in a vacuum often form granular, rather than uniform and continuous, layers.<sup>15,17,18</sup> For example, a 10-nm-thick Cr film deposited by thermal evaporation on a mica substrate showed a distribution of disk-shaped granules with a diameter of  $\sim$  a few tens of nanometer and a height of  $\approx$  2–6 nm, as was evidenced from atomic force microscopy (AFM) studies.<sup>15</sup> Varying the deposition rate modified the average grain size.<sup>17</sup> Even thermally evaporated in a vacuum having a background pressure as low as  $\sim 1 \times 10^{-6}$  mbar, the surfaces of Cr granules became oxidized and formed thin dielectric layers of CrO<sub>x</sub>.<sup>18</sup> In this work, we carried out our thermal evaporation deposition at a pressure

of  $\sim 5 \times 10^{-6}$  mbar so that our films were guaranteed to form metallic Cr granules separated by thin CrO<sub>x</sub> dielectric layers.

To study the conductivity and the tunneling DOS of granular Cr films, we fabricated four Al/AIO<sub>x</sub>/Cr planar tunnel junctions by using the standard thermal evaporation method as described previously.<sup>19</sup> A set of parallel, relatively clean 0.8 or 1 mm wide and 25 nm thick Al films were first deposited on glass substrates held at room temperature. The surfaces of the as-deposited Al films were subsequently oxidized by utilizing plasma discharge to produce a  $\approx$  1.5–2 nm-thick AIO<sub>x</sub> layer. A long Cr electrode (1 mm wide, and 15–30 nm thick) was then deposited across the parallel AIO<sub>x</sub> coated Al strips to complete the tunnel junction geometries. At the same time, the Cr electrode was attached with leads appropriate for four-probe electrical measurements [see Fig. 1(a) for a schematic diagram]. The resistivities of our Al reference electrodes were typically 13 (16)  $\mu\Omega$  cm at 4 (300) K, corresponding to the product  $k_F \ell \approx 54$  at 4 K, where  $k_F$  is the Fermi wavenumber and  $\ell$  is the electron mean free path. The conductivity of the Cr electrode in each set of junctions was adjusted by varying the mean Cr film thickness and the deposition rate between 0.01 and 1.5 nm/s. To achieve a very low conductivity in the junction D, the Cr film was deposited onto a cold substrate held at liquid-nitrogen temperature, by employing a very low deposition rate of  $\sim 0.01$  nm/s. The tunneling differential conductances,  $G(V, T) = dI(V, T)/dV$ , across the junctions were measured by utilizing the standard lock-in technique, where  $I$  is the tunneling current between the Al and Cr electrodes, and  $V$  is the voltage dropped across the insulating barrier [see Fig. 1(b) for a schematic diagram].<sup>19</sup> Before performing any detailed measurements of  $G(V, T)$  curves, we checked to ensure the high quality of each tunnel junction by measuring the superconducting gap of the clean Al electrode at 0.25 K. (Our Al electrodes became superconducting at  $\approx$  1.8–2 K.) Table I lists the values for the relevant parameters of the four Cr electrodes comprising the tunnel junctions A–D studied in this work.

## III. RESULTS AND DISCUSSION

### A. Temperature dependence of conductivity and differential conductance curves

Figure 2(a) shows the variation in normalized resistivity,  $\rho(T)/\rho(280 \text{ K})$ , with temperature for the four Cr electrodes

TABLE I. Values for relevant parameters of the Cr electrodes in Al/AIO<sub>x</sub>/Cr tunnel junctions.  $A_j$  is the junction area and  $R_j$  is the junction resistance at 300 K.  $t$  is the thickness,  $\rho$  ( $R_{\square}$ ) is the resistivity (sheet resistance) at 2.5 K, and  $N_{\text{Cr,d}}(0)$  is the DOS at the Fermi energy.  $k_F \ell$  was calculated by using the Drude model. The diffusion constant  $D$  was evaluated through the Einstein relation  $\rho^{-1} = N_{\text{Cr,d}}(0)e^2 D$ . The values of  $k_F \ell$  and  $D$  were evaluated for 2.5 K.  $E_c$ ,  $\sigma_0$ ,  $g$ , and  $a$  are defined in Eq. (1). Notice that the values of  $a$  are only listed for reference because our Cr granules are disk-shaped rather than spherical.

Sample	$A_j$ (mm <sup>2</sup> )	$R_j$ (k $\Omega$ )	$t$ (nm)	$\rho$ ( $\mu\Omega$ cm)	$R_{\square}$ ( $\Omega$ )	$N_{\text{Cr,d}}(0)$ (J <sup>-1</sup> m <sup>-3</sup> )	$k_F \ell$	$D$ (cm <sup>2</sup> /s)	$E_c$ (meV)	$\sigma_0$ ( $\Omega^{-1}$ cm <sup>-1</sup> )	$g$	$a$ (nm)
A	$0.8 \times 1.0$	1.0	30	115	38.3	$2.4 \times 10^{47}$	5.1	1.4	5	8690	62	5.5
B	$0.8 \times 1.0$	4.5	25	154	61.6	$1.8 \times 10^{47}$	4.1	1.4	7	6560	42	5.0
C	$1.0 \times 1.0$	11	15	290	193	$1.1 \times 10^{47}$	2.6	1.3	4	3500	10	2.2
D	$0.8 \times 1.0$	4.0	25	5060	2024	$2.8 \times 10^{46}$	0.23	0.28	22	260	0.96	2.8

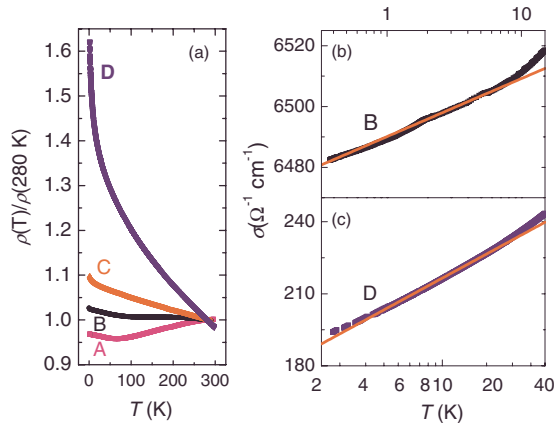


FIG. 2. (Color online) (a) Normalized resistivity,  $\rho(T)/\rho(280 \text{ K})$ , as a function of temperature of the Cr electrodes in junctions A–D, as indicated. Conductivity as a function of temperature of the Cr electrodes in (b) junction B and (c) junction D. The straight solid lines are least-squares fits to Eq. (1).

of our tunnel junctions. Except for the junction A, the resistivities of all other samples monotonically increase with decreasing temperature. However, the amounts of the resistivity rise are much smaller than what would be expected for samples falling deep on the insulating side, where resistivity should show  $\rho \propto \exp(\sqrt{T_0}/T)$  dependence and rapidly increase with decreasing temperature. This result immediately reflects that the intergrain tunneling conductances  $g$  in our Cr electrodes must be large. In particular, a  $\ln T$  dependence of the conductivity was found in all Cr electrodes at liquid-helium temperatures. Figures 2(b) and 2(c) show that the  $\sigma \propto \ln T$  law holds for more than one decade of temperature in the junctions B and D, respectively. Correspondingly, the normalized differential conductances,  $G(V)/G(70 \text{ mV})$ , of these tunnel junctions reveal essentially symmetric dips centered at zero-bias voltage, Fig. 3. It is clearly seen that the differential conductance dips are markedly more pronounced in junctions comprised of more disordered Cr electrodes. The

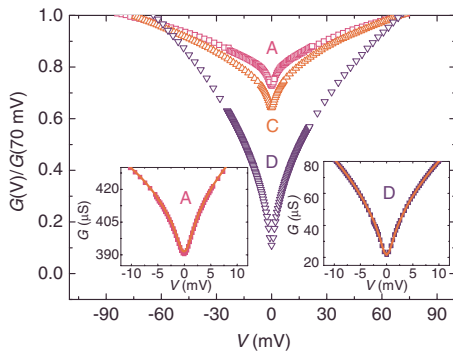


FIG. 3. (Color online) Normalized differential conductance,  $G(V)/G(70 \text{ mV})$ , as a function of bias voltage for junctions A, C, and D, as indicated. Notice that the  $G(V)$  curves are essentially symmetric around zero-bias voltage. Insets: The left (right) inset shows the  $G(V)$  curve of junction A (D) in zero magnetic field (symbols) and in a perpendicular magnetic field of 4 T (solid curve). Notice that the magnetic field causes a negligible change. Data were taken at 2.5 K.

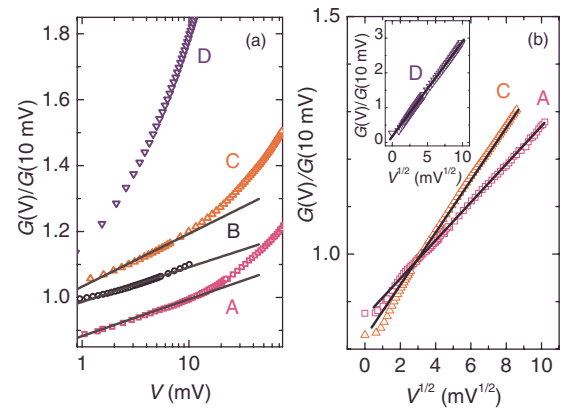


FIG. 4. (Color online) (a) Normalized differential conductance,  $G(V)/G(10 \text{ mV})$ , as a function of bias voltage for junctions A–D, as indicated. The straight solid lines are least-squares fits to Eq. (2). For clarity, the data for the junctions B, C, and D have been vertically shifted up by 0.1, 0.2, and 0.8, respectively. (b)  $G(V)/G(10 \text{ mV})$  versus  $\sqrt{V}$  for junctions A, C, and D, as indicated. The straight solid lines are guide to the eyes. Data were taken at 2.5 K.

relative changes in  $G(V)$  at 2.5 K are  $[G(0) - G(70 \text{ mV})]/G(70 \text{ mV}) = -0.27, -0.36$  and  $-0.89$  in the junctions A, C and D, respectively. These magnitudes of relative change in  $G(V)$  are more than one order of magnitude larger than what would be expected from the EEI effect in weakly disordered homogeneous conductors.<sup>4</sup>

We have measured the differential conductance curves in external magnetic fields applied perpendicular to the junction plane. The left (right) inset of Fig. 3 shows the  $G(V)$  curves of the junction A (D) in zero magnetic field (symbols) and in a magnetic field of 4 T (solid curve). These two insets firmly demonstrate that the magnetic field caused a negligible change ( $\leq 0.05\%$ ) in  $G(V)$ . A negligible magnetic field effect, together with the strong dependence of the magnitudes of  $G(V)$  dips on the level of disorder displayed in the main panel of Fig. 3, strongly suggests that our observed large conductance dips must be associated with some sort of disorder effect in the granular Cr electrodes. In other words, any magnetic origins, which have long been suspected to play an important role in Cr comprised junctions, can be ruled out.<sup>13,14</sup>

Figure 4(a) shows the normalized  $G(V)/G(10 \text{ mV})$  spectra of our Al/AI<sub>2</sub>O<sub>3</sub>/Cr junctions at 2.5 K in the positive bias voltage regime. [Recall that our  $G(V)$  curves are essentially symmetric around the zero-bias voltage.] Specifically, we found that in the junctions A–C,  $G(V)/G(10 \text{ mV}) \propto \ln V$  for  $V \leq 10 \text{ mV}$ , and it crosses over to the  $G(V)/G(10 \text{ mV}) \propto \sqrt{V}$  law at higher bias voltages, Fig. 4(b). On the other hand, in the junction D, the  $G(V)/G(10 \text{ mV}) \propto \sqrt{V}$  law was obeyed from relatively low bias voltages all the way up to a notably high bias voltage of  $\sim 100 \text{ mV}$ , see the inset of Fig. 4(b).

## B. Comparison with theoretical predictions

In this subsection, we compare our experimental results with available theoretical predictions concerning disordered

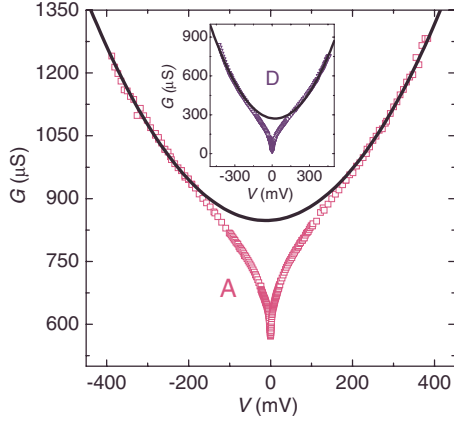


FIG. 5. (Color online)  $G(V)$  spectra of junctions A and D at 2.5 K in a wide bias voltage interval, as indicated. The symbols are the experimental data and the solid curves are parabolic fits. The solid curves in junctions A and D are described by  $G_{\text{para}}(V) = 848 + 0.067V + 0.0028V^2$  and  $G_{\text{para}}(V) = 272 - 0.12V + 0.0026V^2$ , respectively, where  $G_{\text{para}}$  is in microsiemens and  $V$  in millivolt.

conductors. We first explain that our results cannot be described by the conventional EEI effect originally developed for weakly disordered homogeneous conductors by Altshuler and co-workers.<sup>4,20,21</sup> (The WL effects are even less important, because, on one hand, the electron dephasing length is short in granular samples<sup>22</sup> and, on the other hand, our Cr electrodes are magnetic.<sup>23</sup>) Then, we discuss our observations in terms of the recent theory of granular metals formulated by Efetov and Tschersich<sup>2</sup> and Beloborodov and co-workers.<sup>1,3</sup>

### 1. Ruling out the weak-localization and electron-electron interaction effects

In order to understand our  $G(V)$  results in a quantitative manner, we first determine the parabolic background tunneling conductance (PBTC) in our junctions.<sup>24</sup> It is known that a PBTC is characteristic of every metal-insulator-metal tunnel junction at high bias voltages, where the disorder induced suppression in DOS, regardless of its origin, is insignificant.<sup>2,4</sup> Figure 5 shows the  $G(V)$  curves in a wide range of  $V$  for the junctions A and D. It is seen that  $G(V) \propto V^2$  (the solid curves) in the wide interval of  $|V| \approx 200\text{--}400$  mV. This observation confirms that our measured zero-bias conductance dips are superimposed on a PBTC. From this fitted PBTC, we can extrapolate the zero-bias conductance,  $G_{\text{para}}(0)$ , of an as would be ideal Al/AIO<sub>x</sub>/Cr tunnel junction consisting of a “clean” Cr electrode. The extrapolated value of  $G_{\text{para}}(0)$  contains the information about the DOS at  $E_F$  in a clean Cr electrode, because for small bias voltages and at low temperatures,  $G_{\text{para}}(0)$  can be approximated by  $G_{\text{para}}(0) = PN_{\text{Al}}(0)N_{\text{Cr,c}}(0)$ , where  $P$  is the electron tunneling rate (which depends on the barrier height and width), and  $N_{\text{Al}}(0)$  and  $N_{\text{Cr,c}}(0)$  are the DOS at  $E_F$  in the Al and clean Cr electrodes, respectively.<sup>25</sup> The DOS at  $E_F$  in our granular and *disordered* Cr electrode,  $N_{\text{Cr,d}}(0)$ , can then be evaluated through the relation  $N_{\text{Cr,d}}(0) = N_{\text{Cr,c}}(0) \times [G(0)/G_{\text{para}}(0)]$ , where the measured zero-bias conduc-

tance  $G(0) = PN_{\text{Al}}(0)N_{\text{Cr,d}}(0)$ . With the literature value of  $N_{\text{Cr,c}}(0) = 3.5 \times 10^{47} \text{ J}^{-1} \text{ m}^{-3}$  in clean Cr metal,<sup>26</sup> our extracted magnitudes of  $N_{\text{Cr,d}}(0)$  are listed in Table I.

For two-dimensional (2D) weakly disordered homogeneous conductors, Altshuler and co-workers<sup>4,20</sup> have predicted that the EEI effect causes a minute suppression in the DOS around zero-bias voltage

$$\frac{\delta N_2(\epsilon)}{N_2(0)} = \lambda_2 \frac{e^2 R_{\square}}{8\pi^2 \hbar} \ln \left[ \frac{\epsilon}{\hbar D} \left( \frac{t}{2\pi} \right)^2 \right], \quad (3)$$

where  $N_2(0)$  is the measured DOS at  $E_F$ ,  $\lambda_2$  is the electron-electron coupling constant in 2D,  $R_{\square}$  is the sheet resistance,  $D$  is the electron diffusion constant, and  $t$  is the sample thickness. The tunneling electron energy  $\epsilon$  is measured relative to  $E_F$ . For a given film thickness, as  $|V|$  increases, the effective dimensionality of the EEI effect will cross over from 2D to 3D at  $|V| \geq V_c \approx (4\pi^2 \hbar D) / (et^2)$ .<sup>27</sup> In 3D, the EEI correction to the DOS is given by<sup>4,21</sup>

$$\frac{\delta N_3(\epsilon)}{N_3(0)} = \frac{\lambda_3 \sqrt{\epsilon}}{4\sqrt{2}\pi^2 (\hbar D)^{3/2} N_3(0)}, \quad (4)$$

where  $N_3(0)$  is the measured DOS at  $E_F$ , and  $\lambda_3$  is the electron-electron coupling constant in 3D.

At first glance, since our junctions A and B are nominally weakly disordered ( $k_F \ell \approx 4\text{--}5$ ), one might attempt to attribute our  $G(V) \propto \ln V$  results to the 2D EEI effect. However, such an interpretation can be ruled out as follows. By comparing our measured  $\ln V$  dependence of  $G(V)$  in the junction A with the prediction of Eq. (3), we obtained a value  $\lambda_2 \approx 448$ . This value is one order of magnitude larger than the theoretical prediction of  $\lambda_2 = \ln[(\hbar D \kappa^4 t^2) / (4\pi^2 \epsilon)] \approx 43$ , where  $\kappa = (me^2 k_F t) / (2\pi^2 \hbar^2 \epsilon_0)$ , and  $\epsilon_0$  is the permittivity of the vacuum<sup>4</sup> (we took a typical electron energy  $\epsilon = 5$  meV). Furthermore, in the high bias voltage regime, a comparison of our measured  $G(V) \propto \sqrt{V}$  result with Eq. (4) yielded a value of  $\lambda_3 \approx 88$ , which is also far higher than the theoretical prediction of  $\lambda_3 \approx 2$ .<sup>4</sup> Therefore, our observed giant  $G(V)$  dips cannot be due to the EEI effect in weakly disordered homogeneous conductors. Definitely, the granularity in the structure of our Cr electrodes must play an important role,<sup>15,17,18</sup> as to be addressed below.

### 2. Logarithmic temperature dependence of conductivity

Let us discuss that our results can be satisfactorily, but not fully, interpreted in terms of the recent theory of granular metals. We have fitted our measured  $\sigma \propto \ln T$  results with Eq. (1). To carry out the least-squares fits, we assume a value of the charging energy  $E_c \approx 10k_B T^*$ , where  $T^*$  is the temperature below which the  $\sigma \propto \ln T$  law holds. Taking this value of  $E_c$  and the sample dimensionality  $d=3$  (because our Cr granules are disk-shaped with a height of  $\approx 0.5\text{--}2.5$  nm, which is much smaller than our mean film thickness of 15–30 nm), we have extracted the values of the parameters  $\sigma_0$  and  $g$  in our samples (see Table I). It should be noted that the extracted  $\sigma_0$  and  $g$  values are insensitive to the choice of  $E_c$  value, because  $E_c$  appears in the argument of a logarithmic function.<sup>28</sup> Inspection of Table I indicates that in the junctions A–C we obtained  $g \gg 1$ . This result is in good consistency with the

prerequisite for Eq. (1) to be applicable. On the other hand, our extracted value of  $g \approx 1$  in the junction D implies that this sample falls marginally inside the regime of validity of Eq. (1). We notice that Eq. (1) was formulated by considering a periodic cubic array of uniformly sized grains and neglecting dispersion of the intergrain tunneling conductance,<sup>1</sup> while our samples contained random arrays of varying-sized, disk-shaped granules.<sup>15</sup> Therefore, a close quantitative comparison of our (and other groups'<sup>8,29,30</sup>) experiment with theory is not possible at this stage.

Another important feature of the predictions of the theory of granular metals is that Eq. (1) should be valid at any magnetic field. This is indeed confirmed by our experiment. We have measured  $\sigma(T)$  of the Cr electrode in the junction A between 2 and 20 K in both zero magnetic field and in a perpendicular magnetic field of 4 T. The measured values are the same to within our experimental uncertainty. On contrary, the WL effect, if any exists, should be very sensitive to and suppressed by even a small magnetic field.<sup>23</sup> If the EEI effect were responsible, we should then have observed a  $\sqrt{T}$ , but not a  $\ln T$ , dependence in this sample in this temperature interval.<sup>31</sup> Therefore, both the WL and EEI effects are irrelevant to our observations in Figs. 2(a)–2(c).

The mean energy level spacing  $\delta$  in our Cr granules may be evaluated as follows. We have carried out AFM studies of a film deposited under conditions similar to those used for the fabrication of the junction B. We found that the Cr film formed a granular structure consisting of disk-shaped grains of  $\approx 60 \pm 20$  nm in diameter and  $\approx 1.5 \pm 1$  nm in height, along with a few larger aggregations. By taking an average diameter of  $\sim 60$  nm and an average height of  $\sim 1.5$  nm, we obtain an estimate of  $\delta = 1/N_{\text{Cr,d}}(0)\bar{V} \approx 2 \mu\text{eV}$ , where  $\bar{V}$  is the average granule volume. This  $\delta$  value in turn suggests a characteristic temperature  $T_B \approx g\delta/k_B \approx 1$  K above which Eq. (1) is expected to apply. Experimentally, the  $\sigma \propto \ln T$  law in our junction B is observed in the temperature interval 0.3–7 K, see Fig. 2(b). This degree of agreement is satisfactory, considering that the evaluations of parameters in a granular sample unavoidably involve large uncertainties.

Using our estimated values of  $E_C \approx 6$  meV (see Table I) and  $\delta \approx 2 \mu\text{eV}$ , we obtained the ratio  $E_C/\delta \approx 3 \times 10^3$ . This ratio suggests the existence of a broad range for logarithmic corrections to conductivity. However, even under such circumstances, Feigel'man *et al.* have theoretically shown that a simple  $\sigma \propto \ln T$  law should still hold in a wide range of temperature.<sup>32</sup> This prediction is confirmed by the present experiment.

### 3. Differential conductance curves and tunneling density of states

Tuning to the differential conductance curves, we discuss the crossover behavior of  $G(V)$  from the  $\ln V$  to  $\sqrt{V}$  dependence. This occurs at a characteristic bias voltage of  $V_c \approx E_C/e \approx 10k_B T^*/e$ . Theoretically, the application of Eq. (2) requires the condition  $\max(T, \epsilon) \leq E_C$  to be satisfied. That is, at low temperatures such that  $k_B T < \epsilon$ , the  $\nu_3(\epsilon) \propto \ln \epsilon$  law is predicted for the regime  $\epsilon \leq E_C$ . On the other hand, the variation in  $\nu_3$  with  $\epsilon$  in the opposite limit ( $\epsilon \geq E_C$ ) has not been calculated. Experimentally, in the junctions A–C, we ob-

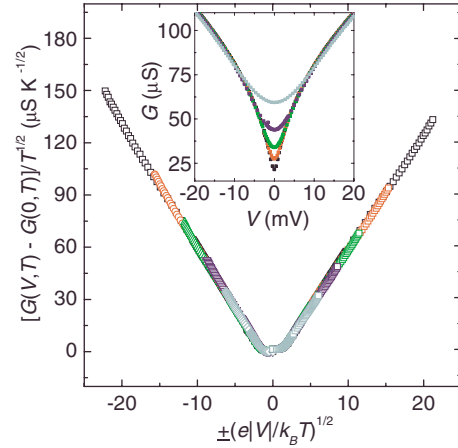


FIG. 6. (Color online) Normalized differential conductance,  $[G(V, T) - G(0, T)]/\sqrt{T}$ , as a function of the combined parameter  $\sqrt{|eV|/k_B T}$  for the junction D at five measurement temperatures. Notice that the data points collapse closely. Inset: unscaled  $G(V)$  versus bias voltage  $V$  at (from bottom up) 2.5, 5.0, 9.0, 16, and 32 K.

served  $G(V) \propto \ln V$  in the low bias voltage regime ( $V \leq V_c \approx E_C/e \approx$  a few to  $\sim 10$  mV). This is qualitatively in line with the prediction of Eq. (2). However, a close comparison with theory cannot be made at this stage because the numerical prefactor  $A$  in Eq. (2) was calculated for the case when the logarithmic term is much smaller than 1.<sup>2</sup> Quantitatively, the magnitudes of the  $G(V)$  dips we observed are much larger than that predicted by Eq. (2). On the other hand, our  $g$  values are larger than the critical intergrain tunneling conductance  $g_C = (2\pi d)^{-1} \ln(E_C/\delta)$  ( $\approx 0.4$ , using the above  $E_C/\delta$  value).<sup>3</sup> Therefore, we do not expect to find a “hard” gap in our samples.<sup>12</sup> Our  $G(V) \propto \sqrt{V}$  results in the high bias voltage regime ( $V \geq V_c$ ) also have to await a future theoretical explanation.<sup>33</sup>

Finally, in the junction D, we did not observe any  $G(V) \propto \ln V$  dependence even at relatively low bias voltages, which may be due to the fact that  $g \approx 1$  in this sample and thus Eq. (2) is marginally applicable. [However, recall that we found the  $\sigma \propto \ln T$  behavior, as predicted by Eq. (1).] In fact,  $G(V) \propto \sqrt{V}$  was observed in this sample in a wide range of  $|V| \approx 1$ –100 mV at 2.5 K, Fig. 4(b). Furthermore, we found that in this particular sample, the scaled differential conductance  $[G(V, T) - G(0, T)]/\sqrt{T}$  versus the combined parameter  $\sqrt{|eV|/k_B T}$  for different measurement temperatures between 2.5 and 32 K collapse closely onto a single curve, Fig. 6. This result strongly suggests the existence of a universal scaling function in the  $g \approx 1$  regime. That is, there exists a function  $f$  such that  $G(V, T) - G(0, T) = \sqrt{T} \times f(\sqrt{|eV|/k_B T})$ , where  $f$  should depend on the combined parameter  $\sqrt{|eV|/k_B T}$ , instead of depending independently on  $eV$  or  $k_B T$ . For comparison, the inset of Fig. 6 shows the unscaled  $G(V)$  versus bias voltage  $V$  at five measurement temperatures. Previously, a universal scaling behavior of differential conductance has been theoretically predicted for 3D weakly disordered homogeneous conductors (Refs. 21 and 27). Our observation of Fig. 6 demonstrates that a universal scaling phenomenon also exists in the granular case. This



issue deserves further theoretical investigations.

#### IV. CONCLUSION

We have measured the conductivities  $\sigma(T)$  in granular Cr electrodes and the differential conductances  $G(V)$  in Al/AlO<sub>x</sub>/Cr tunnel junctions at liquid-helium temperatures. In samples with dimensionless intergrain tunneling conductances  $g \gg 1$ , we found  $\sigma \propto \ln T$  and  $G(V) \propto \ln V$  at low bias voltages. These results are satisfactorily understood in light of the recent theory of granular metals. A crossover of  $G(V)$  from the  $\ln V$  to  $\sqrt{V}$  dependence was observed at high bias voltages. In a sample with  $g \approx 1$ , we found  $\sigma \propto \ln T$  and  $G(V) \propto \sqrt{V}$  in a wide bias voltage interval. Moreover, the normalized differential conductance  $[G(V, T) - G(0, T)]/\sqrt{T}$  reveals a universal scaling behavior with the combined param-

eter  $\sqrt{e|V|/k_B T}$  in a wide range of temperature. This last observation requires a further theoretical explanation. Finally, we would like also to note that, while the theory of granular metals considers a periodic array of uniformly sized grains, in real samples one often has some distribution in granule size. The effect of such size distribution on our results in the present study has yet to be fully addressed.

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in essentially the same temperature interval shown in Fig. 2(b) [Fig. 2(c)]. However, the measured resistance rise is more than a factor of 3 as could be expected from the 2D EEI and WL effects, taking the sheet resistance listed in Table I into calculation.

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