Quadratic temperature dependence of the electron-phonon scattering rate in disordered metals

S. Y. Hsu
Department of Electrophysics, National Chiao Tung University, Hsinchu 300, Taiwan

P. J. Sheng and J. J. Lin
Institute of Physics, National Chiao Tung University, Hsinchu 300, Taiwan

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We have measured the electron-phonon scattering rates $1/\tau_{ep}$ from a weak-localization study of a series of high-resistivity tin-doped $Ti_{73}Al_{27}$ alloys. The resistivities of these alloys are essentially the same and are so high that the electron elastic mean free path l approaches the interatomic spacing, resulting in a very small value of $q_{ph}l \approx 0.0056\,T$ (q_{ph} is the wave number of the thermal phonons, and T is the temperature). Based on as many as about 60 magnetoresistivity measurements between 2 and 22 K, we are able to determine the dependence of $1/\tau_{ep}$ on T to within a great degree of accuracy and find that $1/\tau_{ep}$ varies essentially with T^2 in the dirty limit ($q_{ph}l \ll 1$). This observation is not understood in terms of the current theoretical concept of electron-phonon interaction in disordered metals. [S0163-1829(99)05030-4]

I. INTRODUCTION

The electron-phonon scattering rate $1/\tau_{ep}$ is one of the most important physical quantities of metals. In the case of pure metals, the temperature behavior of $1/\tau_{ep}$ is well understood both theoretically and experimentally.[1] In the presence of strong impurity scattering, however, the issue still remains open. Theoretically, electron-phonon interaction in disordered metals has been studied by a good number of authors[2–5] for over two decades and widely varied results were obtained. Recently, it has been widely accepted that a consensus has finally been reached in theoretical efforts.[3–5] On the other hand, few experiments have successfully provided an overall consistency check for the various aspects of the theoretical predictions. For instance, apart from the dependence on the electron elastic mean free path l, the expected T^4 dependence of $1/\tau_{ep}$ is very frequently (or almost) unseen in experiments.[6] It has been argued that the absence of the expected T^4 law in the various experiments can be ascribed to the fact that most material systems studied previously are not yet strongly disordered enough for the electron-phonon interactions to strictly satisfy the dirty-limit criterion of $q_{ph}l \ll 1$ (where q_{ph} is the wave number of the thermal phonons). Information about $1/\tau_{ep}$ on T in sample systems having smaller values of $q_{ph}l$ than those ever obtained in the literature is therefore of prime importance for a stringent justification for the electron-phonon interaction theory concerning disordered metals.

It is now well established that weak-localization studies can be used to extract various electron dephasing scattering times in disordered metals, including inelastic scattering time, spin-orbit scattering time (τ_{so}), and magnetic spin-spin (or, some kind of intrinsic nonthermal) scattering time (τ_s). In particular, in the case of *three* dimensions, unlike the cases of reduced dimensions, electron-phonon scattering is the *sole*, significant inelastic process while the small energy-transfer (''quasielastic'') electron-electron scattering is not important.[3] Therefore, the value of $1/\tau_{ep}$ can be reliably extracted from weak-localization studies using bulk samples. According to the theory, the weak-localization effects in those disordered systems having moderate to strong spin-orbit scattering are controlled by two parameters, $1/\tau_{so}$ and $1/\tau_\phi(T)$, where the electron dephasing scattering rate $1/\tau_\phi$ reads[7]

$$1/\tau_\phi(T) = 2/\tau_s + 1/\tau_{ep} = 2/\tau_s + AT^p. \tag{1}$$

Notice that, for the reason just discussed above, we have identified the inelastic scattering rate with the electron-phonon scattering rate and written $1/\tau_{ep} = AT^p$, where A characterizes the strength of the electron-phonon interaction and p is the exponent of temperature for $1/\tau_{ep}$. Theoretically, it is expected that ''weakened'' electron-phonon interaction in the presence of strong impurity scattering results in $p=4$ in the dirty limit ($q_{ph}l \ll 1$), compared with $p=3$ in the clean limit ($q_{ph}l \gg 1$).

In this work, we have successfully fabricated a series of bulk crystalline disordered $Ti_{73-x}Al_{27}Sn_x$ alloys with the nominal concentration of tin $0 \lesssim x \lesssim 5$. The tractable doping of minute amounts of tin atoms into the parent $Ti_{73}Al_{27}$ phase enhances spin-orbit scattering in the samples in a controllable manner, while leaving the amounts of disorder, i.e., resistivities, of the samples barely changed. This unique material property allows a convincing consistency check of our experimental method and, particularly, a systematic extraction of the electron-phonon scattering rates to within a great degree of accuracy. Based on as many as about 60 weak-localization-induced magnetoresistivity measurements on the various alloys held at various temperatures between 2 and 22 K, we have drawn a conclusion that $1/\tau_{ep}$ depends essentially quadratically on the temperature T in disordered metals.

II. EXPERIMENTAL METHOD

A series of bulk crystalline disordered $Ti_{73-x}Al_{27}Sn_x$ alloys were fabricated by a standard arc-melting method as described previously.[8] The nominal concentration of tin x had been kept low enough ($x \lesssim 5$) so that all the alloy

samples studied were single phased. X-ray diffraction measurements confirmed that all of our ternary alloy samples possessed a structure similar to that of the parent $Ti_{73}Al_{27}$ phase; no noticeable impurity peaks in the diffraction patterns were found.

Our alloy system, which was derived from the parent $Ti_{73}Al_{27}$ phase (the so-called ''ordered'' α_2 phase in terms of crystallographic nomenclature), had been chosen because the structure and material properties of the $Ti_{73}Al_{27}$ phase were well established in the literature.[9] In addition, it had been one of our aims to compare the electron-phonon scattering rate in this system with that in the dilute $Ti_{100-y}Al_y$ alloy system with $y \lesssim 12$ (the so-called ''disordered'' α phase in terms of crystallographic nomenclature). The electron-phonon scattering rate, particularly its temperature and electron elastic mean free path dependence, has been extensively studied in the latter system.[8] It is thus of interest to examine how the inelastic electron-phonon process might, or might not, change from the α phase to the α_2 phase. Moreover, there are further advantages for choosing $Ti_{73-x}Al_{27}Sn_x$ as our system material: (1) Tractable doping of minute amounts of tin atoms into the parent $Ti_{73}Al_{27}$ phase, which contains no heavy atoms and hence possesses moderate spin-orbit scattering, is metallurgically feasible, causing a gradual increase in the strength of spin-orbit scattering from the moderate toward the strong limit in a controllable manner. This material property allows a convincing consistency check of our experimental method and a systematic extraction of the values of $1/\tau_{ep}$ (see below). (2) The resistivities of these tin-doped alloys are fairly large, being with $\rho(300\ \mathrm{K}) \approx 250\ \mu\Omega$ cm and $\rho_0 = \rho(10\ \mathrm{K}) \approx 225\ \mu\Omega$ cm. Such large amounts of disorder, i.e., resistivities, result in measurable weak-localization-induced magnetoresistivities. More importantly, these *extremely high* values of resistivity (corresponding to an electron elastic mean free path being on the order of the interatomic spacing) put the electron-phonon interaction in this material system *much closer* to the dirty limit ($q_{ph}l \ll 1$) than ever obtained in any other material system previously studied by us, including dilute $Ti_{100-y}Al_y$ alloys[8] and $Au_{50}Pd_{50}$ alloys.[10] (3) The sample resistivities of $Ti_{73-x}Al_{27}Sn_x$ alloys *barely* change with the concentration of tin x. This unique material property therefore enables us to concentrate on the temperature dependence of $1/\tau_{ep}$ while exempting us from any complications that might arise from a variation of $1/\tau_{ep}$ with the disorder. (It is understood that $1/\tau_{ep}$ is likely mean-free-path dependent in the dirty limit.)

The magnetoresistivities of our alloy samples, typically $0.2 \times 1 \times 10\ \mathrm{mm}^3$, were measured by a standard four-probe technique between 2 and 22 K and in magnetic fields below 1.4 T. The magnetoresistivities at various measuring temperatures for each alloy were compared with three-dimensional weak-localization theory[7] to extract the electron dephasing scattering rate $1/\tau_\phi$ and the spin-orbit scattering rate $1/\tau_{so}$. The details of the least-squares fitting procedure had been discussed previously.[8] Here we merely stress that, for every alloy sample studied in this work, the three-dimensional weak-localization predictions with a moderate to somewhat strong spin-orbit scattering rate, depending on the concentration of tin, can well describe our experimental results. Thus, $1/\tau_\phi$ and $1/\tau_{so}$ can be reliably extracted. It is worth stressing that, between the two adjusting parameters, only $1/\tau_\phi$ is temperature dependent while a single value of $1/\tau_{so}$ is used to describe 10 or more magnetoresistivity curves for a given alloy sample.

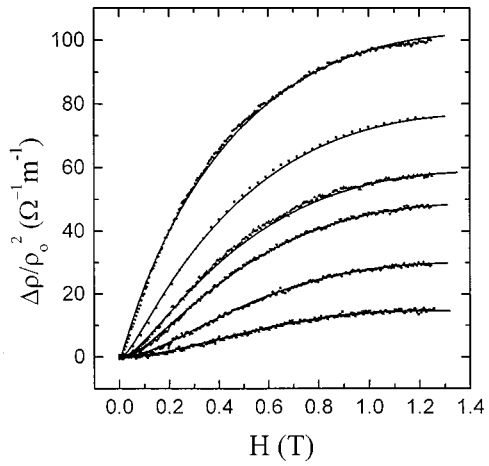

FIG. 1. Normalized magnetoresistivities $\triangle\rho(B)/\rho^2(0)$ as a function of the magnetic field for the $Ti_{70}Al_{27}Sn_3$ alloy at (from top down) 2.00, 5.00, 8.00, 10.0, 15.0, and 20.0 K. The symbols are the experimental results, and the solid curves are the three-dimensional weak-localization predictions.

III. RESULTS AND DISCUSSION

Figure 1 shows the measured, normalized magnetoresistivities $\triangle\rho(B)/\rho^2(0) = [\rho(B) - \rho(0)]/\rho^2(0)$ and the three-dimensional weak-localization predictions for a representative alloy sample, $Ti_{70}Al_{27}Sn_3$, at several measuring temperatures as indicated in the caption to Fig. 1. The symbols are the experimental data and the solid curves are the theoretical results. It is clearly seen that the theoretical predictions can well describe the experimental data. To compute $1/\tau_\phi$ from the electron dephasing scattering field $B_\phi = \hbar/4eD\tau_\phi$ defined in the weak-localization theory, one needs the value of the electron diffusion constant D. Using the measured ρ_0 and the density of states at the Fermi level, $N(0)$, independently determined from specific heat measurements,[11] we estimate the values of D for our alloys through the Einstein relation $1/\rho_0 = N(0)e^2D$. We obtain $D \approx 0.85\ \mathrm{cm}^2/\mathrm{s}$ for all our tin-doped samples having $\rho_0 \approx 225\ \mu\Omega$ cm.

Figure 2 shows the variation of the spin-orbit scattering rate $1/\tau_{so} = 4eDB_{so}/\hbar$ (where B_{so} is defined in the weak-localization theory) with the concentration of tin x for eight $Ti_{73-x}Al_{27}Sn_x$ alloys studied. It is encouraging to see from Fig. 2 that our experimental value of $1/\tau_{so}$ increases linearly from about 5.2×10^{10} to about $3.4 \times 10^{11}\ \mathrm{s}^{-1}$ as x increases from 0 to 5. This observation is strongly suggestive of the fact that our alloy samples possess homogeneous composition and lattice structure at length scales that are considerably smaller than the relevant length scale in the weak-localization problem (i.e., the electron diffusion length). This result thus provides a convincing crosscheck of our experimental method (sample fabrication, for instance) and data analysis. Therefore, our extracted values of $1/\tau_{ep}$ should be very reliable, which in turn makes our determination of the exponent of temperature p of $1/\tau_{ep}$ reliable to within a great degree of accuracy.

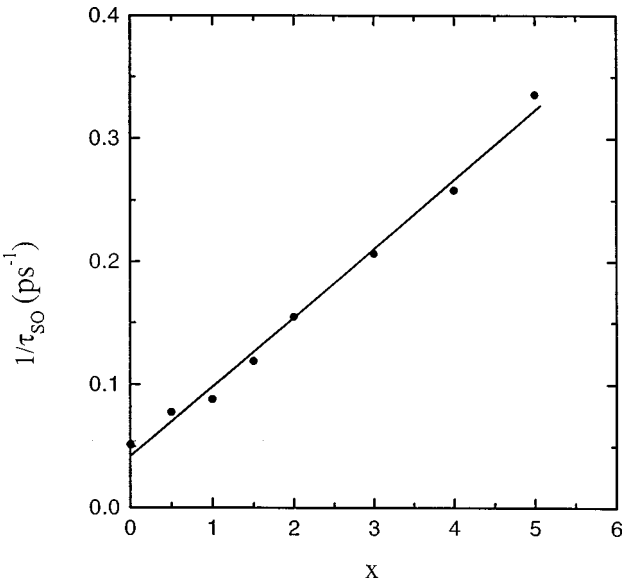

FIG. 2. Spin-orbit scattering rates $1/\tau_{so}$ as a function of the concentration of tin x for several $Ti_{73-x}Al_{27}Sn_x$ alloys. The straight line is a guide to the eye.

Our main results of the present work, i.e., the extracted values of the electron dephasing scattering rate $1/\tau_\phi(T)$ are summarized in Fig. 3. Figure 3 shows the variation of $1/\tau_\phi$ with temperature for eight $Ti_{73-x}Al_{27}Sn_x$ alloys studied. Different symbols designate the $1/\tau_\phi$ for different alloy samples. However, since the values of $1/\tau_\phi$ are very similar for all samples (as is evident in Fig. 3), it is thus not necessary to label explicitly each symbol with its associated particular alloy. Inspection of this figure clearly illustrates that spin-orbit scattering has little, if any, effect on either the magnitude or the temperature dependence of $1/\tau_\phi$ (and hence $2/\tau_s$ and $1/\tau_{ep}$) in disordered metals. (Recall that all the alloy samples studied in this work have very similar resistivities. They should then possess very similar values of $1/\tau_{ep}$, regardless of how $1/\tau_{ep}$ might depend on the electron elastic mean free path l.)

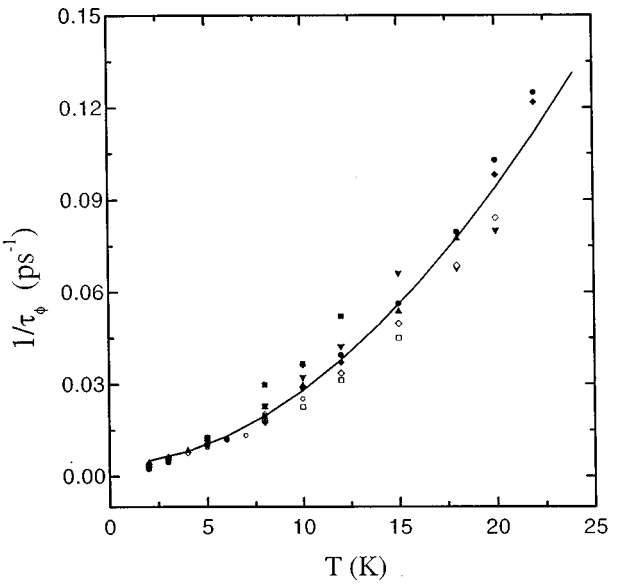

FIG. 3. Electron-phonon scattering rates $1/\tau_{ep}$ as a function of temperature for several tin-doped $Ti_{73}Al_{27}$ alloys. The solid line is a least-squares fit to Eq. (1).

The solid line drawn through the data points in Fig. 3 is a least-squares fit to Eq. 1 with the inelastic electron scattering strength A, the exponent of temperature p, and the residual scattering rate $2/\tau_s$ as adjusting parameters. One sees that Eq. 1 can well describe our experimental data of $1/\tau_\phi$ over the wide temperature range of 2–22 K. Experimentally, as many as about 60 magnetoresistivity curves, corresponding to about 60 experimental data points for $1/\tau_\phi$, have been measured on our various alloys and used in the fits with Eq. 1. Therefore, any appreciable statistical uncertainties in the extraction of the values of the adjusting parameters can be largely minimized. With all the eight alloy samples taken together in the fits, our best fitted values for the adjusting parameters are the following: the inelastic scattering strength $A \approx 2.9\times10^8$ s^{-1}/K^p, the exponent of temperature $p=1.9\pm0.2$,[12] and the residual scattering rate $2/\tau_s \approx 4.0\times10^9$ s^{-1}. Most noticeably, our experimental value of p implies that $1/\tau_{ep}$ varies *essentially quadratically* with temperature in bulk crystalline disordered $Ti_{73}Al_{27}$ alloys. This experimental result of $p\approx2$ is substantially lower than the theoretical value of $p=4$ expected for ''weakened'' electron-phonon scattering in strongly disordered metals. For reference, our fitted value of $1/\tau_{ep}(10\text{ K}) \approx 2.4\times10^{10}$ s^{-1} is reasonably close to that [$\approx(1-3)\times10^{10}$ s^{-1}] observed in the above-mentioned dilute $Ti_{100-y}Al_y$ alloys [where the impurity resistivities $\rho_0 \approx 60-150$ $\mu\Omega$ cm (Ref. 8)].

In order to perform a crosscheck and so as to provide further confirmation of the T^2 dependence of $1/\tau_{ep}$, in addition to tin-doped $Ti_{73}Al_{27}$ alloys, we have in this work made two $Ti_{73-z}Al_{27}Au_z$ alloys with $z=0.5$ and 1, respectively, and measured weak-localization-induced magnetoresistivities to extract $1/\tau_{ep}$. Again, we obtain $p\approx2.1$ in these two gold-doped alloys, supporting our above assertion of an essentially quadratic temperature dependence of $1/\tau_{ep}$ in the $Ti_{73}Al_{27}$ phase. However, because these two gold-doped alloys are already quite close to the limit of strong spin-orbit scattering (due to the large atomic number of the heavy gold atoms), an experimental determination of a precise value of $1/\tau_{so}$ is less feasible and, in fact, is not of much interest in this work.

Since we are most concerned with the electron-phonon scattering in the disordered limit, we examine whether the disorder criterion $q_{ph}l\lesssim1$ is satisfied in the present experiment, where $q_{ph}\approx k_BT/\hbar v_s$ (v_s being the sound velocity) is the wave number of the thermal phonons at temperature T. For the high-resistivity $Ti_{73}Al_{27}$ phase, $v_s\approx4\times10^3$ m/s,[13] and $l\approx1.6$ Å.[14] We notice that, due to the extremely high value of resistivities, this amount of electron mean free path already approaches that of the interatomic spacing, causing a very small value of $q_{ph}l$. Quantitatively, we obtain $q_{ph}l \approx 0.0056\,T$, where T is in K. This indicates that the electron-phonon processes in our alloy samples are well within the disordered limit, i.e., $q_{ph}l\ll1$, even at our highest measuring temperature of 22 K. We stress that the value of $q_{ph}l$ for the present material system is *much lower* than those in the other material systems which have previously been studied by our group, including dilute $Ti_{100-y}Al_y$ alloys[8] [where $q_{ph}l \approx (0.0068-0.019)T$] and $Au_{50}Pd_{50}$ alloys[10] [where $q_{ph}l \approx (0.024-0.078)T$]. The present experimental result thus

provides valuable evidence in supporting the T^2 dependence of $1/\tau_{ep}$ in disordered metals. This observation should cause us to rethink what heretofore has been taken for granted concerning electron-phonon scattering in strongly impure metals. For comparison, we mention that the T^2 dependence of $1/\tau_{ep}$ has also been observed in our dilute $Ti_{100-y}Al_y$ and $Au_{50}Pd_{50}$ alloys. On the other hand, there are few experiments[6,15] reported in the literature that had dealt with samples which had values of $q_{ph}l$ smaller than those in our various samples of either dilute $Ti_{100-y}Al_y$, $Au_{50}Pd_{50}$, or tin-doped $Ti_{73}Al_{27}$ alloys.

Recently, the theory for electron-phonon interaction in disordered metals has been re-examined in the literature. For instance, Rammer and Schmid[3] have treated this problem by considering impurity atoms that move in phase with the other lattice atoms and predicted that $1/\tau_{ep}$ should be weakened and be of the order of $(q_{ph}l)(1/\tau_{ep}^0)$, where $1/\tau_{ep}^0$ is the electron-phonon scattering time in the pure metal ($1/\tau_{ep}^0 \sim T^3$). Microscopically, it is predicted that both longitudinal and transverse phonons would contribute to the weakened electron scattering rate $1/\tau_{ep}$, while only longitudinal phonons are responsible for $1/\tau_{ep}^0$ in the pure case. At low temperatures, since $q_{ph} \approx k_B T/\hbar v_s$, then $1/\tau_{ep}$ should follow the $T^4 l$ law. This prediction has received wide acceptance from the theoretical community, and it has been independently confirmed by the calculations of Reizer and Sergeyev[4], and Belitz.[5] Our experimental result of the dependence of $1/\tau_{ep}$ on the square of temperature (Fig. 3) is, however, in disagreement with this Rammer-Schmid prediction. The physical origin for the T^2 behavior, which has been observed experimentally from time to time in various materials,[16] is still not understood.[17]

Using the values of τ_ϕ and the values of D given above, we estimate the electron dephasing scattering length, $L_\phi = \sqrt{D\tau_\phi}$, for our alloy samples to range from about 300 to about 1600 Å as the temperature decreases from 22 down to 2 K. That is, every alloy sample studied lies well within the three-dimensional regime, justifying our use of the three-dimensional weak-localization predictions to describe the experimental magnetoresistivities.

IV. CONCLUSION

We have measured the electron-phonon scattering rates in a serious of carefully tailored bulk crystalline disordered $Ti_{73-x}Al_{27}Sn_x$ alloys which have a very short electron elastic mean free path being on the order of the interatomic spacing. Such a short electron mean free path causes the electron-phonon interaction to be *well within* the dirty limit in this material. We find an essentially quadratic temperature dependence of the electron-phonon scattering rate $1/\tau_{ep}$, i.e., $1/\tau_{ep} \sim T^2$ at $q_{ph}\, l \ll 1$. This observation is not understood in terms of the current theoretical concept for the weakened electron-phonon interaction in disordered metals.

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[12] If the experimental value of $1/\tau_\phi(T)$ for each of our alloy sample is treated separately and fitted to Eq. (1), then we obtain the exponent of temperature p falling in the range 1.7–2.1 for the various alloys.

[13] This value of v_s is evaluated using the average speed of sound $v_s(\mathrm{Ti}) \approx v_s(\mathrm{Al}) \approx 4000$ m/s.

[14] The value of the Fermi wave number k_F for the $Ti_{73}Al_{27}$ phase is evaluated using $k_F(\mathrm{Ti}) \approx 1.9 \times 10^{10}\ \mathrm{m}^{-1}$ and $k_F(\mathrm{Al}) \approx 1.75 \times 10^{10}\ \mathrm{m}^{-1}$. The electron elastic mean free path is then obtained through the relation $l = 3\pi^2\hbar/(k_F^2 e^2 \rho_0)$.

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