

# Electron-phonon-impurity interference effect in disordered $\text{Au}_{56}\text{Pd}_{44}$ and $\text{IrO}_2$ thick films

S. S. Yeh<sup>1</sup> and J. J. Lin<sup>1,2</sup><sup>1</sup>*Institute of Physics, National Chiao Tung University, Hsinchu 30010, Taiwan*<sup>2</sup>*Department of Electrophysics, National Chiao Tung University, Hsinchu 30010, Taiwan*

Jing Xiunian and Zhang Dianlin

*Institute of Physics, Chinese Academy of Sciences, Beijing 100080, China*

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We have fabricated a series of  $\text{Au}_{56}\text{Pd}_{44}$  thick films with a wide range of residual resistivity  $\rho_0$  varying from 40 to 280  $\mu\Omega$  cm. The resistivities of these films were measured between 15 and 300 K. We found that at temperatures below about  $0.1\theta_D$  ( $\theta_D$  is the Debye temperature), the interference mechanism between the elastic electron scattering and electron-phonon scattering (the electron-phonon-impurity interference effect) contributes significantly to the measured resistivities. Our results support the current theoretical idea that this interference-mechanism-induced resistivity varies with  $\rho_0 T^2$ , where  $T$  is the temperature. Similar observation has also been made in disordered, conducting transition-metal oxide  $\text{IrO}_2$  thick films.

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## I. INTRODUCTION

The magnitude and temperature behavior of the electrical resistivity  $\rho(T)$  in metals are extremely difficult to calculate quantitatively. In the most standard model for the electrical transport in metals,  $\rho(T)$  is presumed to comprise of two terms: the residual resistivity  $\rho_0$  due to electron scattering from random potential and the temperature dependent part of resistivity due to scattering from lattice vibrations (phonons). The latter contribution is known as the Bloch-Grüneisen term, denoted by  $\rho_{\text{BG}}(T)$ . In Matthiessen's rule, these two contributions are expected to be independent of each other, and thus  $\rho(T) = \rho_0 + \rho_{\text{BG}}(T)$ . However, it has been known for years that deviations from the Matthiessen's rule exist in many real conductors.<sup>1</sup> Recently, it becomes clear that such deviations can be particularly noticeable in impure conductors. Theoretically,<sup>2</sup> it was proposed that the interference mechanism between the elastic electron scattering and the electron-phonon scattering would lead to an additional contribution to the resistivity. The contribution due to this so-called electron-phonon-impurity interference effect can dominate over the Bloch-Grüneisen term especially at temperatures below about  $0.1\theta_D$ , where  $\theta_D$  is the Debye temperature. The measured resistivity of a *disordered* metal should then be written as<sup>2</sup>

$$\rho(T) = \rho_0 + \rho_{\text{int}}(T) + \rho_{\text{BG}}(T), \quad (1)$$

where  $\rho_{\text{int}}$  is due to the electron-phonon-impurity interference mechanism, which was not considered in the Matthiessen's rule.

In this work, we shall concentrate on the temperature regime above 15 K where the corrections to the residual resistivity due to the weak-localization and electron-electron interaction effects<sup>3</sup> are essentially negligible (or, the minor contributions of which can be safely subtracted from the measured resistivities).

## II. THEORY

In disordered metals, the electron-phonon interaction is mainly due to two processes. The first process is the usual "pure" electron-phonon scattering similar to that in clean metals. The second process is due to the *inelastic* electron scattering from *vibrating* impurities. As a consequence, a variety of interference processes are generated due to the coexistence of the elastic electron scattering, the "pure" electron-phonon scattering and the inelastic electron scattering. Reizer and Sergeev<sup>2</sup> took into account of all possible electron scattering channels and calculated the contribution from those interference processes to the resistivity ( $\rho_{\text{int}}$ ). In particular, they found that  $\rho_{\text{int}}$  scales with the residual resistivity  $\rho_0$  of the disordered sample and varies with the square of temperature, i.e.,  $\rho_{\text{int}} \propto \rho_0 T^2$  at low temperatures of about  $T < 0.1\theta_D$ . This multichannel interference contribution to resistivity has recently been tested in experiments using thin metals films<sup>4-6</sup> and metal-dielectric composite nanowires.<sup>7</sup> Less extensive studies have been performed on bulk samples.<sup>8,9</sup> On using bulk samples, one of the advantages is that the phonon spectra would be definitely three dimensional;<sup>10</sup> three dimensionality of phonons is a criterion that was originally assumed in the theoretical calculations of Ref. 2.

Under the conditions that  $q_{l,t}l > 1$  and  $\rho_{\text{int}} < \rho_0$  (where  $q_{l,t}$  is the wave number of the longitudinal and transverse thermal phonons, respectively, and  $l$  is the electron mean free path), the correction to the resistivity due to the electron-phonon-impurity interference effect has been explicitly calculated by Reizer and Sergeev. The clean limit criterion of  $q_{l,t}l > 1$  is equivalent to  $q_{l,t}l \approx k_B T l / (\hbar u_{l,t}) > 1$ , where  $u_{l,t}$  is the longitudinal and transverse sound velocity, respectively. Therefore, the clean-limit criterion is satisfied when the measuring temperature is higher than a characteristic temperature of  $T_{l,t}^* \approx \hbar u_{l,t} / (k_B l)$ . The Reizer-Sergeev result for the interference-mechanism-induced resistivity is given by<sup>2</sup>

$$\rho_{\text{int}}(T) = BT^2 \rho_0 \left( \frac{6}{\pi^2} \right) \int_0^{\theta_D/T} \left[ \frac{x^2 e^x}{(e^x - 1)^2} - \frac{x}{e^x - 1} \right] dx, \quad (2)$$

where

$$B = \left[ 2 \left( \frac{u_l}{u_t} \right) \beta_l + \left( \frac{\pi^2}{16} - 1 \right) \beta_l \right] \frac{2\pi^2 k_B^2}{3E_F \rho_F u_l}. \quad (3)$$

Here  $\beta_{l,t}$  is the coupling constant of electrons with longitudinal and transverse phonons, respectively,  $E_F$  ( $p_F$ ) is the Fermi energy (momentum), and  $k_B$  is the Boltzmann constant. At low temperatures ( $T < 0.1\theta_D$ ), the integral in Eq. (2) approaches  $\pi^2/6$ , and thus the temperature dependence of  $\rho_{\text{int}}$  reduces to a simple power law

$$\rho_{\text{int}}(T) = BT^2 \rho_0. \quad (4)$$

Notice that  $d(\rho_{\text{int}}/\rho_0)/dT^2 = B$  is a constant for a given material, being independent of the amount of disorder contained in the sample.

In the jellium model with the Bohm-Staver relation for the sound velocity,<sup>2,11</sup>  $\beta_l = 0.5$  and the two coupling constants are related to each other by  $\beta_l/\beta_t = (u_l/u_t)^2$ . The electron-phonon coupling constants can be explicitly written as  $\beta_{l,t} = (\frac{2}{3}E_F)^2 N(0)/(2\rho_m u_{l,t}^2)$ , where  $\rho_m$  is the mass density, and  $N(0)$  is the electronic density of states (both spins) at the Fermi level. It should be noted that in usual metals,  $u_l/u_t \approx 2-3$ . Thus, inspection of Eq. (3) indicates that the contribution from the interactions of the electrons with the *longitudinal* phonons (the second term in the square parentheses) is *negligibly small* compared with the interactions of the electrons with the *transverse* phonons (the first term in the square parentheses).

It is worth mentioning that, by the same token, current theories<sup>12,13</sup> and experiments<sup>14</sup> have also established that the total electron-phonon scattering rate in impure conductors is dominated by the interactions of electrons with *transverse* rather than with longitudinal phonons.

The contribution of the “pure” electron-phonon scattering to the resistivity in a disordered metal has been calculated by Altshuler.<sup>15</sup> His result at temperatures  $T > (a/l)\theta_D$  (where  $a$  is the lattice spacing) is similar to that given by the Bloch-Grüneisen law<sup>6,16</sup>

$$\rho_{\text{BG}}(T) = \beta_{\text{BG}} T \left( \frac{T}{\theta_D} \right)^4 \int_0^{\theta_D/T} \frac{x^5 dx}{(e^x - 1)(1 - e^{-x})}, \quad (5)$$

where

$$\beta_{\text{BG}} = \frac{\pi \beta_l \tau k_B^5}{2 \hbar \rho_F^4 u_l^4} \rho_0 \theta_D^4. \quad (6)$$

Here  $\tau = l/v_F$  is the electron elastic mean free time, and  $v_F$  is the Fermi velocity.

Comparison of the theoretical predictions Eqs. (2) and (5) reveals that the Bloch-Grüneisen term dominates at high temperatures while the electron-phonon-impurity interference effect dominates at low temperatures of about  $T < 0.1\theta_D$ . Quantitative comparison of these two contributions in our samples is presented in Figs. 2 and 4.

### III. EXPERIMENTAL METHOD

Prototypical disordered Au<sub>56</sub>Pd<sub>44</sub> alloy was selected for this study for the following reasons. The substitutional disorder and the structural disorder for the present concentration ratio of the material provide enormous scattering centers and the level of disorder can be “tuned” by adjusting the dc-sputtering deposition rate,<sup>17</sup> resulting in a wide range of residual resistivity  $\rho_0$  [ $=\rho(10\text{ K})$ ] from  $\approx 40$  to  $\approx 280\ \mu\Omega\text{ cm}$  in our case, yet the alloy is still at the Au-rich side where the electronic structure at the Fermi level is not much complicated by the *d*-band.<sup>18</sup> It is well known that Au and Pd form perfect fcc solid solution and it would be safe to treat the alloy as an isotropic material. The details of the sample preparation were described in Ref. 17. To ensure a three-dimensional phonon spectrum as well as to minimize the weak-localization and electron-electron interaction effects at liquid-helium temperatures, we have made our films sufficiently thick ( $\geq 0.50\ \mu\text{m}$ ) in this study.

In a quantitative comparison of the theory with experiment, the Debye temperature  $\theta_D$  plays an important role in both the  $\rho_{\text{int}}$  and  $\rho_{\text{BG}}$  terms. Therefore, instead of treating  $\theta_D$  as an adjusting parameter in Eqs. (2) and (5), we have experimentally extracted the value of  $\theta_D$  from specific heat,  $C$ , measurements between 0.4 and 40 K. We then evaluated  $\theta_D$  according to the relation  $C/T = \gamma + \alpha T^2$ , where  $\gamma$  and  $\alpha$  are material dependent parameters.<sup>19</sup> From the value of  $\alpha$ , we obtained  $\theta_D \approx 240\text{ K}$  for our Au<sub>56</sub>Pd<sub>44</sub> alloy. This value is reasonable, compared with the  $\theta_D(\text{Au}) \approx 165\text{ K}$  and  $\theta_D(\text{Pd}) \approx 274\text{ K}$ .<sup>20</sup> Since the Debye temperature determined from resistance measurement and that from specific heat measurement differs only slightly,<sup>16</sup> we have fixed  $\theta_D$  to this value in Eqs. (2) and (5) in our data analysis. (We have also treated  $\theta_D$  as a free parameter and found a similar result.)

### IV. RESULTS AND DISCUSSION

#### A. Au<sub>56</sub>Pd<sub>44</sub> thick films

Figure 1 plots the phonon dependent part of the resistivity,  $\Delta\rho = \rho - \rho_0$ , as a function of temperature for a series of Au<sub>56</sub>Pd<sub>44</sub> thick films with various amounts of disorder as indicated in the caption to Fig. 1. According to Matthiessen’s rule,  $\Delta\rho$  should be independent of disorder, and thus a plot of  $\Delta\rho$  versus temperature for all samples should collapse on a same curve. Obviously, this is not the case found in Fig. 1. On the contrary, Fig. 1 reveals that the temperature variation of  $\Delta\rho$  is strongly sample dependent, being larger in films with higher  $\rho_0$ . This observation clearly implies that disorder must play a crucial role in determining the temperature behavior of the measured resistivity.

To illustrate the importance of the electron-phonon-impurity interference term, we plot in Fig. 2 the variation of  $\Delta\rho/\rho_0$  with temperature in double logarithmic scales for three representative samples. The measured data for each film were least-squares-fitted to Eqs. (1), (2), and (5), and the relevant parameters were determined.<sup>21</sup> It is clearly seen that the theoretical predictions (the solid curves) can well describe the experimental results. We notice that at low temperatures of  $T \approx 0.1\theta_D$ , the measured resistivity is a factor of

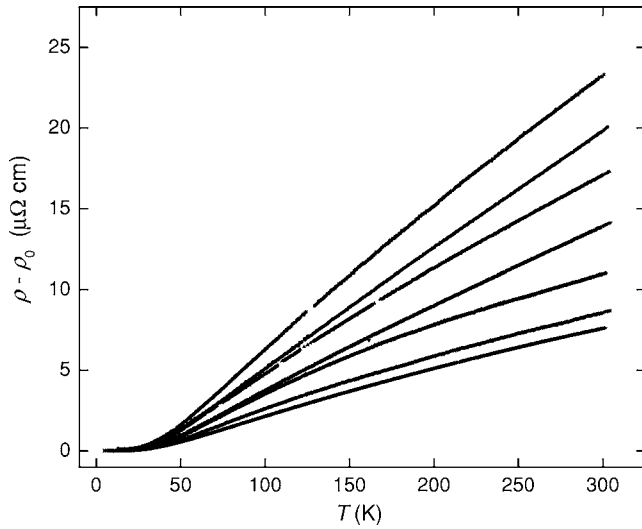


FIG. 1. Variation of  $\Delta\rho = \rho - \rho_0$  with temperature for a series of  $\text{Au}_{56}\text{Pd}_{44}$  thick films. The values of the residual resistivity for the films are (from bottom to top): 39.3, 66.4, 125, 171, 202, 264, and 275  $\mu\Omega$  cm. Notice that  $\Delta\rho(T)$  increases with increasing disorder.

$\sim 3$  (for our cleanest sample) to  $\sim 5$  (for our dirtiest sample) larger than the Bloch-Grüneisen contribution, indicating the dominant role of the electron-phonon-impurity interference mechanism. Thus, the prediction of Eq. (2) is realized in the present material system. As the temperature increases, the

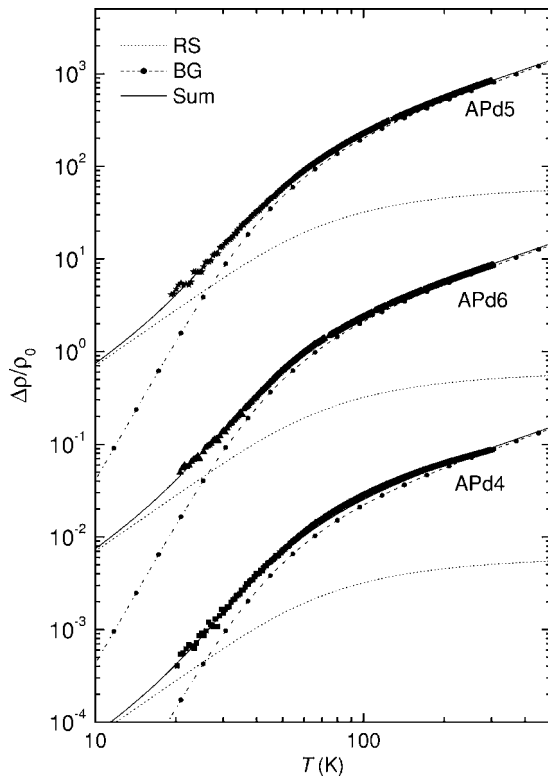


FIG. 2. Variation of  $\Delta\rho/\rho_0 = (\rho - \rho_0)/\rho_0$  with temperature for three representative  $\text{Au}_{56}\text{Pd}_{44}$  thick films as indicated. The theoretical predictions (solid curves) can well describe the experimental data. For clarity, the data for APd6 and APd5 have been multiplied by 100 and 10 000, respectively.

TABLE I. Values of relevant parameters for seven  $\text{Au}_{56}\text{Pd}_{44}$  and two  $\text{IrO}_2$  thick films. The  $\text{Au}_{56}\text{Pd}_{44}$  films are  $\geq 0.50$   $\mu\text{m}$  thick, and the  $\text{IrO}_2$  films are 0.30  $\mu\text{m}$  thick.

Films	$\rho_0$ ( $\mu\Omega$ cm)	$\rho(300 \text{ K})/\rho_0$	$\beta_{\text{BG}}$ ( $\mu\Omega$ cm/K)
APd5	276	1.084	0.30
APd3	265	1.075	0.25
APd6	202	1.086	0.23
APd1	171	1.082	0.18
APd4	125	1.091	0.15
APd2	66.4	1.130	0.12
AP052	39.3	1.194	0.10
Ir300	286	1.208	0.38
Ir100	432	1.085	0.18

Bloch-Grüneisen contribution becomes progressively important. Between 20 and 28 K (depending on disorder), a cross-over happens and the Bloch-Grüneisen term eventually determines the measured resistivity, as expected from the standard electrical-transport theory.

The measured and fitted values for the relevant parameters for our samples are listed in Table I. In performing least-squares fits to Eqs. (1), (2), and (5), we found that a *single* value of  $B = 7.0 \times 10^{-7} \text{ K}^{-2}$  could be used for *all* the  $\text{Au}_{56}\text{Pd}_{44}$  thick films listed in Table I. Since the residual resistivity of our films varies by a factor as large as 7, this result of a *constant*  $B$  provides a strong justification for Eq. (2). This value of  $B$  is a factor of  $\sim 4$  and  $\sim 1.5$  smaller than that found in Au and Al thin films, respectively.<sup>6</sup>

For  $\text{Au}_{56}\text{Pd}_{44}$ , using the free-electron theory, we estimate  $E_F \approx 4.1 \text{ eV}$ ,  $N(0) \approx 1.0 \times 10^{47} \text{ states/J m}^3$ ,  $\rho_m \approx 1.65 \times 10^4 \text{ kg/m}^3$ ,  $u_l \approx 3.5 \times 10^3 \text{ m/s}$ ,  $u_t \approx 1.3 \times 10^3 \text{ m/s}$ , and  $p_F \approx 1.3 \times 10^{-24} \text{ kg m/s}$ . Then, we obtain the theoretical values of  $\beta_l \approx 0.34$  and, from Eq. (3),  $B \approx 7.7 \times 10^{-7} \text{ K}^{-2}$ . This theoretical values of  $B$  is in excellent agreement with the experimental data. For a typical  $\text{Au}_{56}\text{Pd}_{44}$  film with  $\rho_0$  of order 100  $\mu\Omega$  cm, the characteristic temperature  $T_l^*$  above which  $q_l l > 1$  is 5 K. This criterion is satisfied in our measurements.

Figure 3 shows the variation of the fitted Bloch-Grüneisen prefactor  $\beta_{\text{BG}}$ , Eq. (6), with residual resistivity for our  $\text{Au}_{56}\text{Pd}_{44}$  thick films. This figure demonstrates that  $\beta_{\text{BG}}$  increases with increasing  $\rho_0$ . As  $\rho_0$  varies from 40 to 280  $\mu\Omega$  cm,  $\beta_{\text{BG}}$  increases roughly linearly from 0.10 to 0.30  $\mu\Omega$  cm/K. (However, there is no *prior* reason why  $\beta_{\text{BG}}$  should vary linearly with disorder.) This result is in sharp contrast to the behavior of the interference mechanism prefactor  $B$ . As discussed, the prefactor  $B$  for all of our films can be fixed at a single value, being independent of disorder. On the other hand, we stress that, when a wide range of  $\rho_0$  is concerned, Fig. 3 indicates that one can by no means fix  $\beta_{\text{BG}}$  at a single value for all samples. The issue of applying the Reizer-Sergeev theory to a *wide* range of disorder for a given material has not been addressed in previous experiments.<sup>4-9</sup>

We notice that our experimental data of  $\Delta\rho(T)/\rho_0$  *cannot* be described by Eq. (1) even by fixing  $\beta_{\text{BG}}$  to a constant value while allowing  $\theta_D$  to vary from sample to sample, as

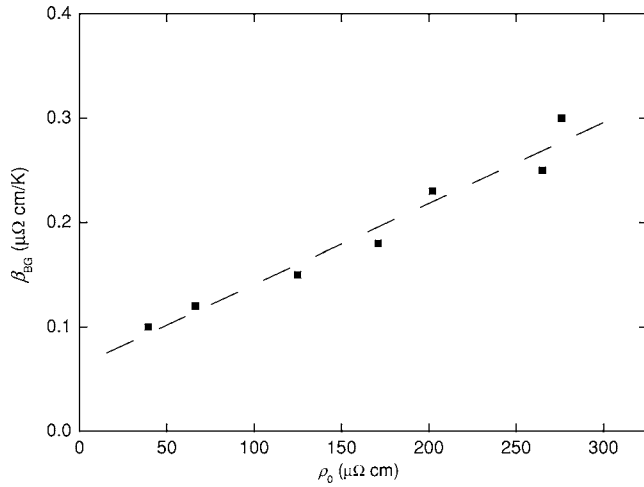


FIG. 3. Variation of the fitted Bloch-Grüneisen prefactor  $\beta_{BG}$  with residual resistivity  $\rho_0$  for  $\text{Au}_{56}\text{Pd}_{44}$  thick films. The dashed line is a guide to the eye.

one might conjecture. The functional form of Eq. (5) indicates that the effect of changing the value of  $\theta_D$  is to significantly alter the temperature behavior of  $\Delta\rho/\rho_0$  (especially at several tens K and higher, or  $T \gtrsim 0.3\theta_D$ ), and thus no longer reproduce our experimental results.

At first glance, a prefactor  $\beta_{BG}$  dependent on disorder is unexpected according to the Bloch-Grüneisen law. However, comparison of Eqs. (6) and (3) suggests that  $\beta_{BG}$  possess a much stronger dependence on the electronic properties of the material than  $B$  does. Therefore, it may not be unplausible to find, in our system, a prefactor  $\beta_{BG}$  revealing a variation with disorder while the prefactor  $B$  remaining essentially constant. Physically, the noticeable dependence of  $\beta_{BG}$  on  $\rho_0$  found in Fig. 3 is likely to result from significant modifications of the electron (and probably phonon) properties of  $\text{Au}_{56}\text{Pd}_{44}$  with increasing disorder. [We should point out that our  $\text{Au}_{56}\text{Pd}_{44}$  thick films are so disordered that  $\rho(300\text{ K})/\rho_0 \approx 1.1-1.2$ .] Indeed, systematic measurements of the electronic-transport properties as a whole, including resistivities, thermoelectric powers, and Hall coefficients, have been undertaken on our films. Our results indicate that lattice disorder renormalize the material parameters, leading to an enhancement in the electron-phonon coupling and a reduction in the Fermi velocity of this material.<sup>22</sup> Further theoretical and experimental investigation is definitely necessary to clarify this issue of disorder variation of  $\beta_{BG}$  in  $\text{Au}_{56}\text{Pd}_{44}$ .

### B. $\text{IrO}_2$ thick films

In order to test the validity of Eq. (2) for a wide range of materials, we have also studied the resistivities of two polycrystalline  $\text{IrO}_2$  thick films between 15 and 300 K. The films were prepared by rf sputtering on glass substrates as described previously.<sup>23</sup> The electronic band structure and electrical-transport properties of  $\text{IrO}_2$  have been established, both theoretically<sup>24,25</sup> and experimentally.<sup>26</sup> It is understood that in this *metallic* material, apart from the usual Bloch-Grüneisen contribution to the resistivity, there is an additional contribution due to the coupling of electrons with

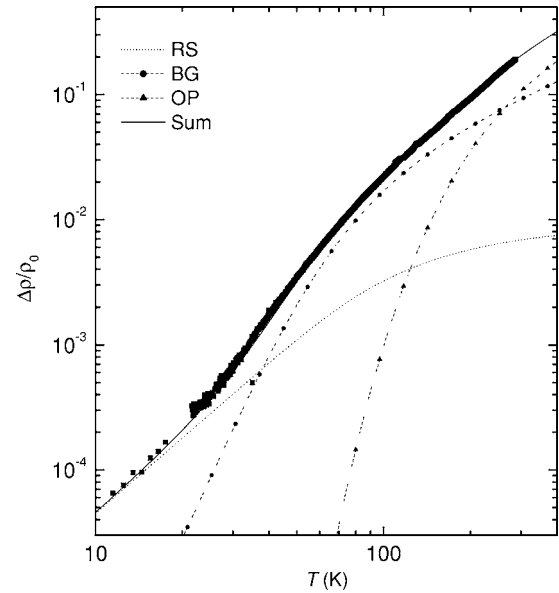


FIG. 4. Variation of  $\Delta\rho/\rho_0 = (\rho - \rho_0)/\rho_0$  with temperature for  $\text{Ir}_{300}$  thick film. The theoretical prediction (the solid curve), Eq. (7), can well describe the experimental data.

optical-mode phonons (this term is important for transition-metal oxides which contain multiatom bases). Therefore, this material can provide a new horizon to test whether the Reizer-Sergeev theory may be applied to *conducting* transition-metal oxides. In the presence of the electron-optical-mode phonon coupling, the total resistivity can be written as:

$$\rho(T) = \rho_0 + \rho_{\text{int}}(T) + \rho_{BG}(T) + \rho_E(T). \quad (7)$$

Here the last term is treated using the Einstein approximation with a single phonon frequency corresponding to the energy  $k_B\theta_E$ , and can be written as<sup>25,26</sup>

$$\rho_E(T) = \beta_E T \left[ \frac{\theta_E/2T}{\sinh(\theta_E/2T)} \right]^2, \quad (8)$$

where  $\beta_E$  is a prefactor whose value depends on the material properties.

Figure 4 shows the variation of  $\Delta\rho/\rho_0$  with temperature in double logarithmic scales for one of our  $\text{IrO}_2$  thick films. (For clarity, we show only one film with various contributions in this plot.) The measured data were least-squares-fitted to Eq. (7) and the relevant parameters were determined and listed in Table I.<sup>27</sup> It is clearly seen that the theoretical prediction (the solid curve) can well describe the experimental result. In particular, at low temperatures of  $T \approx 0.1\theta_D$ , the contribution from the interference mechanism is a factor of  $\sim 10$  larger than that from the Bloch-Grüneisen term. Thus, the prediction of Eq. (2) is established, for the first time, in conducting transition-metal oxides. At higher temperatures, the usual Bloch-Grüneisen and the optical-mode contributions become progressively dominant. Eventually, at sufficiently high temperatures (well above  $\theta_D$ ), the



electron-optical-mode phonon coupling term would surpass the Bloch-Grüneisen term which is due to electron-acoustic-mode phonon coupling.

Quantitatively, we obtained the electron-phonon-impurity interference prefactor  $B \approx 4.5 \times 10^{-7} \text{ K}^{-2}$  in these two films. This value is close to that ( $\approx 7.0 \times 10^{-7} \text{ K}^{-2}$ ) found in our  $\text{Au}_{56}\text{Pd}_{44}$  thick films. This agreement, on one hand, supports the idea that  $\text{IrO}_2$  possess usual metallic behavior and, on the other hand, establishes the wide validity of the Reizer-Sergeev theory.

## V. CONCLUSION

Using a series of  $\text{Au}_{56}\text{Pd}_{44}$  and two  $\text{IrO}_2$  thick films with different amounts of disorder for resistivity measurements, we have found that the contribution from the electron-

phonon-impurity interference effect to resistivity  $\rho_{\text{int}}$  is significant. This interference mechanism dominates over the usual Bloch-Grüneisen term at temperatures below  $0.1\theta_D$ . Our observation confirms the predicted temperature and disorder behavior of  $\rho_{\text{int}} \propto \rho_0 T^2$ . Moreover, our results imply that the electron and phonon properties (e.g., the electron-phonon coupling and Fermi velocity) of  $\text{Au}_{56}\text{Pd}_{44}$  may be significantly modified in the presence of strong disorder. This last observation deserves further investigation.

## ACKNOWLEDGMENTS

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- <sup>21</sup>In generating the least-squares-fits of our measured normalized resistivities  $\Delta\rho/\rho_0$  to Eqs. (1), (2), and (5), we had performed numerical integrations of the integrals in Eqs. (2) and (5). In addition, we had adjusted the prefactor  $\beta_{\text{BG}}$  in Eq. (5) such that the Bloch-Grüneisen term exactly reproduced the measured  $\Delta\rho/\rho_0$  at room temperature.
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- <sup>27</sup>For these two films, we determined  $\theta_D \approx 370 \text{ K}$  and  $\theta_E \approx 850 \text{ K}$ . This value of  $\theta_E$  was adopted from Ref. 26. The functional form of Eq. (8) indicates that the magnitude of  $\rho_E$  is not very sensitive to some variation in  $\theta_E$ . Also, from Refs. 25 and 26 it is known that  $\beta_E = 0.5\beta_{\text{BG}}$ . Therefore, only  $\beta_{\text{BG}}$  needs to be treated as a free parameter in the comparison of the theory with experiment.