Electron-electron scattering times in low-diffusivity thick RuO_2 and IrO_2 films

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We have systematically measured the electron dephasing scattering times from a three-dimensional weak-localization study of low-diffusivity thick RuO_2 and IrO_2 films. We find that the inelastic electron scattering rate $1/\tau_{in}$ varies essentially linearly with the temperature and there is essentially no dependence of $1/\tau_{in}$ on the electron elastic mean free path. This observation is understood in terms of the current theoretical concept for electron-electron scattering in strongly disordered bulk conductors. [S0163-1829(99)02601-6]

I. INTRODUCTION

Over the last 15 years, it has been realized that weak-localization effects result in noticeable magnetoresistivities in *weakly disordered* conductors at low temperatures and in low magnetic fields. Analysis of those ''anomalous'' magnetoresistivities has proved to provide fairly quantitative information of the various dephasing scattering mechanisms of the electron wave functions, including inelastic, spin-orbit, and magnetic spin-spin scattering processes.[1] In particular, the inelastic electron dephasing scattering times have been intensively investigated and become very well understood in one and two dimensions (i.e., small-diameter wires and thin films, respectively). It is now widely accepted that the dominating inelastic dephasing process in reduced dimensions is the (''quasielastic'') electron-electron scattering at small energy transfers.[2] In three-dimensional weakly disordered conductors, on the other hand, both theoretical and experimental studies have found that electron-phonon scattering is the sole, important inelastic mechanism while the small energy-transfer electron-electron scattering is usually negligibly weak.[3] Thus, it can be considered that the inelastic dephasing scattering process in every dimension in the weakly disordered regime is now well identified.[4] In this work, we shall concentrate on the three-dimensional case. The usual materials used for studies in this case are thick granular films,[5] thick quench-condensed metal films,[6] doped semiconductors,[7] and metallic glasses,[8] but not bulk metallic oxides.[9] Obviously, it would be of great interest to obtain information on the inelastic electron dephasing scattering times in metallic oxides and compare them with those in the just mentioned more conventional conductors. To our knowledge, there has been no report on the weak-localization effects and the inelastic electron dephasing scattering times in the RuO_2 and IrO_2 dioxide compounds which crystallize in the rutile structure. As a matter of fact, the electronic structure and the Boltzmann transport properties of these two rutiles have been extensively studied in the literature,[10,11] partly because of their exhibiting metallic conductivity resulting from the partially filled Ru $4d$ and Ir $5d$ states.

The electron dephasing scattering time, τ_ϕ, in disordered conductors can be reliably determined from weak-localization studies, and it reads[1,2]

$$\frac{1}{\tau_\phi(T)} = \frac{2}{\tau_s} + \frac{1}{\tau_{in}(T)}. \tag{1}$$

In Eq. (1) τ_s is a temperature-independent residual scattering time (caused by magnetic spin-spin scattering or quantum zero-point fluctuations[12]), and $1/\tau_{in} = AT^p$ is due to inelastic electron scattering. For the case of diffusive electrons in the weakly disordered regime, it is established that the temperature exponent $p = 2$, 3, or 4 for electron-phonon scattering,[3] and $p = d/2$ for small energy-transfer electron-electron scattering (where $d = 2$ or 3 is the sample dimensionality).[2] In addition to the knowledge of the temperature dependence of τ_{in}, information about the disorder (or, the electron elastic mean free path l) dependence of the inelastic scattering strength A is of prime importance for a critical discrimination among the various theoretical models for the inelastic electron processes in impure conductors. In this work, we will show that the temperature and disorder behavior of τ_{in} in low-diffusivity thick RuO_2 and IrO_2 films is distinctly different from that expected in the weakly disordered bulk conductors.

II. EXPERIMENTAL METHOD

Seven RuO_2 and IrO_2 films with thickness of about 3000 Å were deposited by reactive RF magnetron sputtering. The target was a 1 inch diameter Ru or Ir metal with a purity of 99.99%. The films were deposited onto fused silica substrates. The substrate temperature during deposition, which was monitored using a thermocouple attached to the substrate holder, was kept in the range of 100–400 °C. Before the deposition, the sputtering chamber was evacuated to a pressure of $\sim 1\times 10^{-6}$ torr. The O_2 and Ar gases were introduced into the sputtering chamber using independently

TABLE I. Values of relevant parameters for disordered thick RuO_2 and IrO_2 films. t designates film thickness. A is the inelastic electron scattering strength and p is the temperature exponent of the inelastic electron scattering time, i.e., $1/\tau_{in}=AT^p$. Owing to uncertainties in the geometrical dimensions, the absolute values of ρ are accurate to about 10% or higher. The values of D are estimated to be accurate to about 30%. ρ is in $\mu\Omega$ cm, and A in $1/K^p$ s.

Film	t (Å)	ρ (300 K)	ρ (10 K)	D (cm^2/s)	A	p	$2/\tau_s$ (s^{-1})
Ru200[a]	3000	482	413	0.29	0.30	1.37 ± 0.13	5.5×10^{10}
Ru300	3000	389	311	0.39	0.35	1.22 ± 0.22	7.1
Ru350	3300	397	301	0.40	0.51	1.11 ± 0.07	3.9
Ru400	3000	268	161	0.75	0.71	0.91 ± 0.10	2.4
Ir100	3000	407	387	0.33	0.37	1.35 ± 0.06	2.8×10^{10}
Ir300	3000	286	247	0.52	0.75	1.00 ± 0.07	6.8
Ir350	3300	333	269	0.48	0.61	0.99 ± 0.14	8.0

[a]The number following the symbol (Ru or Ir) indicates the substrate temperature used for sputtering deposition of the particular film.

controlled fine needle valves. The O_2 was maintained at a pressure of 3×10^{-3} torr, and the total working gas pressure ($Ar+O_2$) was kept at a constant level of about 7.5×10^{-3} torr. The distance between substrate and target was fixed at 3 cm. The sputtering rate was about 200 Å/min for a sputtering power of 100 W. Characterization of the crystallinity of the films was performed using x-ray diffraction. The basic rutile structure of RuO_2 and IrO_2 was retained.[13] In general, the diffraction peaks shifted to smaller angles and the linewidths of the peaks increasingly broadened as the substrate temperature decreased. The shift to lower angles and the increase of the broadening of the diffraction peaks were attributed to microcrystallinity and local disorder. The relevant parameters for our films are listed in Table I. Notice that our films were fairly thick, with thicknesses of about 3000 Å. These films should therefore possess three- rather than two-dimensional physical behavior (see below and Ref. 14). Also, the residual resistivities ρ(10 K) ($\gtrsim160\ \mu\Omega$ cm or much higher) of these films were much higher than those (of order 1 $\mu\Omega$ cm or smaller[15]) of single-crystal RuO_2 and IrO_2, suggesting that our films were very disordered.

The magnetoresistivities of our films were measured by a standard four-probe technique between 2 and 20 K and in magnetic fields below 1 T. The magnetoresistivities at various measuring temperatures for each film were compared with three-dimensional weak-localization theory[1] to extract the electron dephasing time τ_ϕ. The details of the least-squares fitting procedure had been discussed previously.[16] Here we merely stress that, for every film studied in this work, the three-dimensional weak-localization predictions in the limit of *strong* spin-orbit scattering, as appropriate for RuO_2 and IrO_2, can well describe our experimental results. Thus, τ_ϕ could be reliably extracted. Since τ_ϕ is given in the weak-localization theory through the ''characteristic'' field defined by $B_\phi=\hbar/4eD\tau_\phi$ (where $\hbar$ and e have the usual meaning, and D is a diffusion constant), one needs the value of D to compute $\tau_\phi(T)$ from the fitted $B_\phi(T)$. To do this, we use the experimentally measured electronic specific heat $\gamma T=305T$ and $287T$ J/m^{3}K for RuO_2 and IrO_2 (Ref. 17), respectively, to calculate the electronic density of states at the Fermi level $N(0)$, using the free-electron model $\gamma=\pi^2k_B^2(1+\lambda)N(0)/3$, where λ (≈0.5, Ref. 11) is the electron-phonon enhancement factor. The electron diffusion constant D is then obtained through the Einstein relation $D=1/\rho e^2N(0)$, where ρ is the measured impurity resistivity ρ(10 K). Our best values of D thus obtained are listed in Table I. We note that these values of D are *relatively low*, being only on the order of $\sim10^{-5}$ m^2/s. With the values of D and ρ(10 K) being available, there then remains only one adjustable parameter involved in the comparison (i.e., least-squares fitting) of experiment with theory: the temperature-dependent B_ϕ. (As mentioned, the spin-orbit scattering field B_{so} can essentially be set to infinity in the least-squares fits for both RuO_2 and IrO_2 films.)

III. RESULTS AND DISCUSSION

Figure 1 shows the measured, normalized magnetoresistivities $\Delta\rho(B)/\rho^2(0)=[\rho(B)-\rho(0)]/\rho^2(0)$ and the three-dimensional weak-localization predictions for a representative film, Ru350, at three measuring temperatures of (from top to bottom) 3.0, 10.0, and 17.0 K. The symbols are the experimental data and the solid curves are the theoretical results. It is clearly seen that the theoretical predictions agree

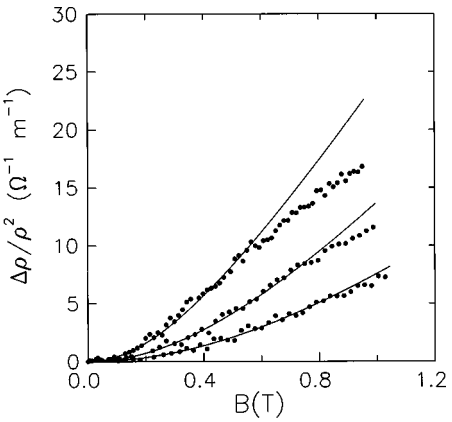

FIG. 1. Normalized magnetoresistivity $\Delta\rho(B)/\rho^2(0)$ as a function of the magnetic field for the Ru350 film at (from top to bottom) 3.00, 10.0, and 17.0 K. The symbols are the experimental results, and the solid curves are the three-dimensional weak-localization predictions.

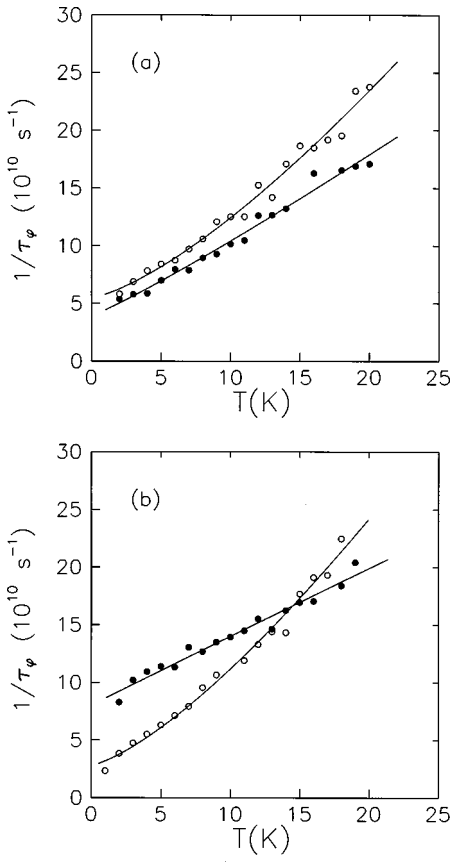

FIG. 2. Electron dephasing scattering rate $1/\tau_\phi$ as a function of temperature for thick (a) Ru200 (open circles) and Ru350 (closed circles) films, and (b) Ir100 (open circles) and Ir350 (closed circles) films. The solid lines are the least-squares fits to Eq. (1).

very well with the experimental data in low magnetic fields. The discrepancies in higher magnetic fields especially at low measuring temperatures are expected and are only drawn for reference. (It is known that, in higher magnetic fields, electron-electron interaction effects would also contribute to the magnetoresistivities which need be included in order to fully account for the experimental data.)

Our main results of this work, i.e., the extracted values of the electron dephasing scattering time $\tau_\phi(T)$, are summarized in Figs. 2 and Table I. Figure 2(a) shows the variations of $1/\tau_\phi$ with temperature for two representative RuO_2 films: Ru200 (open circles) and Ru350 (closed circles); while Fig. 2(b) shows the variations of $1/\tau_\phi$ with temperature for two representative IrO_2 films: Ir100 (open circles) and Ir350 (closed circles). The solid lines drawn through the data points in Figs. 2(a) and 2(b) are least-squares fits to Eq. (1) with the inelastic electron scattering strength A, the temperature exponent p, and the residual scattering rate $2/\tau_s$ as adjusting parameters. Inspection of Figs. 2(a) and 2(b) clearly indicates that Eq. (1) can well describe our experimental data of $1/\tau_\phi$ over the wide temperature range of 2–20 K. Experimentally, as many as 16–18 magnetoresistivity curves, corresponding to 16–18 experimental data points for $1/\tau_\phi$, have been measured for every film. Therefore, any appreciable experimental uncertainties in the extraction of the values of the adjusting parameters from comparison of the experimental data with Eq. (1) can be largely minimized. Notice that only *one* inelastic process is included in our analysis and is

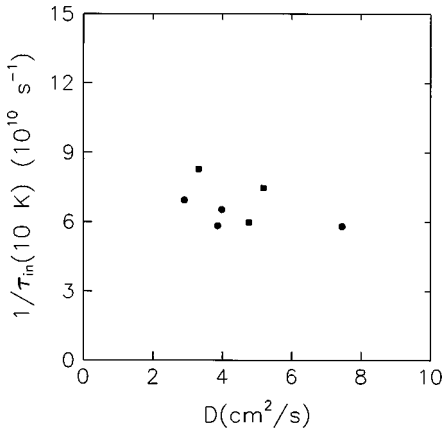

FIG. 3. Fitted inelastic electron scattering rate at 10 K, $1/\tau_{in}$ (10 K), as a function of diffusion constant D for thick RuO_2 (circles) and IrO_2 films (squares). Notice that $1/\tau_{in}$ (10 K) depends very weakly on D.

already sufficient to fully account for the experimental results. That is, there is no need to invoke two or more inelastic electron processes (and thus more adjusting parameters) to fully describe the experimental $1/\tau_\phi$. Our best fitted values of the inelastic scattering strength A, the temperature exponent p, and the residual scattering rate $2/\tau_s$ for all films are listed in Table I. Inspection of Table I reveals an average experimental value of $p \approx 1.13$ for the seven films studied, strongly suggesting that the inelastic electron scattering rate $1/\tau_{in}$ varies *essentially linearly* with the temperature T in these two dioxides. This value of p is *significantly low* compared with that ($p=2-4$) established for the electron-phonon scattering in three-dimensional weakly disordered conductors. Therefore, electron-phonon scattering can be safely ruled out as the responsible inelastic dephasing process in these two dioxides.

Figure 3 shows the variation of the ''fitted'' inelastic electron scattering rate $1/\tau_{in}$ at (a representative temperature) 10 K with diffusion constant D for four RuO_2 (circles) and three IrO_2 films (squares) studied in this work. This figure indicates that the values of $1/\tau_{in}$(10 K) for these two dioxides are fairly similar. More importantly, it is apparent that the values of $1/\tau_{in}$(10 K) are very similar for all samples, regardless of the difference (by a factor ~ 2.6) in the values of the diffusion constant D for the various films. This result is strongly suggestive of a *disorder insensitive* inelastic electron dephasing process operating in impure thick RuO_2 and IrO_2 films. This very weak dependence on D (or, mean free path l) again strongly suggests that the responsible inelastic dephasing process in our thick RuO_2 and IrO_2 films is *not* caused by electron-phonon scattering. If electron-phonon scattering were the dominating inelastic process, a noticeable dependence of $1/\tau_{in}$ on l would have most likely been observed.[18]

In the case of three dimensions, observations of a linear dependence of $1/\tau_{in}$ on the temperature have been reported in thick granular aluminum films,[19] thick scandium films,[20] doped semiconductors,[21] and heavily doped conjugated polymers[22] on the metallic side of the metal-insulation transition. Recently, these observations have been attributed to the electron-electron scattering in impure conductors near a mobility edge. Belitz and Wysokinski[23] have calculated the

inelastic quasiparticle lifetime due to a Coulomb interaction in disordered bulk metals, their calculation being perturbative with respect to the screened Coulomb interaction, but for an arbitrary disorder. They found that the inelastic electron-electron scattering is very sensitive to the critical, as opposed to diffusive, current dynamics in systems near the Anderson transition. In particular, they observed a linear temperature dependence of the inelastic scattering rate. They also predicted that the inelastic scattering rate should be disorder independent. Their predictions are in line with our experimental results for thick RuO_2 and IrO_2 films which have very low values of diffusion constant D. Quantitatively, our experimental value of $1/\tau_{in}$ is reasonably close to that found in, e.g., thick scandium films just mentioned, and is comparable with the theoretical value of Belitz and Wysokinski. It is worth pointing out that, although a linear dependence of $1/\tau_{in}$ on temperature for the predicted electron-electron scattering time has previously been reported, the independence of $1/\tau_{in}$ on disorder has never been tested in almost all of the above-mentioned material systems (except in the case of thick scandium films[20]). The present experimental results thus provide valuable systematic evidence (the temperature as well as the disorder dependences) in supporting the basic predictions of the Belitz-Wysokinski theory.

Strictly speaking, in applying the Belitz-Wysokinski theory to the case of our RuO_2 and IrO_2 films, one should notice that it is not totally indisputable. According to the theory, a linear T dependence of the inelastic electron-electron scattering rate should be realized only in strongly disordered metals in the vicinity of a metal-insulator transition. In particular the prediction of a disorder-independent inelastic scattering time holds only in the extremely disordered regime. As for our RuO_2 and IrO_2 films, although they are rather disordered as mentioned, they are probably not close enough to the mobility edge. Typically, a resistivity of a few thousand $\mu\Omega$ cm is what one would assume to be needed in order to be in the quantum-critical state for which this theory is applicable. For example, $1/\tau_{in} \sim T$ has been observed in thick granular aluminum films for which $\rho(300\ \mathrm{K}) \approx 2000-6000\ \mu\Omega$ cm.[19] On the other hand, however, as long as the microscopic disorder is concerned, one can think that our RuO_2 and IrO_2 films are far more homogeneous than granular aluminum films. In high-resistivity granular films where a linear T dependence of $1/\tau_{in}$ is observed, most of the resistivities are actually contributed from the grain boundaries while the metal grains might only be weakly disordered. Then, it is conjectured that the criterion for the applicability of the Belitz-Wysokinski theory might be less stringent than originally evaluated. Another point which should be briefly addressed is that an inspection of the values of the temperature exponent p for $1/\tau_{in}$ in Table I reveals that p moves closer to 1 as the disorder is reduced (rather than as it increases). At first glance, this observation seems to argue against the Belitz-Wysokinski picture. Again, however, it is conceived that this discrepancy might be removed and a more quantitative agreement between the theory and measurement would be achieved if our films could be made more resistive. In short, we think it is suitable to say that the Belitz-Wysokinski theory is the *most* (and only) plausible *existing* theory that contains the most fundamental attributes we see in our data for $1/\tau_{in}$ (independence from disorder and a linear T dependence). Alternatively, to be alert, one should not completely ignore the possibility that there might be as yet unidentified dephasing mechanisms that are also important in RuO_2 and IrO_2 dioxides (and in other low-diffusivity materials).

Using the values of A and p and the values of D given in Table I, we estimate the inelastic electron dephasing scattering length, $L_{in}=(D\tau_{in})^{1/2}$ [$>L_\phi=(D\tau_\phi)^{1/2}$], for our films to range from about 350 to about 1300 Å as the temperature decreases from 20 down to 2 K. That is, every film studied lies well in the three-dimensional regime, justifying our use of the three-dimensional weak-localization predictions to describe the experimental magnetoresistivities. Again, we point out that our films are not two dimensional, and thus the observed linear T behavior of $1/\tau_{in}$ is not due to the two-dimensional small energy-transfer electron-electron processes usually operating in weakly disordered thin metal films in which an exponent $p=1$ has been established theoretically[2] and experimentally.[24]

IV. CONCLUSION

We have observed an essentially linear temperature dependence of the inelastic electron scattering rate $1/\tau_{in}$ in a series of low-diffusivity thick RuO_2 and IrO_2 films. We have also observed that $1/\tau_{in}$ depends very weakly on the electron elastic mean free path. Our results are qualitatively understood in terms of the current theoretical concept that considers the electron-electron scattering in strongly disordered bulk conductors.

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