Effect of microstructures on the electron-phonon interaction in the disordered metals 
\textbf{Pd}_{60}\textit{Ag}_{40}

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(Dated: November 16, 2018)

PACS numbers: 73.61.At, 72.10.Di, 72.15.Rn

I. INTRODUCTION

The electron-phonon (\textit{e-\textit{ph}}) scattering time, \(\tau_{e\textit{-ph}}\), is one of the most important physical quantities in metals and superconductors. For instance, it determines the dephasing (also called the phase-breaking or decoherence) time for the electron wave function, the cooling time for an electron gas, and the relaxation time for the order parameter in a superconductor. The \textit{e-\textit{ph}} scattering time also plays a crucial role in the development of novel mesoscopic devices such as sensitive low-temperature bolometers \cite{1}. The \textit{e-\textit{ph}} scattering time in the presence of multiple (elastic) impurity scattering has been intensively calculated by several authors \cite{2, 3, 4}, but the current understanding of the temperature and electron elastic mean free path, \(\ell\), dependences of \(\tau_{e\textit{-ph}}\) is still incomplete. In particular, different temperature and disorder dependences of \(\tau_{e\textit{-ph}}\) have been reported, both theoretically and experimentally \cite{5, 6}. Recently, it was proposed that, in addition to the dependence on the total level of disorder, the \(T\) and \(\ell\) dependence of \(\tau_{e\textit{-ph}}\) might be fairly sensitive to the microscopic quality of the disorder \cite{7, 8, 9}. It has also been conjectured that the contribution due to the Umklapp process of impurity scattering may be important \cite{10}. In this work, we have fabricated two series of \textit{Pd}_{60}\textit{Ag}_{40} thick films by DC-sputtering and RF-sputtering deposition techniques. The palladium-silver alloys are chosen because Pd and Ag form perfect fcc solid solution through the alloy series \cite{11}. Also, since the masses of the Pd and Ag atoms are quite similar, the vibrational spectrum of the lattice does not change significantly through the alloy series \cite{12}. The low-field magnetoresistances of our films are measured at liquid-helium temperatures, and are compared with the weak-localization theoretical predictions to extract the values of the \textit{e-\textit{ph}} scattering time. Our results for the temperature and electron mean free path dependence of \(\tau_{e\textit{-ph}}\) and their implications are described below.

II. EXPERIMENTAL METHOD

Our films were prepared from a 99.995\% pure \textit{Pd}_{60}\textit{Ag}_{40} (hereafter refereed to as \textit{PdAg}) target. Two series of thick films were fabricated, one by DC-sputtering and the other by RF-sputtering deposition technique. The films were deposited onto glass substrates held at room temperature. In both cases, a background pressure of \(3 \times 10^{-6}\) torr was reached before an argon atmosphere of \(3.8 \times 10^{-3}\) torr was introduced to initiate the deposition process. A same sputtering gun was used for these two deposition methods, but with the gun being connected to either a DC or a RF power supply. The distance between the sputtering target and the glass substrates was the same for both methods. The sputtering power was progressively adjusted to “tune” the deposition rate, which resulted in different amounts of disorder, i.e., the residual resistivities \(\rho_0 [= \rho(10\, K)]\), in the films. For the DC-sputtering (RF-sputtering) case, the deposition rate was varied from 30 to 230 (19 to 333) \(\AA/\text{min}\), and values of \(\rho_0\) ranging from 281 to 183 (74 to 178) \(\mu\Omega\, \text{cm}\) were obtained.

The sample structures of our films were carefully studied by performing the powder diffraction on an MAC MXP18 x-ray diffractometer. The x-ray power was 10 kW and the scanning speed was 6 degrees per minute. In all cases, we found our samples to reveal very similar diffraction patterns, which clearly suggested that both the DC and RF sputtered films possessed the same fcc lattice structure characteristic to that of the \textit{PdAg} alloys. Representative x-ray diffraction patterns for two DC and

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two RF sputtered films are shown in Fig. 1. Most of our films had a thickness \( t \gtrsim 4000 \) Å. This thickness ensured that the weak-localization effects were three-dimensional in our samples. It also ensured that the thermal phonons were unambiguously three-dimensional, i.e., the wavelength of the most probable thermal phonons was always smaller than the film thickness at our measurement temperatures. This latter condition greatly eliminated any complications that might result from phonon confinement effects. (In reduced-dimension systems, modifications to the phonon spectrum and the speed of sound might be significant, which could lead to non-straightforward temperature and disorder behavior of \( \tau_{\text{ep}} \).

Our values of the diffusion constant, \( D \), were evaluated through the Einstein relation \( \rho_0^{-1} = D e^2 N(0)/(1 + \lambda) \), where \( N(0) \) is the electronic density of states at the Fermi level, and \( \lambda \) is the \( e\)-\( ph \) mass enhancement factor. The values of \( N(0) \) were calculated from the independently determined electronic specific heat: \( \gamma T = \frac{1}{2} \pi^2 k_B^2 N(0) T \). For PdAg \( 40 \), \( \lambda \approx 0.43 \), and \( \gamma \approx 3.3 \) mJ/mol K\(^2 \). Then, we obtained \( D \approx (100/\rho_0) \) cm\(^2\)/s, where \( \rho_0 \) is in \( \mu\Omega \) cm. Table 1 lists the values of the relevant parameters for our films studied in this work.

### III. RESULTS AND DISCUSSION

The normalized magnetoresistivities, \( \Delta \rho(B)/\rho^2(0) = (\rho(B) - \rho(0))/\rho^2(0) \), for the PdAg17 thick film at several temperatures are plotted in Fig. 2. The symbols are the experimental data and the solid curves are the three-dimensional weak-localization theoretical predictions [13]. It is clearly seen that the weak-localization predictions can describe well our experimental data. Therefore, the electron dephasing time \( \tau_{\phi} \), which is the key parameter in the weak-localization theory, can be reliably extracted. Since PdAg has a very strong spin-orbit scattering, \( \tau_{\phi} \) is the only adjusting parameter in the comparison of the theory with experiment. (That the spin-orbit scattering is strong in PdAg is evident in the shape of the positive magnetoresistivity curves shown in Fig. 2.) The details of our data analysis procedure was discussed previously [13].

In three dimensions, the total electron dephasing rate that governs the weak-localization effects is given by [13]

\[
\frac{1}{\tau_{\phi}(T, \ell)} = \frac{1}{\tau_{\phi}^0(\ell)} + \frac{1}{\tau_{\text{ep}}(T, \ell)},
\]

where \( \tau_{\phi}^0 = \tau_{\phi}(T \rightarrow 0) \) depends very weakly on temperature, if at all, and is called the saturated dephasing time. Whether \( \tau_{\phi}^0 \) should reach a finite or an infinite value as \( T \rightarrow 0 \) is currently under vigorous experimental and theoretical investigations [8]. At finite temperatures, the dominating inelastic electron process in three dimensions is solely due to the \( e\)-\( ph \) scattering, while the Nyquist electron-electron scattering is negligibly small [1, 2, 3, 4, 9, 6]. Usually, one writes \( 1/\tau_{\text{ep}} = A_{\text{ep}} T^p \) over the limited temperature range accessible in a typical experiment, where \( A_{\text{ep}} \) characterizes the strength of the \( e\)-\( ph \) coupling, and \( p \) is an effective exponent of temperature. According to current understanding, \( p \) lies between 2 and 4 [2, 3, 3, 3, 3, 3, 3, 3].

The extracted \( \tau_{\phi}(T) \) between 0.5 and 20 K for each of our films is least-squares fitted to Eq. (1), and the fitted values of the relevant parameters \( (\tau_{\phi}^0, A_{\text{ep}}, \text{and } p) \) are listed in Table 1. Figure 3 shows a plot of the variation of \( 1/\tau_{\phi} \) with temperature for the PdAg17 thick film. The symbols are the experimental data. The thick solid
creasing level of disorder ($\rho$) is very similar, i.e., $1/\tau_{ep}$ is the fitted electron dephasing time as $T \to 0$. $\tau_{ep}$ and $p$ are the fitted strength of $e$-$\phi$ coupling and effective exponent of temperature, respectively, in $1/\tau_{ep} = A_{ep} T^p$.}

| Sample   | $t$ (Å) | $\rho_0$ (µΩ cm) | $D$ (cm$^2$/s) | $k_F \ell$ | $\tau_{ep}^0$ ($10^{-10}$ s) | $A_{ep}$ (10$^5$ s$^{-1}$ K$^{-p}$) | $p$  |
|----------|---------|-------------------|----------------|------------|-----------------------------|-----------------------------------|-----|
| PdAg11†  | 3900    | 281               | 0.36           | 0.93       | 2.8                         | 1.5                               | 2.3±0.1 |
| PdAg15†  | 5100    | 235               | 0.43           | 1.1        | 1.1                         | 2.4                               | 1.9±0.2 |
| PdAg08†  | 3900    | 224               | 0.45           | 1.2        | 6.7                         | 2.0                               | 2.4±0.1 |
| PdAg12†  | 4800    | 183               | 0.55           | 1.4        | 3.7                         | 2.8                               | 2.2±0.1 |
| PdAg18   | 5000    | 178               | 0.56           | 1.5        | 4.3                         | 0.40                              | 2.4±0.1 |
| PdAg17   | 4500    | 101               | 0.99           | 2.6        | 1.6                         | 2.2                               | 2.2±0.1 |
| PdAg19   | 4000    | 98                | 1.0            | 2.6        | 1.6                         | 2.9                               | 2.3±0.1 |
| PdAg14†  | 4100    | 90                | 1.1            | 2.9        | 1.1                         | 2.5                               | 2.2±0.2 |
| PdAg16   | 3300    | 74                | 1.3            | 3.5        | 0.97                        | 3.6                               | 2.2±0.1 |

The curve drawn through the data points is obtained with $\tau_{ep}^0$, $A_{ep}$, and $p$ as free parameters. In this case, we obtain a temperature exponent $p = 2.2 \pm 0.1$. For comparison, we have also least-squares fitted the measured $1/\tau_{ep}$ with Eq. (1), but with $p$ fixed at an integer value of either 2, 3, or 4 (while allowing $\tau_{ep}^0$ and $A_{ep}$ to vary). The dotted, dashed, and thin solid curves in Fig. 3 plot the fitted results with $p = 2, 3,$ and 4, respectively. It is clearly seen that our temperature dependence of $1/\tau_{ep}$ can be best described with an exponent $p$ equal or close to 2. In fact, we have found that the temperature behavior of $\tau_{ep}$ for all films listed in Table I is very similar, i.e., $1/\tau_{ep}$ demonstrates an essentially quadratic temperature dependence.

Figure 3: Electron dephasing rate as a function of temperature for the PdAg17 thick film. The thick solid curve drawn through the data points is a least-squares fit to Eq. (1), using $p$ as a free parameter. The dotted, dashed, and thin solid curves are least-squares fits to Eq. (1) with $p$ fixed at 2, 3, and 4, respectively (see text).

Taken together, Fig. 3 and Fig. 4 demonstrate that the $e$-$\phi$ scattering in PdAg possesses an anomalous temperature and disorder dependence of $1/\tau_{ep} \propto T^2 \ell$. This dependence is insensitive to the fabrication method. Such a $T^2 \ell$ behavior is totally unexpected, even qualitatively, in terms of the current theoretical concepts for the $e$-$\phi$ interaction in impure conductors. According to the “orthodox” $e$-$\phi$ interaction theory for disordered metals [11, 12, 13], that assumes a coherent motion of the impurity atoms with the deformed lattice atoms at low temperatures, one should expect a $T^4 \ell$ dependence. Recently, it was speculated that, in real metals containing heavy (light) impurities and tough boundaries, the impurity and/or boundary atoms might not move in phase with...
potential scatterers drastically changes the e-phonon interaction, and the relaxation rate is proportional to $T^2 L^{-1}$, where $L$ is the electron mean free path with respect to the static impurities ($L > \ell$). Experimentally, a $T^4$ temperature dependence has been observed very recently in disordered Hf and Ti thin films. A $T^4$ dependence has been previously observed in Bi thin films over a very limited temperature range of 0.6–1.2 K. However, to the best of the authors’ knowledge, the combined $T^4\ell^1$ law has never been confirmed in real conductors thus far. On the other hand, a distinct $T^2\ell^{-1}$ dependence has been observed in Ti$_{1-x}$Al$_x$ and Ti$_{1-x}$Sn$_x$ alloys. Previously, a $T^2\ell$ dependence was independently found in AuPd thick films ($t > 4000$ Å) and Nb thin films ($t \lesssim 200$ Å). In the present case of PdAg thick films, the masses of the Pd and Ag atoms are quite similar, and the films are three-dimensional. Therefore, it is not clear how the Sergeev-Mitin theory evoking heavy (light) impurities and tough boundaries can apply to this case.

The criterion for the e-phonon interaction to satisfy the dirty-limit condition is $qT\ell \ll 1$, where the wave number of the thermal phonons $q_T \approx k_B T/\hbar v_s$, and $v_s$ is the speed of sound. Taking $v_s \approx 2600$ m/s and $\ell \approx 2–8$ Å, we obtain $qT \approx (0.01–0.04) T$ for our PdAg thick films. The phase-breaking lengths $\sqrt{D\tau_p}$ in our films are calculated to be 690–1500 Å at 2 K. (The dephasing length essentially saturates below about 2 K.) This length scale justifies the use of three-dimensional weak-localization theory to describe our experimental magnetoresistivities.

IV. CONCLUSION

We have measured the e-phonon scattering time $\tau_{ep}$ in DC and RF sputtered PdAg thick films. In both series of films, we observe an anomalous $1/\tau_{ep} \propto T^2\ell$ temperature and disorder dependence. Moreover, the e-phonon coupling is found to be much stronger in the DC than RF sputtered films. This observation strongly indicates that the e-phonon interaction not only is sensitive to the total level of disorder but also is sensitive to the microscopic quality of the disorder. These results pose a new theoretical challenge.

Acknowledgments

We are grateful to V. Mitin, A. Sergeev, and G. Y. Wu for valuable discussions. This work was supported by the Taiwan National Science Council through Grant Nos. NSC90-2112-M-009-037 and NSC90-2119-M-007-004.
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