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# Screening of the effect of delocalization on disordered transition metals by including the d states in the conduction band hybridization

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**Abstract.** The critical resistivity required for the temperature coefficient of electrical resistivity to change sign, for weakly disordered transition metals, may be larger than the Mooij universal boundary. A theoretical modification of the conduction electron wavefunction that involves including the d-state hybridization for the transition metals has a screening effect on the electron-phonon interaction and enhances the effect of localization on the inelastic scattering.

## 1. Introduction

Localization in disordered metals has been extensively studied for more than a decade. Mooij [1] observed a change in sign of the temperature coefficients of resistivity ( $\text{TCR} = \delta\rho/\rho \delta T$ , where  $\rho$  is the resistivity) within a critical narrow range of resistivity ( $\rho_c = 100\text{--}150 \mu\Omega \text{ cm}$ ), which afterwards served as a universal boundary. Recently, Tsuei [2] proposed that the Mooij correlation was non-universal, based on the concept of competition between the quantum mechanical effects of incipient localization [3] and the classical Boltzmann electron transport.

In transition metals or silicides, the conduction electron wavefunction can be mixed with the d-states. This hybridization implies a screening of inelastic scattering, which assists the localization by enhancing the quantum coherent interference. Consequently, the critical resistivity  $\rho_c$  necessary for the TCR to change sign, increases. This presumption has been verified by the measurement of the resistivity of ion-implanted  $\text{C}_{54}\text{-TiSi}_2$  films.

## 2. Theory

### 2.1. Conductivity concerning quantum coherent interference

The discovery of negative TCR in disordered metals invokes a move to solid-state theory, since the classical Boltzmann transport mechanism predicts an increase of resistivity with increasing temperature irrespective of whether the scattering is elastic or inelastic. The use of quantum interference successfully clears this ambiguity by proposing that during inelastic scattering, both the phase and amplitude of the electron momentum ( $k$ ) are changed, making the probability of coherent backscattering ( $k$  becomes  $-k$ )

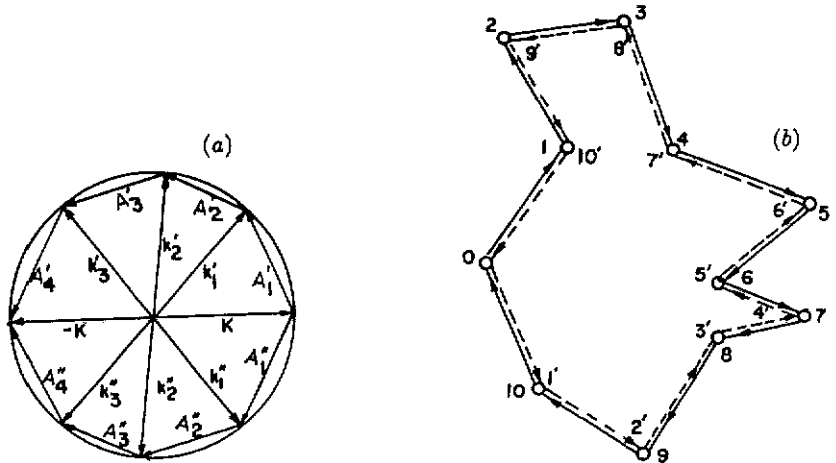


Figure 1. (a) Momentum change diagram for the elastic scattering process yielding echo in weak localization and, (b) diffusion path of the conduction electrons. The echo can occur either through the intermediate states  $K \rightarrow K_1 \rightarrow K'_2 \dots \rightarrow K_n = -K$  or  $K \rightarrow K'_1 \rightarrow K''_2 \dots \rightarrow K''_n = -K$ .

unfavourable [4]. Here, it is noticeable that the carrier motion is delocalized and the diffusion length is elongated. The origin of the negative TCR can be summarized as the following.

(i) Weak localization ( $K_f l_e \gg 1$ ); where  $K_f$  is the electronic Fermi wave-vector, and  $l_e$  is the elastic mean free path [5]. The incoherent impurity scattering is dominant only at low temperatures. As the temperature increases, the coherent acoustic-phonon scattering increases and the resistivity also increases. At high temperatures, the inelastic optical-phonon scattering intervenes and a negative TCR may reappear.

(ii) Strong localization ( $K_f l_e \ll 1$ ) [6]. If the number of impurity atoms increases, or the crystalline structure is heavily damaged, the electron wavefunction spans only a few lattices and fades exponentially. The conductivity is therefore essentially due to electron tunnelling between localized states. Negative TCR arises from phonon-assisted hopping conduction.

(iii) Kondo effect [7]. The spin of the conduction electrons will be flip-flopped by the impurity spins during collisions and the momentum phase will be randomized. The coherent length in quantum interference is reduced, resulting in a delocalization of the conduction.

(iv) Spin-orbit coupling effect [8]. To recover the original state, the orbital motion is required to have an angular revolution of  $4\pi$  via the spin-orbit interaction. However, in backward scattering, the phase of the electronic wave-vector only changes by  $2\pi$ , which implies a destructive interference by spin-orbit coupling occurring in heavy atoms.

To describe the scattering process in the momentum space, the electron wavefunction is imposed on a phase factor  $e^{ik \cdot r}$  and an amplitude  $|A|$ . After an elastic scattering time  $\tau_0$ , the wave amplitude  $|A|$  is unchanged while the wave-vector  $k$  changes to  $k'$ . As shown in figure 1 there are two ways to rotate  $k$ , one is counter clockwise (denoted by  $A'$ ) and the other is clockwise (denoted by  $A''$ ). After  $n$  scattering steps  $A'$  and  $A''$  may both equal  $-A$ , and then the process is backscattered.

Since, at the final momentum  $A' = A''$ , the total intensity is [4]

$$|A' + A''|^2 = |A'|^2 + |A''|^2 + 2A'A''^* = 4|A|^2. \tag{1}$$

If  $A'$  and  $A''$  were not coherent then the total scattering intensity of the two complementary sequences would only be  $2|A|^2$ . The reduction of scattering intensity due to incoherent quantum interference is neglected in classical Boltzmann transport. This incoherent scattering will delocalize the momentum and enhance current conduction.

If the elastic scattering time is  $\tau_0$ , the uncertainty principle implies electronic energy straggling near the Fermi surface by  $\Delta E = \pi\hbar/\tau_0$ . Converting into momentum space, we find  $\Delta E = V_f\Delta p = \hbar V_f\Delta k$  which implies that the total diffusion area of electrons in the  $k$ -space is  $2\pi k\Delta k = 2\pi k_f \cdot \pi/V_f\tau_0 = 2\pi^2 k_f/l_e$ , where  $l_e = V_f\tau_0$  is the elastic mean free path. If after time  $t = n\tau_0$ , the electrons are backscattered and create coherent interference, then the electrons will diffuse a length  $x = (nD\tau_0)^{1/2}$ . In momentum space, this corresponds to a coherent area of  $\pi q^2 = \pi(1/x)^2 = \pi/Dt$ . Therefore, the probability that backscattering occurs at time  $t$  is

$$I_{\text{coh}} = \frac{(\pi/Dt)}{(2\pi^2 k_f/l_e)} = \frac{\tau_0}{\pi k_f l_e t}. \tag{2}$$

At low temperatures, with  $\tau_0 \ll \tau_i$ , the electrons are accelerated along the applied field  $E$  during the relaxation time  $\tau_0$ . Since the inelastic scattering (with characteristic time  $\tau_i$ ) tends to randomize the scattering amplitude and phase, the probability of backscattering (echo) will occur after  $\tau_0$  (with  $n \geq 1$ ), but before  $\tau_i$ . Therefore, the effective relaxation time for forward propagating electrons is

$$\tau_0 - \int_{\tau_0}^{\tau_i} I_{\text{coh}} dt = \tau_0 \left[ 1 - \frac{1}{\pi k_f l_e} \ln(\tau_i/\tau_0) \right]. \tag{3}$$

The conductivity will be reduced by this backward diffusion, or

$$\sigma = \frac{ne^2\tau_0}{m} \left[ 1 - \frac{1}{\pi k_f l_e} \ln(\tau_i/\tau_0) \right]. \tag{4}$$

The above equation indicates that increasing the inelastic scattering (reducing  $\tau_i$ ) causes delocalization and increases the conductivity.

### 2.2. Conduction wavefunction with $d$ - $p$ state hybridization

The same result as shown in (4) can be derived from the point of view of quantum diffusion. Due to scattering, the conduction electron wavefunction in the lattice is not a single plane wave but is a wavepacket  $2|\Psi_K\rangle$  mixing all plane waves  $|k + q\rangle$  with  $q < \pi/L$ , and is written as

$$|\Psi_K\rangle = (1 + R)^{-1/2} [ |K\rangle + \sum_q a(q) |k + q\rangle ] \tag{5}$$

where

$$R = \sum_q |a(q)|^2 = \sum_q \frac{\langle k + q | V_i | k \rangle^2}{|E_{k+q}^{(0)} - E_k^{(0)}|^2} = \sum \frac{|U(q)|^2}{\left[ \frac{\hbar^2}{2m} (2kq + q^2) \right]^2} \tag{6}$$

and  $U(q)$  is the Fourier transform of the disorder potential  $\sum_j V_j(r - R_j)$  due to atomic scatterers at site  $R_j$ . If  $\Psi_K$  is substituted into the Kubo–Greenwood equation, we find

the conductivity [3]

$$\sigma = \sigma \left| \left\langle \Psi_K \left| \frac{d}{dx} \right| \Psi_K \right\rangle \right|^2 = \frac{\sigma_B}{(1+R)^2} \approx (1-2R)\sigma_B \quad \text{for } R \ll 1. \quad (7)$$

Therefore, random addition of plane waves of  $|k\rangle$  in all directions will reduce the conductivity. The quantum diffusion due to the scattering from a coulomb potential

$$v(r-R_i) \sim 1/(r-R_i)$$

leads to

$$a(q) = \frac{1}{[\hbar\pi N(E_f D)]^{1/2}} \frac{e^{i\phi(q)}}{q}$$

where  $N(E_f)$  is the density of states near  $E_f$ , and  $\phi(q)$  is a random phase relating to the state  $|k\rangle$ . In a two-dimensional system  $R \sim \sum |a(q)|^2 = [\ln(L_i/l_e)](\pi k_f l_e)^{-1}$  which reduces (7) into (4).

In silicides, the wavefunction of the conduction electrons may hybridize using the d-states of transition metals and p-states of silicon [9]. The valence charge density is then redistributed during silicide formation.

The first-order perturbation theory exploiting the orthogonalized pseudo-potential wavefunction implies the true wavefunction is

$$|\Psi_K\rangle = |k\rangle + \sum_q \frac{|k+q\rangle\langle k+q|W|k\rangle}{E_k - E_{k+q}} - \sum_{q,d} \frac{|K+q\rangle\langle K+q|\Delta|d\rangle\langle d|\Delta|k\rangle}{(E_k - E_{k+q})(E_d - E_k)} \\ - \sum_a |a\rangle\langle a|k\rangle - \sum_d |d\rangle\langle d|k\rangle + \sum_d \frac{|d\rangle\langle d|\Delta|k\rangle}{E_d - E_k} \quad (8)$$

where  $|a\rangle$  is the core state,  $\Delta$  is the pseudo-potential excluding the d-state contribution, and  $W$  is the modified pseudo-potential due to d-p hybridization and is given by

$$W|\varphi_k\rangle = V|\varphi_k\rangle + \sum_a (E - E_a)|a\rangle\langle a|\varphi_k\rangle + \sum_d [(E - E_d)|d\rangle\langle d|\varphi_k\rangle + |d\rangle\langle d|\Delta|\varphi_k\rangle \\ + \Delta|d\rangle\langle d|\varphi_k\rangle]. \quad (9)$$

The Fermi golden rule gives the implicit transition rate for elastic scattering

$$\frac{1}{\tau_0} = \frac{2\pi}{\hbar} |\langle \varphi_{k+q}^{(0)} | v_i | \varphi_k^{(0)} \rangle|^2 N_1(E_f) \quad (10)$$

where  $N_1(E_f)$  refers to the density of states for a given spin direction. Therefore, the correlation function  $R$  in (6) can be modified as

$$R = \frac{\hbar}{4\pi^2 \tau_0 N_1(E_f)} \int_{q_{\min}}^{q_{\max}} \frac{q dq}{|E_k^{(0)} - E_{k+q}^{(0)}|^2} \quad (11)$$

where the denominator is

$$E_{k+q}^{(0)} - E_k^{(0)} = \frac{\hbar^2 k q \cos \theta}{m} + \sum_d [(k+q|d\rangle\langle d|\Delta|k+q\rangle + \langle k+q|\Delta|d\rangle\langle d|k+q\rangle \\ - \langle k|d\rangle\langle d|\Delta|k\rangle - \langle k|\Delta|d\rangle\langle d|k\rangle] \quad (12)$$

and  $\theta$  is the angle between  $k$  and  $q$ . To determine the integration lower and upper limits,

we may apply the uncertainty principle

$$\Delta p \Delta \chi = q \hbar \Delta \chi = \hbar \quad \text{or} \quad q = (\Delta \chi)^{-1}. \quad (13)$$

The maximum and minimum values of  $q$  correspond to the smallest and largest distances the carrier can diffuse in various collisions. Therefore

$$q_{\min} = \frac{1}{L} \quad \text{if the crystal length } L > L_i = (D\tau_i)^{1/2}$$

$$q_{\min} = \frac{1}{L_i} \quad \text{if } L < L_i$$

and

$$q_{\max} = \frac{1}{l_c} \quad \text{for } l_c \ll L_i. \quad (14)$$

### 2.3. Example of $\text{TiSi}_2$ with C54 structure

The TEM (transmission electron microscope) electron diffraction pattern and the SEM (scanning electron microscope) cross-sectional view [10] indicate that  $\text{TiSi}_2$  are face-centred orthorhombic C54 with  $a_0 = 0.8253$  nm,  $b_0 = 0.4783$  nm and  $c_0 = 0.8540$  nm. Each unit cell contains eight Ti and sixteen Si atoms. The pseudo-potential  $\delta V$  arising from the valence charge density, may fluctuate due to the distortion of the d-state in the metal as well as from the overlap potential from neighbouring silicons, and can be written as [11]

$$\delta V(r) = a_0 + Ze^2 r^2 / 2r_c^3 \quad (15)$$

where  $r_c = (3\Omega_0/4\pi)^{1/3} \approx 2.16 \times 10^{-8}$  cm,  $\Omega_0$  is the volume of each unit cell, and  $Z = 4.817$  is the valence electron charge. The pseudo-potential for the 3d electron is

$$\Delta = \frac{Ze^2 r^2}{2r_c^3} - \int_0^\infty \frac{p_{3d}^2(r) Ze^2 r^2}{2r_c^3} dr = (5.521 \times 10^4 r^2 - 8.382 \times 10^{-12}) \text{ esu}. \quad (16)$$

Also, the matrix

$$\langle k|3d\rangle \langle 3d|\Delta|k\rangle = \frac{4\pi}{\Omega_0} 5P_2(\cos \theta) \int_0^\infty dr j_2(kr) r P_{3d} \int_0^\infty dr j_2(kr) r \Delta P_{3d}(r) \quad (17a)$$

which can be evaluated providing  $k = k_f = (9\pi Z/4)^{1/3} \times 1/r_c = 1.501 \times 10^8 \text{ cm}^{-1}$ ,  $P_2(\cos \theta) = 3/2 \cos^2 \theta - 1/2$ , and  $j_2(kr)$  is the spherical Bessel function. Therefore (12) can be calculated to yield

$$E_{k+q}^{(0)} - E_k^{(0)} = \frac{k\hbar^2}{m} q \cos \theta - 7.82 \times 10^{-21} q = \frac{C\hbar V_f}{\sqrt{2}} q \quad (17b)$$

where  $C = 0.94$ ,  $k$  is substituted by  $k_f = 1.501 \times 10^8 \text{ cm}^{-1}$ ,  $\langle \cos^2 \theta \rangle^{1/2} = 1/\sqrt{2}$ . Substituting (17) into (11), we find

$$R = \frac{\hbar}{4\pi^2 \tau_0 N_1(E_f)} \int_{1/L_i}^{1/l_c} \frac{2q dq}{C^2 \hbar^2 V_f^2 q^2} = \frac{1}{C^2 \pi k_f l_c} \ln \frac{L_i}{l_c}. \quad (18)$$

Here, we use  $N_1(E_f) = m/2\pi\hbar^2$  for the 2D system [13]. The conductivity is modified to be

$$\sigma = \sigma_B \left[ 1 - \frac{2}{C^2 \pi K_f l_c} \ln \left( \frac{L_i}{l_c} \right) \right]. \quad (19)$$

Inspecting (19), we can see that the hybridization of the conduction wavefunction from the d-states of Ti atoms has the effect of screening the electron-phonon interaction, and hence helps localizing the coherent scattering time by a factor of  $C^{-2}$ .

In Boltzmann transport, all the possible scattering mechanisms are treated the same if the scattering cross-sections have the same dependence on the wave-vector. Neglecting the inelastic electron-electron interaction, the effective mean free path of the conduction electrons follows  $1/l(T) = 1/l_c + 1/l_i$  and the conductivity

$$\sigma_B = e^2 k_f l(T) / 2\pi\hbar \approx e^2 k_f l_c (1 - l_c/l_i) / 2\pi\hbar \quad \text{if } l_i \gg l_c = \sigma_B(0) + \sigma_B(T) \quad (20)$$

where  $\sigma_B(0) = e^2 k_f l_c / 2\pi\hbar$ , and  $\sigma_B(T) = e^2 k_f l_c^2 / (2\pi\hbar l_i)$  are the temperature independent and dependent terms. The localization effects cause the conductivity to have a larger temperature dependence than  $\sigma_B(T)$  and therefore we need a correction to the Boltzmann conductivity. With  $L_i = (1/2l_c l_i)^{1/2}$  for the 2D system, (19) can be rewritten as

$$\begin{aligned} \sigma &= \sigma_B - \frac{e^2}{2C^2\pi^2\hbar^2} (1 - l_c/l_i) (\ln l_i - \ln 2l_c) \\ &= \sigma_B + \sigma_L = \sigma_B(0) + \sigma_B(T) + \sigma_L(0) + \sigma_L(T) \end{aligned} \quad (21)$$

where

$$\sigma_L(0) = \frac{e^2}{2C^2\pi^2\hbar} \ln(2l_c) \quad \text{and} \quad \sigma_L(T) = -\frac{e^2}{2C^2\pi^2\hbar} \left[ \ln l_i + \frac{l_c}{l_i} \ln(2l_c) \right].$$

The temperature dependent terms  $\sigma_B(T)$  represent the delocalization of the zero-temperature, quantum corrected conductivity  $\sigma(0)$ , by inelastic scattering, and  $\sigma_L(T)$  describes the degradation of the coherent quantum interference by inelastic scattering and various thermally excited inelastic scatterings. For metals with  $l_i \gg l_c$ , we can assume  $\sigma \approx \sigma_B(0)$  to a first order approximation, and it is straightforward then to derive

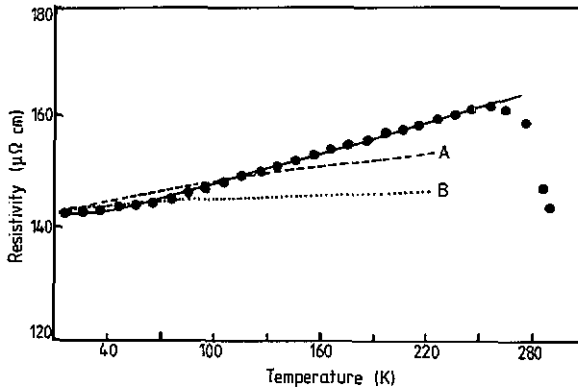
$$a = \frac{1}{\sigma(T)} \frac{\delta\sigma(T)}{\delta T} \approx -\frac{e^2}{2\pi^2\hbar} \left[ -\frac{\pi k_f l_c^2}{l_i^2(T)} + \frac{1}{C^2} \frac{1}{l_i(T)} \right] \frac{dl_i(T)}{dT}. \quad (22)$$

Since  $dl_i(T)/dT < 0$ , and assuming  $C = 1$  and  $K_f l_c \approx 1$ , it can be assumed that  $a(T)$  will be always positive (non-metallic). However, above some temperature  $T_M$  when  $k_f l_c$  increases and becomes much greater than 1, the sign of  $a(T)$  may change and become negative (metallic) [3]. Since  $l_c$  is inversely proportional to the impurity concentration, the critical temperature  $T_M$  decreases as the implanted ion dose decreases, which is what we have proved in this experiment. The  $T_M$  which usually occurs below 10 K for a weakly disordered system is a cross-over from a weak temperature dependence on  $\sigma$  (dominated by the correlation effect) to a strong one (dominated by localization). The same experimental result has been demonstrated for  $\text{Ge}_{1-x}\text{Au}_x$  alloy [14].

There is another cross-over temperature which occurs at much lower temperatures because the conduction is dominated by inelastic electron-electron interaction rather than the inelastic electron-phonon interaction. With the inclusion of d-p hybridization (i.e.  $C \neq 0$ ) the critical condition for  $a$  begins to change sign (i.e.  $a = 0$ ), which occurs when  $l_i = \pi C^2 k_f^2 l_c$ . Then, the critical resistivity for the 2D system is

$$\rho_c = \rho_B(0) \left[ 1 - \frac{1}{C^2\pi k_f l_c} - \frac{1}{C^2\pi k_f l_c} \ln \frac{C^2\pi k_f l_0}{2} \right]^{-1}.$$

Because of the hybridization, the factor  $C (< 1)$  makes  $\rho_c$  increase. This is a possible reason for the non-universality of the Mooij correlation which limits  $\rho_c$  to be within 100–150  $\mu\Omega$  cm.



**Figure 2.** The  $\rho$ - $T$  measurement for the  $\text{As}^+$  implanted  $\text{Ti}_2\text{Si}$  of 125 nm. The solid line is the theoretical fitting by (21) with parameters given by  $l_e = 0.728$  nm,  $l_i = 1.25 \times 10^3$  nm/ $T$  for ion energies of 100 keV at a dose of  $4 \times 10^{15}$   $\text{cm}^{-2}$ . Curve fittings for A and B are given by  $\rho = 138.8 + 0.97T^{1/2}$  and  $\rho = 138.8 + 1.4 \ln T$   $\mu\Omega$  cm, respectively.

### 3. Experimental technique

To explore the localization in transition metal silicides, we have measured the temperature dependent resistivity of  $\text{As}^+$  implanted C54- $\text{TiSi}_2$  films. Titanium films with thicknesses of 100 ~ 125 nm were electron gun deposited on (111), 1 ~ 10  $\Omega$  cm silicon substrates under a pressure of  $1 \times 10^{-6}$  torr at a deposition rate of 0.2 nm  $\text{min}^{-1}$ . The samples were isothermally annealed at 800  $^\circ\text{C}$  for 30 min in an oil-free high vacuum furnace. The TEM electron diffraction patterns indicate that [10, 15] two preferred orientations of [102] and [101]  $\text{TiSi}_2$  can be epitaxially grown on (111) Si at local areas with a grain size of about 4  $\mu\text{m}$ . The van der Pauw method was used to perform the resistivity measurement, since the  $\text{TiSi}_2$  and Si interface can form a Schottky barrier contact, which can effectively prevent the current flowing from the under-layered Si substrates.

Figure 2 is the resistivity versus temperature ( $\rho$ - $T$ ) measurement for the  $\text{As}^+$  implanted  $\text{TiSi}_2$  at a dose of  $5 \times 10^{15}$   $\text{cm}^{-2}$ . A theoretical fitting with (21) implies that the simulated elastic and inelastic mean free paths (e.g.  $l_e = 0.728$  nm and  $l_i = 1.25 \times 10^3$  nm/ $T$ ) satisfactorily agree with the reported values for disordered metallic conductors [16]. For this short mean free path, a simulation based on a 3D weak localization mechanism may yield a better agreement with the experimental work. Equation (21) can be approximated by  $\rho(T) \sim \rho_0 + \rho' \ln T$  in the low temperature regions under the weak localization conditions which are relevant to the experimental data below  $T < 90$  K. We have also plotted the  $\rho \alpha T^{1/2}$  dependences which predict a better agreement with the experimental data at  $90 \text{ K} < T < 160 \text{ K}$  implying that the electron-phonon interaction prevails in the presence of disorder at high temperatures. As shown in figure 2, the slope of the TCR appears to be negative near 280 K. This negative  $d\rho/dT$  at high temperatures may be ascribed to power law 'localization effects' [3]. The critical resistivity  $\rho_c$  as shown in the implanted  $\text{TiSi}_2$  exceeds the Mooij universal boundary.

### 4. Conclusions

In this work, we have first calculated the effect of d-state hybridization of the conduction wavefunction on the resistivity of weakly localized  $\text{TiSi}_2$  films. The probability of con-



duction electrons being scattered from  $k$  to  $k + q$  states by inelastic scattering is reduced by a factor of  $C^2 \approx 0.88$  due to the screening of the electron-phonon interaction by this hybridization. It should be noted that the critical resistivity  $\rho_c$  increases until the temperature coefficient of resistivity changes signs.

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