Anomalous temperature and disorder dependences of electron-phonon scattering rate in impure $V_{1-x}Al_x$ alloys

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Using weak-localization studies, we have measured the electron-phonon scattering time $\tau_{ep}$ in a series of $V_{1-x}Al_x$ alloys having the characteristic of $ql \ll 1$, where $q$ is the wave number of the thermal phonons and $l$ is the electron elastic mean free path. We find an anomalous electron-phonon scattering rate obeying a quadratic temperature dependence as well as a linear mean-free-path dependence, i.e., $\tau_{ep}^1 \sim T^2l$. This result cannot be explained in terms of existing theories for the electron-phonon interaction in impure conductors.

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I. INTRODUCTION

Disordered A15 compounds continue to be an interesting subject in condensed-matter physics due to their unusual normal-state electrical-transport properties. Physically, it has been established over the last two decades that both weak-localization (WL) and electron-electron interaction (EEI) effects play essential roles in understanding the electronic conduction in disordered metals. Although both these effects introduce temperature-dependent corrections to resistivity at low temperatures, it is known that, in the case of three-dimensional (3D) disordered metals, the temperature dependence of conductivity in zero magnetic field is dominated by the EEI correction, and the field dependence in low fields is dominated by the WL correction. Particularly, it is established that analysis of the magnetoresistances due to the WL effects could provide quantitative information of various electron dephasing times for the electron waves, such as inelastic, spin-orbit, and the “zero-temperature” scattering times. Among these three dephasing scattering times, spin-orbit and zero-temperature scattering times are (presumed to be) independent of temperature, only the inelastic-scattering time is temperature dependent and is often of greatest interest, since it provides information for the microscopic interactions between the electrons and electrons and between the electrons and phonons. Many theoretical and experimental works reported in the literature have established that the Nyquist electron-electron scattering is the dominant dephasing process in reduced-dimensional systems.

On the contrary, in the case of 3D, electron-electron scattering is not important, only electron-phonon ($e$-$ph$) scattering is the significant inelastic process. Although in the clean limit of $ql \gg 1$ (where $q$ is the wave number of the thermal phonons and $l$ is the electron elastic mean free path), the temperature behavior of the $e$-$ph$ scattering time $\tau_{ep}$ is well understood, the nature of $e$-$ph$ interaction in the dirty limit of $ql \ll 1$ remains a long-standing puzzle. For almost three decades, many theoretical efforts have been made to study the $e$-$ph$ scattering time in impure materials, reaching at quite different predictions. Experimentally, the theoretically predicted $T^4$ dependence of $\tau_{ep}^1$ in the dirty limit has recently been reported in a few cases, but only over very limited temperature ranges. Apart from a good deal of studies reported in the literature, to the authors’ knowledge, the behavior of $\tau_{ep}$ in A15 compounds has not been studied by any experimental and, particularly, the WL method. In this work, we thus propose to systematically measure $\tau_{ep}$ from WL studies in a series of disordered $V_{1-x}Al_x$ alloys whose compositions are close to the superconducting A15 $V_3Al$ compound.

II. EXPERIMENTAL METHOD

A series of $V_{1-x}Al_x$ alloys were prepared by a standard arc-melting method. Appropriate amounts of V (99.7% pure) and Al (99.999% pure) were melted several times. Some of the melted ingots were placed at 800°C for 48 h for homogeneity. Aluminum atoms were introduced into the V host to serve as compositional disorder in the samples. In this work, the nominal Al concentration $x$ was kept in the range of 0.18–0.26 so that all the samples fabricated were essentially single phased and possessed the cubic crystal structure characteristic to the superconducting A15 $V_3Al$ compound. X-ray-diffraction studies confirmed that our alloys possessed bcc structure and the lattice of the alloys underwent an expansion with increasing solute contents of Al. The measured lattice parameters $a$ of our alloys are listed in Table I.

We chose Al as impurity atom mainly because the presence of Al in a V host could result in relatively large values of residual resistivity, which in turn produced comparatively pronounced 3D WL-induced magnetoresistances. The values of impurity resistivity $\rho_0$ [ps (10 K)] and relevant parameters for different alloys are listed in Table I. Our experimental values of $\rho_0$ varied from 144 to 212 $\mu\Omega$ cm, corresponding to values of $k_p$ ranging from about 3.1 to about 4.6, where we used the evaluated Fermi wave number $k_p = 1.84 \times 10^{14}$ m$^{-1}$. These values of $\rho_0$ are consistent with previous measurements. It should be noted that it has previously been established, both experimentally and theoretically, that $\rho_0$ increases linearly with increasing $x$ over a wide range.
TABLE I. Values of relevant parameters for V_{1−x}Al_x alloys. a is the lattice parameter for the bcc structure of our alloys.

<table>
<thead>
<tr>
<th>Sample</th>
<th>ρ_0 (μΩ cm)</th>
<th>ρ(300 K)/ρ_0</th>
<th>a (Å)</th>
<th>D (cm²/s)</th>
<th>k_p l</th>
<th>T_c (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>x=0.18</td>
<td>114</td>
<td>1.03</td>
<td>3.0520</td>
<td>1.59</td>
<td>4.6</td>
<td>0.77</td>
</tr>
<tr>
<td>x=0.20</td>
<td>147</td>
<td>1.02</td>
<td>3.0530</td>
<td>1.55</td>
<td>4</td>
<td>1.75</td>
</tr>
<tr>
<td>x=0.22</td>
<td>183</td>
<td>0.995</td>
<td>3.0543</td>
<td>1.25</td>
<td>3.6</td>
<td>0.98</td>
</tr>
<tr>
<td>x=0.24</td>
<td>203</td>
<td>1.02</td>
<td>3.0597</td>
<td>1.12</td>
<td>3</td>
<td>1.60</td>
</tr>
<tr>
<td>x=0.26</td>
<td>212</td>
<td>0.981</td>
<td>3.0677</td>
<td>1.08</td>
<td>3.1</td>
<td>0.63</td>
</tr>
</tbody>
</table>

from x=0.05 to 0.35 in V_{1−x}Al_x. In contrast to a naive speculation, the residual resistivity ρ_0 in this alloy system does not reveal a minimum at the stoichiometric value of x ≈ 0.25.

The samples were cut into specimens of rectangular shape, typically ≈0.2 × 0.2 × 10 mm³, for resistivity measurement. Platinum electrodes were spot welded onto the samples. Resistances and magnetoresistances were measured by using the standard four-probe technique, and were performed on either a ⁴He or a ³He cryostat.

Upper critical fields B_{c2} for our alloys were measured and the values of the electron diffusion constant D were then calculated by using the relation

\[ D = (4k_B/\pi e)|dB_{c2}/dT| \]

where k_B is the Boltzmann constant and e is the electronic charge. We had also measured the electronic specific heat, γT = π²κ_F²N(0)T/3, for two alloys (x=0.18 and 0.20) to deduce the e-ph enhanced electronic density of states at the Fermi level N(0). The values of D were then obtained through the Einstein relation 1/ρ_0 = De²N(0)/(1 + λ), where the e-ph coupling constant λ was taken to be 0.46 (Ref. 18).

We observed that the values of D deduced from B_{c2} measurement and γT measurement were consistent to within 20%. Taking the average of D values measured by these two ways, we obtained the relation D = (229/ρ_0)cm²/s for our V_{1−x}Al_x alloys studied in this work, where ρ_0 is in μΩ cm.

\[ -\alpha = \frac{4k_B}{\pi e^2} \]  

where α is a disorder dependent constant. A plot of Δρ(T)/ρ_0 = [ρ(T) − ρ_0]/ρ_0 are of the order a tenth of a percent. From about 3T_c up to 20 K, the change in resistivity obeys the relation

\[ \Delta \rho(T) = -\alpha \sqrt{T} \]  

III. RESULTS AND DISCUSSION

We have performed measurements of the resistivities and magnetoresistivities of polycrystalline disordered V_{1−x}Al_x alloys. The superconducting transition temperature T_c of stoichiometric A15 V_3Al compound is known to be 9.6 K, whereas for our highly resistive V_{1−x}Al_x alloys investigated in this work, T_c reduces to between 0.6 and 1.8 K. An erratic variation is observed in the values of T_c listed in Table I. The presence of imperfections, which cannot be ruled out in the disordered alloy systems, may be attributed to such behavior of T_c. This large reduction in T_c from 9.6 K down to ~1 K makes these tailored samples advantageous for quantitative study of τ_{ep} for over a wide range of temperature from about 2 or 3 K up to 20 K. This is because at temperatures above but sufficiently close to T_c, scattering of the conduction electrons by superconducting fluctuations might also contribute to the total inelastic electron time (τ_l), in addition to the e-ph scattering. This situation makes it less desirable for a quantitative separation of τ_{ep} from the measured τ_l.

In order to provide knowledge about the electrical-transport properties of the investigated alloys, we have carefully characterized the temperature and disorder dependences of resistivity in zero magnetic field. Such information would be useful for good understanding and theoretical calculations of τ_{ep} (to be addressed below) for general disordered metals. We find that, at low temperatures, the resistivity of all alloys increases with reducing temperature. The magnitudes of the normalized resistivity rise Δρ(T)/ρ_0 = [ρ(T) − ρ_0]/ρ_0 are of the order a tenth of a percent. From about 3T_c up to 20 K, the change in resistivity obeys the relation

\[ \Delta \rho(T) = -\alpha \sqrt{T} \]  

where α is a disorder dependent constant. A plot of Δρ(T) with √T for different samples are shown in Fig. 1. The straight solid lines in the figure are obtained from least-squares fits with Eq. (1), taking α as an adjusting parameter for each alloy. As mentioned, in 3D disordered metals, the temperature dependence of resistivity at zero magnetic field...
is dominated by the EEI correction.\textsuperscript{2} According to the theory of EEI,\textsuperscript{2} apart from the $\sqrt{T}$ dependence, the resistivity correction $\Delta \rho(T)$ is also a function of disorder and proportional to $\rho_D^2/\sqrt{T}$ or $\rho_0^{5/2}$. The inset of Fig. 1 shows the variation of the resistivity slope $\alpha = -\Delta \rho(T)/\sqrt{T}$ with $\rho_0^{5/2}$ for V$_{1-x}$Al$_x$ alloys. It is seen that the slope of the resistivity rise indeed varies linearly with $\rho_0^{5/2}$, firmly supporting the EEI predictions. This observation is suggestive of the fact that the EEI effects are completely controlled by the total amount of disorder (i.e., $\rho_0$) contained in the sample. In short, our samples behave as a “standard” disordered metal insofar as the zero-field resistivity behavior is concerned. Therefore, $\tau_{ep}$ deduced for this standard material deserves a theoretical explanation.

Magnetoresistances of our alloys are measured under magnetic fields below 1 T and in the temperature range of 2–20 K. The measured magnetoresistances are then compared with the 3D WL theoretical predictions, with the Maki-Thompson correction term taken into account,\textsuperscript{19} to extract the values of the electron dephasing time $\tau_e$. The inset of Fig. 2 shows the variation of the magnetoresistance with magnetic field of V$_{0.80}$Al$_{0.20}$ sample. The points are the experimental data and the solid lines are the theoretical best-fitting values. The details of our fitting procedure had been discussed previously.\textsuperscript{19} As expected, the 3D WL theoretical predictions can well reproduce the measured magnetoresistances, and the values of $\tau_e$ are reliably extracted from these fits. Figure 2 is a plot of the experimental $\tau_e^{-1}$ as a function of temperature for the representative V$_{0.80}$Al$_{0.20}$ alloy. The symbols are the experimental data. The solid curve is the least-square fit to

$$1/\tau_e(T) = 1/\tau_0 + 1/\tau_{ep}(T) = 1/\tau_0 + A_{ep} T^p,$$

where the adjusting parameters are the zero-temperature dephasing time $\tau_0$, the strength of $e$-ph coupling $A_{ep}$, and $p$ the exponent of temperature for $\tau_{ep}^{-1}$. In this case, we obtain the best-fit value of $p=2.15\pm 0.16$. The long-dashed and short-dashed curves are also least-square fits to Eq. (2), but now $p$ was taken as fixed at 3 and 4, respectively, while $\tau_0$ and $A_{ep}$ were still allowed to vary. Inspection of this figure indicates that the temperature-dependent part of our measured $\tau_{ep}^{-1}$, i.e., $\tau_e^{-1}$ can only be described with a quadratic temperature dependence (the solid curve). Therefore, Eq. (2) can well describe our experimental $\tau_{ep}$. The values of our best fitted adjusting parameters for different alloys are $\tau_0=(2.0-6.0)\times 10^{-11}$ s, $A_{ep}=(3.7-7.6)\times 10^{5}$ s$^{-1}$ K$^{-p}$, and $p=2.02-2.18$. Our experimental value of $p$ clearly implies that $\tau_{ep}^{-1}$ varies essentially with $T^p$. Similar quadratic temperature behavior had previously been observed in several bulk alloys, such as Ti$_{1-x}$Al$_x$ alloys\textsuperscript{20} and Ti$_{0.73-x}$Al$_{0.27}$Sb$_x$ alloys.\textsuperscript{21} The magnitudes of the fitted $\tau_{ep}$ are comparable with those observed in these titanium alloys.

In the study of the $e$-ph interaction in impure systems, it is of great fundamental importance, but much more difficult, to measure the elastic mean-free-path dependence of $\tau_{ep}$. In this work, we have succeeded in addressing this issue using several V$_{1-x}$Al$_x$ alloys containing various amounts of disorder. We have quantitatively determined the values of $\tau_{ep}$ for five samples. In Fig. 3, we plot the variation of $\tau_{ep}$ with the diffusion constant $D$ for five alloys at a representative temperature of 10 K. Inspection of Fig. 3 clearly reveals that $\tau_{ep}^{-1}(10$ K) varies linearly with $D$ for the samples investigated. This observation firmly establishes the variation $\tau_{ep}^{-1} \sim D \sim l$ in highly resistive V$_{1-x}$Al$_x$ alloys. Similar temperature and disorder behavior of $\tau_{ep}^{-1} \sim T^2$ has previously been reported in three-dimensional Au$_{0.50}$Pd$_{0.50}$ (Ref. 22) and Ag$_{0.40}$Pd$_{0.60}$ (Ref. 23) films, and two-dimensional Sb (Ref. 24) and Nb (Ref. 25) films.

Theoretically, $e$-ph interaction in impure metals has been studied by many authors over the years. Very recently, Sergeev and Mitin\textsuperscript{26} have reconsidered the $e$-ph time in a disorder conductor. They found that, in the case where the random scattering potential is completely dragged by phonons, the effective $e$-ph interaction would decrease due to disorder, resulting in $\tau_{ep}^{-1} \sim T^2$ in the dirty limit.\textsuperscript{3} Moreover, they realized that, in real experiments, the imperfections (defects, impurities, boundaries, etc.) in a disordered conductor would
often not be completely dragged by the phonons. Under such a circumstance of incomplete drag, they observed that the interaction between electrons and transverse phonons would dominate over the interaction between electrons and longitudinal phonons, causing an anomalous $T^2\tau_{ep}^{-1}$ dependence of $\tau_{ep}$ in the dirty limit, where $\tau$ is the electron mean free path with respect to the "static" impurities.

We return to our experimental result of $\tau_{ep}^{-1} \sim T^2\!l$ in disordered $V_{1-x}$Al$_x$ alloys. This observation cannot be understood in terms of the theories of Rammer and Schmid, and Sergeev and Mitin. In fact, it cannot be explained in terms of any existing e-ph interaction theory for impure metals. One might argue that the discrepancy between theory and experiment is not entirely unexpected. For instance, the existing theories are formulated on the basis of a simple model with a spherical Fermi surface. This is certainly inconsistent with the case of the present experimental material system, $V_{1-x}$Al$_x$, where the Fermi surface is complex. Modification of the phonon spectrum from the standard Debye square law in the low-frequency region, in the presence of strong disorder, might also need to be considered in any attempt to formulate a realistic e-ph scattering time.

Since we are most concerned with $\tau_{ep}$ in the dirty limit, we examine whether the disorder criterion $ql \leq 1$ is satisfied in this experiment. Taking $\nu_s \approx 4.1 \times 10^3$ m/s (Ref. 27) for $V_{1-x}$Al$_x$ alloys, we obtain $ql \approx k_B T l / \hbar \nu_s \approx (0.0054 - 0.0079) T$, where $T$ is in kelvin. Notice that, due to the high values of $\rho_0$, the mean free path is very short in these alloys, which produces a very small magnitude of $ql$. Therefore, the e-ph processes in our samples well satisfy the dirty limit criterion of $ql \ll 1$ even at our highest measurement temperatures of 20 K.

IV. CONCLUSION

We have studied the electrical-transport properties of bulk $V_{1-x}$Al$_x$ alloys at low temperatures. We observe, in zero magnetic field, the resistivity rise, $\Delta \rho(T) \sim -\rho_0^{5/2} / \sqrt{T}$, is well described by the EEI effects in 3D. In addition, we have measured $\tau_{ep}$ in the dirty limit of $ql \ll 1$. Our result reveals an unusual variation of $\tau_{ep}^{-1} \sim T^2\!l$. Such a variation has previously also been observed in a "normal" (i.e., non-A15) metal alloy AuPd, but a pertinent theoretical understanding is still lacking.

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