Electron-electron interaction and weak-localization effects in Ti-Al alloys

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We have measured the resistivities ρ of $Ti_{1-x}Al_x$ alloys ($0.00 \le x \le 0.19$) between 4 and 300 K. As the temperature T is lowered to liquid-helium temperatures, a resistivity rise $[\rho(T)-\rho(25\ \mathrm{K})]/\sqrt{T} \sim -\rho^{2.5}(25\ \mathrm{K})$ is observed. This functional form is due to electron-electron interaction effects in a three-dimensional disordered metal. Particularly, the absolute magnitude of the measured resistivity rise is consistent within ~25% with the theoretical prediction. At room temperature, the temperature coefficient of resistivity $(1/\rho)(d\rho/dT)$ decreases monotonically with increasing x or, equivalently, ρ. The essential characteristics of the variation of $(1/\rho)(d\rho/dT)$ with ρ is explained in terms of a theory that considers a competition between the quantum-mechanical effects of weak localization and the classical Boltzmann electron transport.

I. INTRODUCTION

Over the past decade or so, it has been realized that electron-electron interaction and weak-localization effects are indispensable in understanding electronic conduction in disordered systems.[1-5] Intensive work, both theoretical and experimental, has been performed in this direction, and it is now quite well accepted that these two effects are basically understood. At low temperatures (and in zero magnetic field), both effects can result in a noticeable temperature-dependent correction $\Delta\rho(T)$ to the impurity resistivity ρ_e. It has also been realized that both effects depend strongly on system dimensionality. In fact, most experimental work has been performed on two-dimensional systems, e.g., thin metal films[5] and semiconductor inversion layers.[6] Some experimental work has also been done on one-dimensional wires[7-9] and three-dimensional bulk samples.[10,11] Good agreement between theory and experiment has generally been obtained. On the other hand, what is less understood is the variation of $\Delta\rho$ with the disorder ρ_e of the sample. In two dimensions, experimental observations agree well with theoretical prediction.[5] However, it has been found experimentally in one dimension that $\Delta\rho/\rho$ varies linearly with ρ_e,[7] while the theory predicts that $\Delta\rho/\rho$ should scale with the square root of ρ_e. In three dimensions, to the best of our knowledge, this issue has not been experimentally checked.

At higher temperatures, weak-localization effects have recently been proposed to be essential in explaining the variation of the temperature coefficient of resistivity, $\mathrm{TCR} \equiv \alpha(\rho) \equiv (1/\rho)(d\rho/dT)$, with resistivity ρ in a disordered metallic system.[12] In the literature, it has been known for more than 20 years that the TCR for a material decreases *monotonically* with increasing ρ. This behavior has been observed in many crystalline disordered metallic systems and amorphous metals, and is usually referred to as the "Mooij correlation."[13] However, a quantitative, microscopic explanation of this widely observed phenomenon had not been available for a long time. In 1986, Tsuei[12] pointed out that the Mooij correlation could be quantitatively understood in terms of a competition between the quantum-mechanical effects of weak localization and the classical Boltzmann electron transport. Indeed, his theory successfully explained the correlation between α and ρ in α-particle irradiated $LuRh_4B_4$. His work suggested a simple and viable way of probing the interplay between weak-localization and inelastic-scattering processes (such as electron-phonon interaction) at a temperature where the usual magnetoresistance measurements were not useful, since at, say, room temperature an extremely high magnetic field (≈hundreds T) would be required to achieve a magnetic dephasing length shorter than the inelastic dephasing length.[5]

In this work we have carried out measurements of the electrical resistivities of $Ti_{1-x}Al_x$ (Ti-Al) alloys, with $0.00 \le x \le 0.19$, between 4 and 300 K. This system is a crystalline disordered metal. Aluminum atoms are gradually introduced into a titanium host to serve as static disorder. The degree of disorder is carefully controlled by the amount of aluminum doped. In this way, we have been able to "tune" a very wide range of the degree of disorder, e.g., the values of ρ_e of our samples can differ by as much as a factor of ~110, depending on the value of x. This enables us to study systematically the dependence on disorder of the electron-electron interaction and the weak-localization effects. Previously, three-dimensional interaction and weak-localization effects have been studied in thick granular films,[11] doped semiconductors,[14] and metallic glasses,[10] but not in crystalline metallic alloys.

This paper is organized as follows. In Sec. II, we describe our experimental method for sample fabrication and resistance measurements. In Sec. III our experimental results are presented and compared with the pertinent theories, i.e., the electron-electron interaction theory and the weak-localization theory. Section IV contains our conclusion.

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II. EXPERIMENTAL METHOD

$Ti_{1-x}Al_x$ alloys with *nominal* aluminum concentration $0.00 \le x \le 0.19$ were prepared by the standard arc-melting method. Appropriate amounts of Ti (99.995% pure) and Al (99.999% pure) were arc melted several times. The melted ingots were placed at 900 °C for order annealing for one week. Rectangular samples, typically $0.7 \times 0.7 \times 10$ mm^3, were sliced from the ingots for resistance measurements. Platinum electrodes were soldered onto the samples. dc resistances were measured using the standard four-probe technique, using a Keithley 220 current source and a Keithley 181 nanovoltmeter. The measurements were performed in a storage liquid-helium Dewar and a closed-cycle refrigerator. For the measurements at liquid-helium temperatures, the sample resistances were continuously monitored by a personal-computer-based data-acquisition system while the cryostat[15] cooled down very slowly. Care was taken to avoid any appreciable Joule heating effects.

III. RESULTS AND DISCUSSION

Figure 1 shows the resistivity rise $\Delta\rho(T)=\rho(T)-\rho(25\text{ K})$ as a function of $\sqrt{T}$ for three representative Ti-Al alloys with $\rho(25\text{ K})=143$ (open circles), 167 (open triangles), and 204 $\mu\Omega$ cm (open squares), respectively. Clearly, as the temperature is lowered below about 25 K, the resistivity rises with the square root of T. The relative rise $\Delta\rho/\rho(25\text{ K})$ in the temperature range 4–25 K is of the order of a tenth of a percent. Also, the slope of this rise, i.e., the rise between a fixed temperature range, depends strongly on the samples. For the same with a higher value of $\rho(25\text{ K})$ or ρ_e, the slope is larger. In fact, in this work we have fabricated a series of alloys with ρ_e ranging from a few to ~ 210 $\mu\Omega$ cm and carefully measured $\Delta\rho$ for each sample. This has enabled us to determine a quantitative variation of $\Delta\rho$ with ρ_e. In Fig. 2 we plot the slope of the resistivity rise, $-\Delta\rho/\sqrt{T}$, as a function of $\rho(25\text{ K})$ for various alloys. The symbols are the

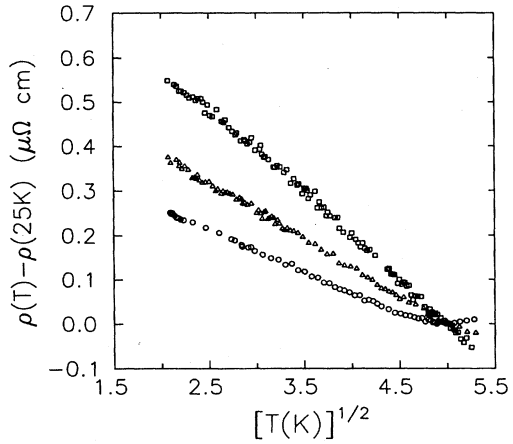

FIG. 1. Resistivity rise $\Delta\rho$ as a function of the square root of temperature T for three representative Ti-Al alloys. The alloys have $\rho(25\text{ K})$ of 143 (open circles), 167 (open triangles), and 204 $\mu\Omega$ cm (open squares), respectively.

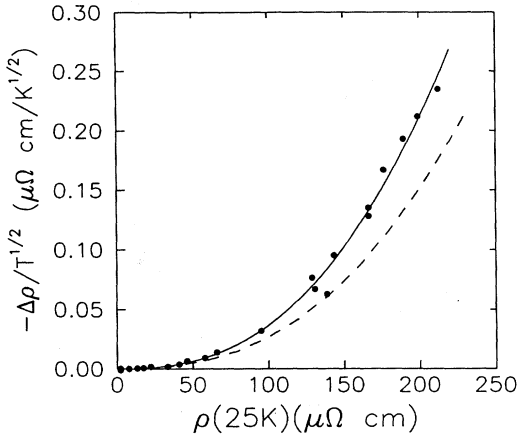

FIG. 2. The slope of the resistivity rise $-\Delta\rho/\sqrt{T}$ as a function of $\rho(25\text{ K})$ for various Ti-Al alloys. The least-squares-fitted solid curve is described by $-\Delta\rho/\sqrt{T}=6.41\times10^{-6}+3.30\times10^{-7}\rho^{2.52}$ (in $\mu\Omega\,\text{cm/K}^{1/2}$). The dashed curve is the prediction of Eq. (2) with values of $F'=0$, and $\gamma_V^*=205$ J/m^3 K^2.

experimental data and the least-squares-fitted solid curve is described by $6.41\times10^{-6}+3.30\times10^{-7}\rho^{2.52}$ (in $\mu\Omega\,\text{cm/K}^{1/2}$). Thus, we see that a dependence $\Delta\rho\sim-\rho_e^{2.5}\sqrt{T}$ is unambiguously observed.

Theoretically, the effects of electron-electron interactions in three dimensions cause a resistivity rise at low temperatures given by[3,4,10]

$$\Delta\rho_I=-\frac{1.3e^2}{4\pi^2\hbar}\left(\tfrac{4}{3}-\tfrac{3}{2}F'\right)\rho^2\left[\frac{k_BT}{2\hbar D}\right]^{1/2}, \quad (1)$$

where e is the electronic charge, k_B is the Boltzmann constant, $\hbar$ is Planck's constant divided by 2π, F' is a screening factor averaged over the Fermi surface, and D is the diffusion constant. In a metal with strong electron-phonon coupling, the value of F' could be negative.[4,10] Experimentally, the value of F' has been found to be small (i.e., close to zero) in most normal metals.[16] To facilitate a quantitative comparison with experimental result, we rewrite Eq. (1) in the following form:

$$\Delta\rho_I=-1.3\sqrt{\tfrac{3}{2}}\left(\tfrac{4}{3}-\tfrac{3}{2}F'\right)\frac{e^3}{4\pi^3\hbar^{3/2}k_B^{1/2}}\rho^{5/2}\sqrt{\gamma_V^*T}\ . \quad (2)$$

Here, γ_V^*T is the electronic specific heat per unit volume at temperature T after removal of the enhancement factor $(1+\lambda)$ due to electron-phonon interaction which is not effective in transport coefficients.[8]

To quantitatively compare our experimental result with Eq. (2), we use *independently* determined parameters for elemental Ti from the literature. Using $\gamma=3.35$ mJ/mol K^2 or $\gamma_V=315$ J/m^{3}K^2 (Ref. 17) and $\lambda\approx0.54$ (Ref. 18) to obtain $\gamma_V^*=\gamma_V/(1+\lambda)=205$ J/m^3 K^2, we then obtain readily from Eq. (2) a theoretical value $-\Delta\rho_I/\sqrt{T}=2.0(4/3-3F'/2)\times10^{-7}\rho^{2.5}$ (in $\mu\Omega\,\text{cm/K}^{1/2}$) for Ti. We assume that this value is also good for $Ti_{1-x}Al_x$, at least for small x. Since the magnitude of F' is usually small (say, ≈0) in metals, we see that this theoretical value is consistent to within $\sim25\%$ with our experimental observation. (The dashed curve in Fig.

2 represents this prediction with $F'=0$.) Note that there has been *no* adjusting parameter used. Therefore, we conclude that the electron-electron interaction theory alone can well describe the low-temperature resistance rise, both T dependence and magnitude, in Ti-Al alloys. ($F'\approx -0.2$ will bring our experimental result to be in full agreement with theory.) The present result is consistent with previous experimental observations that in metals, the electron-electron interaction effects dominate over the weak-localization effects in zero magnetic field (and at low temperatures).[16] We also note that Fig. 2 experimentally confirms the prediction that $\Delta\rho_I$ should vary with $\rho_e^{2.5}$ in a three-dimensional disordered metallic system. For comparison, we point out that, in one dimension, the theory predicts that $\Delta\rho/\rho\sim\sqrt{\rho_e}$,[3,4] while experimentally $\Delta\rho/\rho\sim\rho_e$ was observed in various metal wires with ρ_e in the range from 30 to 400 $\mu\Omega$ cm.[7] It was only when $\rho_e<30$ $\mu\Omega$ cm that the predicted $\sqrt{\rho_e}$ dependence was observed in Cu and Au wires.[7,8] It was conjectured[8] that the linear ρ_e dependence of $\Delta\rho/\rho$ at $\rho_e>30$ $\mu\Omega$ cm might signify a breakdown of the interaction theory when $k_F l_e\rightarrow 1$ (k_F is the Fermi wave number of the material and l_e is the elastic mean free path). Our present observation, however, suggests that the electron-electron interaction theory is valid up to at least $\rho_e\sim 220$ $\mu\Omega$ cm in Ti-Al. At this high value of ρ_e, $l_e\approx 3$ Å and $k_F l_e\approx 4$, compared with $k_F l_e\approx 30$ in a Cu wire with $\rho_e\approx 30$ $\mu\Omega$ cm ($k_F=1.46\times 10^8$ and 1.36×10^8 cm^{-1} for Ti and Cu, respectively[19]). Thus, our result provides strong support for the electron-electron interaction theory. Our result shows that Eq. (2) is still valid even when the magnitude of $k_F l_e$ is of order unity. Our results suggests that further work is needed to clarify the variation of $\Delta\rho$ with ρ_e in one dimension.

It has been known for 20 years that, for a given disordered metallic system, the temperature coefficient of resistivity $(1/\rho)(d\rho/dT)$ decreases with increasing resistivity ρ (the so-called Mooij correlation).[13] However, a quantitative understanding of this behavior had not been available for a long time. In 1986, Tsuei[12] pointed out that weak-localization effects were the key to this problem. Tseui observed that the Mooij correlation stemmed mostly from a competition between the quantum-mechanical effects of incipient localization and the classical Boltzmann electron transport. Thus, the total conductivity of a disordered metal is given by $\sigma(T)=\sigma_B(T)+\sigma_{\rm WL}(T)$, where σ_B is the usual Boltzmann term and $\sigma_{\rm WL}$ is a correction term due to the weak-localization effects.[12] At a finite temperature, the competition between the degradation of weak-localization effects as a result of inelastic scattering and the conventional thermal excitation of various inelastic processes determines the size and the sign of TCR. The TCR, $\alpha=-(1/\sigma)(d\sigma/dT)$, can be expressed as[12]

$$\alpha(T)=\frac{P}{T}\left\{\frac{l_e l_i}{(l_i+l_e)^2}-\frac{3\sqrt{3}l_e^{1/2}}{2(k_F l_e)^2 l_i^{1/2}}\right\}\times\left\{\frac{l_i}{l_e+l_i}+\frac{3l_e}{(k_F l_e)^2}\left[\frac{1}{L_i}-\frac{1}{l_e}\right]\right\}^{-1}, \tag{3}$$

where l_i is the inelastic mean free path, $L_i=\sqrt{l_e l_i/3}$ is the inelastic diffusion length, and P is an integer determining the temperature dependence of l_i [i.e., $l_i(T)=AT^{-P}$, where A is a constant]. Usually, $P=1$ for $T>200$ K. In deriving Eq. (3), the following (three-dimensional) result for $\sigma_{\rm WL}$ has been used:[3,12,20]

$$\sigma_{\rm WL}(T)=\frac{e^2}{\pi^2\hbar}\left\{\frac{1}{L_i(T)}-\frac{1}{l_e}\right\}. \tag{4}$$

Figure 3 plots the temperature dependence of resistivity below 300 K for various Ti-Al alloys. Clearly, as the disorder ρ is increased, Matthiesen's rule is not followed, and the TCR decreases monotonically. Figure 4 plots the variation of the TCR with ρ for our Ti-Al alloys at 300 K. In this figure, the symbols are the experimental data and the dashed curve is the prediction of Eq. (3). This dashed curve is drawn using the values of $P=1$ and $l_i=15$ Å. We see that the essential characteristics of the experimental data α as a function of disorder ρ, especially for $\rho>100$ $\mu\Omega$ cm, can quite reasonably be described by the form of Eq. (3). Thus, the present observation supports Tsuei's theory that there is a unique and monotonic correlation between α and ρ for a specific disordered metallic system. Our result also indicates that weak-localization effects are important in Ti-Al even at room temperature (where interaction effects are negligible[3,4,12]).

Consider the inferred value of l_i which is one of the most important quantities in weak-localization theory. Assume that the dominating inelastic mean free path at room temperature is due to electron-phonon scattering. Theoretically, the electronic mean free path by phonon scattering $l_{\rm ph}$ can be estimated using the expression[21]

$$l_{\rm ph}=\frac{\hbar\langle v_F^2\rangle^{1/2}}{2\pi\lambda_{\rm tr}k_B T}, \tag{5}$$

where $\langle v_F^2\rangle^{1/2}$ is the root-mean-square Fermi velocity, and $\lambda_{\rm tr}$ is the transport electron-phonon coupling con-

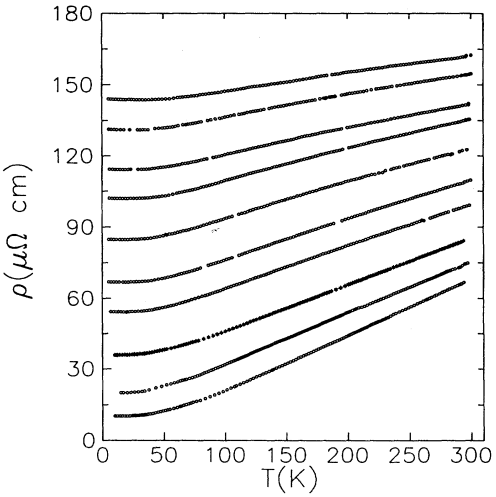

FIG. 3. Resistivity as a function of temperature T for various Ti-Al alloys with (from bottom to top) $x=0.0038$, 0.0076, 0.024, 0.040, 0.053, 0.072, 0.086, 0.102, 0.118, and 0.135, respectively.

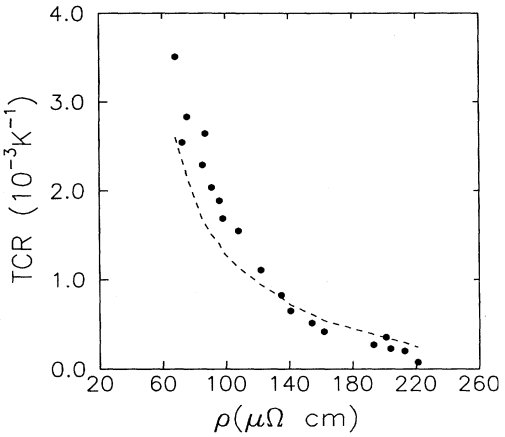

FIG. 4. Variation of the temperature coefficient of resistivity (TCR) with resistivity ρ for various Ti-Al alloys at 300 K. The dashed curve is the prediction of Eq. (3) with values of $P=1$, $k_F=1.46\times10^8$/cm, and $l_i=15$ Å.

stant. Using the values from the recent band-theory calculations for elemental Ti:[18] $\lambda_{tr}=0.54$ and $\langle v_F^2\rangle^{1/2}=0.32\times10^8$ cm/s, we obtain $l_{ph}\approx23$ Å at 300 K. This value is a factor ~1.5 higher than the experimental value (≈15 Å). Considering that a detailed comparison between theory and experiment is extremely difficult, and especially that there is *no* adjusting parameter used, we conclude that the form of Eq. (3) can already provide quite useful information on the inelastic-scattering processes. Incorporation of, say, a disorder-dependent l_i into Eq. (3) will likely further improve the general agreement between theory and experiment.

IV. CONCLUSION

We have shown experimentally that both electron-electron interaction and weak-localization effects are important in Ti-Al alloys. At liquid-helium temperatures, the electron-electron interaction effects dominate which results in a resistivity correction to the residual resistivity. Without using any adjusting parameters, the absolute magnitude of our result is within $\sim25\%$ of the theoretical prediction. At room temperature, weak-localization effects are essential in quantitatively explaining the variation of the temperature coefficient of resistivity with resistivity.

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[1] *Anderson Localization,* edited by T. Ando and H. Fukuyama (Springer, New York, 1988).
[2] *Localization, Interaction, and Transport Phenomena,* edited by B. Kramer, G. Bergmann, and Y. Bruynseraede (Springer, New York, 1985).
[3] P. A. Lee and T. V. Ramakrishnan, Rev. Mod. Phys. **57**, 287 (1985).
[4] B. L. Al'tshuler and A. G. Aronov, in *Electron-Electron Interactions in Disordered Systems,* edited by A. L. Efros and M. Pollak (Elsevier, Amsterdam, 1985).
[5] G. Bergmann, Phys. Rep. **107**, 1 (9184).
[6] See, for example, T. A. Polyanskaya and Yu. V. Shmartsev, Fiz. Tekh. Poluprovodn. **23**, 3 (1989) [Sov. Phys. Semicond. **23**, 1 (1989)].
[7] J. T. Masden and N. Giordano, Phys. Rev. B **31**, 6395 (1985); N. Girodano, in *Physics in One Dimension,* edited by J. Bernasconi and T. Schneider (Springer, New York, 1981), p. 310.
[8] A. E. White, M. Tinkham, W. J. Skocpol, and D. C. Flanders, Phys. Rev. Lett. **48**, 1752 (1982).
[9] J. B. Pieper, J. C. Price, and J. M. Martinis, Phys. Rev. B **45**, 3857 (1992); J. J. Lin and N. Giordano, *ibid.* **33**, 1519 (1986); S. Wind, M. J. Rooks, V. Chandrasekhar, and D. E. Prober, Phys. Rev. Lett. **57**, 633 (1986).
[10] See, for example, M. A. Howson and B. L. Gallagher, Phys. Rep. **170**, 265 (1988).
[11] F. Komori, S. Okuma, and S. Kobayashi, J. Phys. Soc. Jpn. **56**, 691 (1987); D. S. McLachlan, Phys. Rev. B **28**, 6821 (1983); K. C. Mui, P. Lindenfeld, and W. L. Mclean, *ibid.* **30**, 2951 (1984); T. Ohyama, M. Okamoto, and E. Otsuka, J. Phys. Soc. Jpn. **54**, 1041 (1985).
[12] C. C. Tsuei, Phys. Rev. Lett. **57**, 1943 (1986); in *Anderson Localization* (Ref. 1), p. 143.
[13] J. H. Mooij, Phys. Status Solidi A **17**, 521 (1973).
[14] D. J. Newson, M. Pepper, H. Y. Hall, and J. H. Marsh, J. Phys. C **18**, L1041 (1985); R. C. Dynes, T. H. Geballe, G. W. Hull, Jr., and J. P. Garno, Phys. Rev. B **27**, 5188 (1983).
[15] E. T. Swartz, Rev. Sci. Instrum. **57**, 2848 (1986); **58**, 881 (1987).
[16] See, for example, G. Bergmann, Phys. Rev. B **28**, 515 (1983); Solid State Commun. **49**, 775 (1984); R. S. Markiewicz and C. J. Rollins, Phys. Rev. B **29**, 735 (1984); J. J. Lin and N. Giordano, *ibid.* **35**, 545 (1987).
[17] C. Kittel, *Introduction to Solid State Physics* (Wiley, New York, 1986), p. 141.
[18] B. A. Sanborn, P. B. Allen, and D. A. Papaconstantopoulos, Phys. Rev. B **40**, 6037 (1989).
[19] *Physics Vade Mecum,* edited by H. L. Anderson (AIP, New York, 1980), p. 290.
[20] M. Kaveh and N. F. Mott, J. Phys. C **15**, L707 (1982).
[21] P. B. Allen, Phys. Rev. B **3**, 305 (1971).