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To cite this article: Mi-Ae Park *et al* 2001 *Supercond. Sci. Technol.* **14** L31

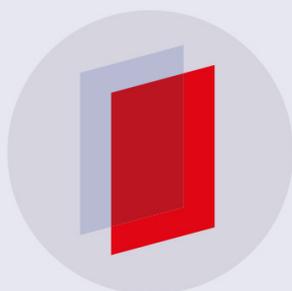
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RAPID COMMUNICATION

A new method of probing the phonon mechanism in superconductors, including MgB₂

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Received 1 May 2001

Abstract

Weak localization has a strong influence on both the normal and superconducting properties of metals. In particular, since weak localization leads to the decoupling of electrons and phonons, the temperature dependence of resistance (i.e. λ_{lr}) decreases with increasing disorder, as manifested by Mooij's empirical rule. In addition, Testardi's universal correlation of T_c (i.e. λ) and the resistance ratio (i.e. λ_{lr}) follows. This understanding provides a new means to probe the phonon mechanism in superconductors, including MgB₂. The merits of this method are its applicability to any superconductor and its reliability because the McMillan's electron–phonon coupling constant λ and λ_{lr} change in a broad range, from finite values to zero, due to weak localization. Karkin *et al*'s preliminary data of irradiated MgB₂ show the Testardi correlation, indicating that the dominant pairing mechanism in MgB₂ is a phonon-mediated interaction.

1. Introduction

The recent discovery of superconductivity in MgB₂ at 39 K by Akimitsu and co-workers [1] has renewed our interest in superconductivity. There are already many experimental and theoretical investigations [2–15]. For instance, the isotope effect coefficient of Boron is measured to be about 0.26–0.3 [2, 3] and temperature-dependent resistivity measurements indicate that MgB₂ is a highly conducting material with $\lambda_{lr} \leq 0.6$ and room-temperature resistance ratio (RRR) of $RRR = 25.3$ [4]. The electron–phonon coupling constant λ is estimated to be about 0.6–0.7 based on low-temperature specific heat measurements [5, 6], whereas the phonon density-of-states measurements suggest $\lambda \sim 0.9$ [7]. Tunnelling measurements of the energy gap show the BCS form with some variations of the maximum gap values [8–10]. On the other hand, electronic and phononic structures have been computed by numerical methods [11–15]. It has been found that the (possibly anharmonic [15]) Boron bond stretching modes are strongly coupled to the $p_{x,y}$ electronic bands. The McMillan's electron–phonon coupling constant λ is calculated

to be about $\lambda \sim 0.7$ – 0.9 [11–15]. These investigations seem to be consistent with the BCS phonon-mediated superconducting behaviour.

However, there is no definite experimental evidence as yet. It is also not clear whether MgB₂ is an intermediate-coupling or strong-coupling superconductor [11–15], even though it is plausible that the high-frequency boron phonon modes may lead to strong electron–phonon coupling and the high T_c [11–15]. From a fundamental point of view it remains to be clarified whether the conventional strong-coupling theory can explain the high $T_c = 39$ K. In other words, what is the maximum T_c which can be produced by the phonon-mediated interaction [16, 17]? In this context it is clear that MgB₂ will lead us to refine our understanding of superconductivity.

There are basically two methods to probe the phonon mechanism directly. One is the isotope effect measurement [2, 3] and the other is the tunnelling measurement of the electron–phonon spectral density [18]. Although the existence of the boron isotope effect on T_c is strong evidence for the importance of the phonon mechanism [2, 3], the observed reduced isotope effect requires more investigation on the

pairing mechanism [3]. Unfortunately, the tunnelling data are not available at this moment.

In this rapid communication we introduce a new method of probing the phonon mechanism in superconductors, including MgB_2 . This method is based on the correlation of the McMillan's electron-phonon coupling constant λ in superconductivity and λ_{ir} in the phonon-limited resistivity of the normal transport phenomena [19,20]. In most s-orbital metals λ and λ_{ir} are almost the same in magnitude [19,20]. Testardi *et al* [21,22] found that disorder significantly decreases both quantities and leads to the universal correlation of T_c and the resistance ratio, which may be called the Testardi correlation. In fact, this experimental result is a manifestation of the weak localization effect on the electron-phonon interaction [23]. More precisely, weak localization leads to the decoupling of electrons and phonons and thereby gives rise to the Testardi correlation. Therefore, if MgB_2 shows the Testardi correlation we may say that the dominant pairing mechanism of MgB_2 is the phonon-mediated interaction. This correlation has already been confirmed in A-15 compounds [21,22] and ternary superconductors [24]. Another experimental manifestation of the weak localization effect on the electron-phonon interaction is the Mooij rule [25]. This rule states that as the system becomes disordered the temperature dependence of the resistivity decreases, that is the coupling between electrons and phonons weakens. This provides another test for the importance of the phonon mechanism in superconductors.

The main advantages of this new method are its wide applicability to any superconductor and its reliability, because the McMillan's coupling constant λ and λ_{ir} can be varied from finite values to zero. Since the temperature dependence of the resistivity at room temperature is dominated by the electron-phonon interaction, the decrease of λ_{ir} clearly signals the reduction of the electron-phonon interaction and thereby probes the importance of the phonon mechanism in superconductors. This method may also provide crucial information on the pairing mechanism in exotic superconductors, such as fullerene superconductors, organic superconductors, heavy fermion superconductors, high- T_c cuprates, and Sr_2RuO_4 .

2. Manifestations of weak localization effect on the electron-phonon interaction

In this section we point out that Testardi's correlation of T_c and the resistance ratio and the Mooij rule are caused by the weak localization of electrons in disordered systems.

2.1. Testardi's correlation of T_c and the resistance ratio

In the 1970s Testardi *et al* [21,22] found the universal correlation of T_c and the resistance ratio in A-15 compounds, such as Nb-Ge, V_3Si , and V_3Ge . Since T_c and the resistance ratio are determined by McMillan's electron-phonon coupling constant λ and λ_{ir} respectively, this means the correlation between λ and λ_{ir} . The McMillan's coupling constant λ is defined by [16]

$$\lambda = 2 \int \frac{\alpha^2(\omega)F(\omega)}{\omega} d\omega = N_0 \frac{\langle I^2 \rangle}{M \langle \omega^2 \rangle} \quad (1)$$

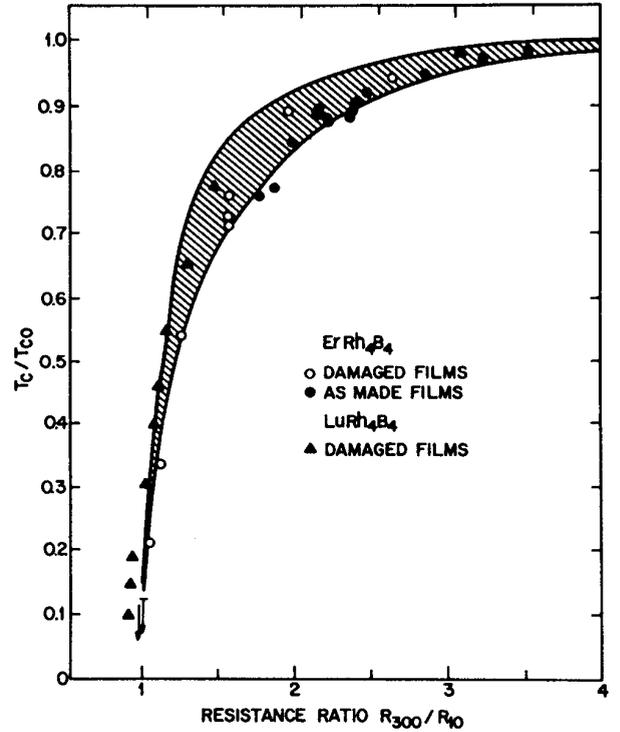


Figure 1. T_c/T_{c0} against the resistance ratio. The shaded region represents the T_c -resistance-ratio correlation band for A-15 compounds from Testardi *et al* [21,22]. The circles are for ErRh_4B_4 and the triangles for LuRh_4B_4 ; the data are from Dynes *et al* [24].

where $F(\omega)$ is the phonon density of states and M is the ionic mass. $\langle I^2 \rangle$ and $\langle \omega^2 \rangle$ are the average over the Fermi surface of the square of the electronic matrix element and the phonon frequency. The RRR is given as

$$\frac{\rho(300\text{ K})}{\rho_0} = \frac{\rho_0 + \rho_{ph}(300\text{ K})}{\rho_0} \quad (2)$$

where ρ_0 and ρ_{ph} denote the residual resistivity and the phonon-limited resistivity. The phonon-limited resistivity ρ_{ph} at high temperature is defined by

$$\rho_{ph}(T) = \frac{4\pi mk_B T}{ne^2 \hbar} \int \frac{\alpha_{ir}^2 F(\omega)}{\omega} d\omega = \frac{2\pi mk_B T}{ne^2 \hbar} \lambda_{ir}. \quad (3)$$

Here α_{ir} includes an average of a geometrical factor $1 - \cos \theta_{\vec{k}\vec{k}'}$. Inserting (3) into (2) we obtain

$$\frac{\rho(300\text{ K})}{\rho_0} = 1 + \frac{2\pi \tau k_B \times 300\text{ K}}{\hbar} \lambda_{ir}. \quad (4)$$

Figure 1 shows the correlation of T_c and the resistance ratio for A-15 compounds and ternary superconductors. Data are from Testardi *et al* [21,22] and Dynes *et al* [23]. The shaded region denotes the correlation band for A-15 compounds. It is clear that as the system becomes disordered by radiation damage or substitutional alloying, both λ and λ_{ir} decrease and thereby reduce T_c and the resistance ratio. This behaviour exemplifies strong correlations between the physical properties in normal and superconducting states.

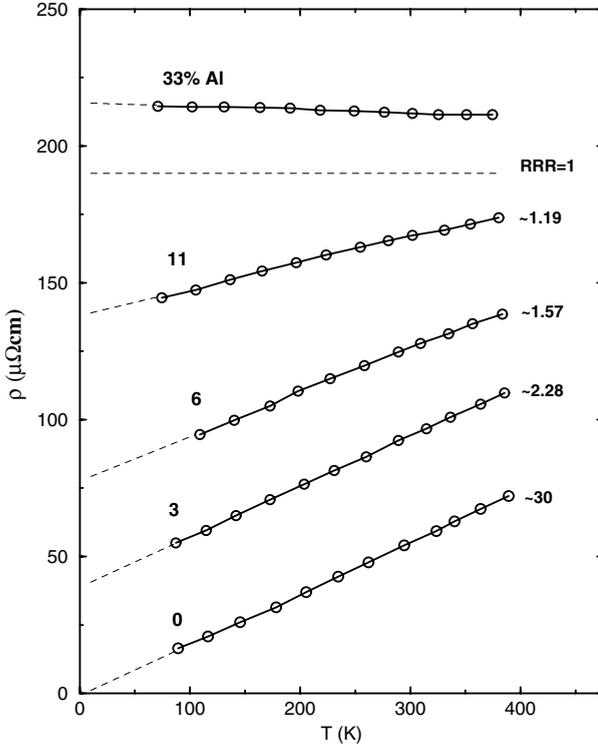


Figure 2. Resistivity against temperature for Ti and TiAl alloys containing 0, 3, 6, 11, and 33% Al; the data are from Mooij [25]. The dashed lines were used to estimate the RRR .

2.2. The Mooij rule

Mooij [25] pointed out that the size and sign of the temperature coefficient of resistivity (TCR) in many disordered systems correlate with its residual resistivity ρ_0 as follows:

$$\begin{aligned} d\rho/dT > 0 & \quad \text{if } \rho_0 < \rho_M \\ d\rho/dT < 0 & \quad \text{if } \rho_0 > \rho_M. \end{aligned} \quad (5)$$

Thus, the TCR changes the sign when ρ_0 reaches the Mooij resistivity $\rho_M \sim 150 \mu\Omega \text{ cm}$. In other words, as the system becomes disordered, the TCR decreases.

Now we show that the Testardi correlation is equivalent to the Mooij rule. Since the resistivity at temperature T is given by

$$\rho(T) = \rho_0 + \rho_{ph}(T) \quad (6)$$

the TCR is determined mainly by ρ_{ph} , i.e.

$$d\rho/dT = d\rho_{ph}/dT \cong \frac{2\pi mk_B}{ne^2\hbar} \lambda_{tr}. \quad (7)$$

Note that since the TCR is controlled by λ_{tr} , the decrease of the TCR due to disorder means the reduction of λ_{tr} , which is the essence of the Testardi correlation. Therefore, both the Testardi correlation and the Mooij rule are manifestations of the weak localization correction to the electron–phonon interaction; that is to λ and λ_{tr} .

Figure 2 shows the resistivity as a function of temperature for pure Ti and TiAl alloys containing 3, 6, 11, and 33% Al; the data are from Mooij [25]. The TCR decreases as the residual resistivity increases due to disorder; note that the RRR

decreases accordingly. The dashed lines are our conjectured data points to estimate RRR . Rough estimated values are ~ 30 , ~ 2.28 , ~ 1.57 , and ~ 1.19 for Al concentrations of 0, 3, 6, and 11%. When the RRR is about one, equation (3) tells us that λ_{tr} (and λ) is zero. If the system shows superconductivity, T_c should drop to zero at this point, which is in agreement with the Testardi correlation of T_c and the resistance ratio.

2.3. Weak localization correction to McMillan's coupling constant λ and λ_{tr}

We briefly review the derivation by Park and Kim [23]. Since the equivalent electron–electron potential in the electron–phonon problem is determined by the phonon Green's function, $D(x - x')$, the Fröhlich interaction at a finite temperature for an Einstein model is given by

$$\begin{aligned} V_{nn'}(\omega, \omega') &= \frac{I_0^2}{M\omega_D^2} \int \int d\mathbf{r} d\mathbf{r}' \psi_n^*(\mathbf{r}) \psi_n^*(\mathbf{r}') \\ &\quad \times D(\mathbf{r} - \mathbf{r}', \omega - \omega') \psi_n(\mathbf{r}') \psi_n(\mathbf{r}) \\ &= \frac{I_0^2}{M\omega_D^2} \int |\psi_n(\mathbf{r})|^2 |\psi_n(\mathbf{r}')|^2 d\mathbf{r} \frac{\omega_D^2}{\omega_D^2 + (\omega - \omega')^2} \\ &= V_{nn'} \frac{\omega_D^2}{\omega_D^2 + (\omega - \omega')^2} \end{aligned} \quad (8)$$

where

$$\begin{aligned} D(\mathbf{r} - \mathbf{r}', \omega - \omega') &= \sum_{\mathbf{q}} \frac{\omega_D^2}{(\omega - \omega')^2 + \omega_D^2} e^{i\mathbf{q}\cdot(\mathbf{r}-\mathbf{r}')} \\ &= \frac{\omega_D^2}{(\omega - \omega')^2 + \omega_D^2} \delta(\mathbf{r} - \mathbf{r}'). \end{aligned} \quad (9)$$

Here ω means the Matsubara frequency and ψ_n denotes the scattered state. I_0 is the electronic matrix element for the plane wave states. Accordingly, the McMillan's electron–phonon interaction coupling constant λ is given by

$$\lambda = N_0 \langle V_{nn'}(0, 0) \rangle = N_0 \frac{I_0^2}{M\omega_D^2} \left\langle \int |\psi_n(\mathbf{r})|^2 |\psi_n(\mathbf{r}')|^2 d\mathbf{r} \right\rangle. \quad (10)$$

This expression shows that the McMillan's coupling constant is basically determined by the short time density correlation function [23, 26], since the phonon-mediated interaction is retarded for $t_{ret} \sim 1/\omega_D$:

$$\lambda = N_0 \frac{I_0^2}{M\omega_D^2} \left[1 - \frac{3}{(k_F \ell)^2} \left(1 - \frac{\ell}{L} \right) \right]. \quad (11)$$

Here ℓ and L denote the elastic mean free path and the inelastic diffusion length, respectively. Subsequently, one finds

$$\lambda_{tr} = 2 \int \alpha_{tr}^2(\omega) F(\omega) \omega d\omega \cong N_0 \frac{I_0^2}{M\omega_D^2} \left[1 - \frac{3}{(k_F \ell)^2} \right]. \quad (12)$$

We have used the fact that L is effectively infinite at $T = 0$. It is worth noting that the weak localization correction term is the same as that of the conductivity.

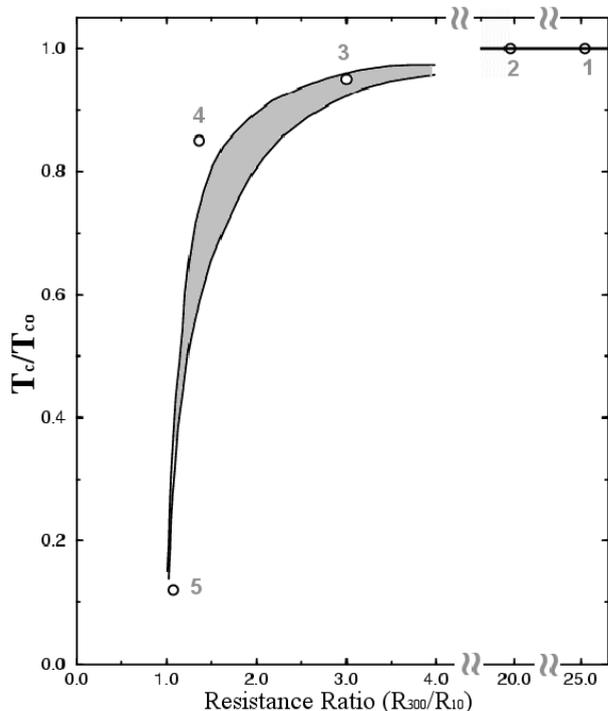


Figure 3. T_c -resistance-ratio correlation band for A-15 compounds with the data of MgB_2 superimposed; the data are from Canfield *et al* [4] (sample 1), Finnemore *et al* [30] (sample 2), Jung *et al* [31] (sample 3), and Karkin *et al* [29] (samples 4 and 5).

3. Using weak localization to probe the phonon mechanism in magnesium diboride

Since weak localization of the electrons occurs for the mean free path of the order of 10 \AA [23], heavy doses of radiation or high concentrations of impurities are required to see the effect of weak localization. At the same time the disordered samples should be macroscopically homogeneous. Consequently, recent impurity doping experiments [27, 28] in MgB_2 were not successful in seeing this effect, while Karkin *et al* [29] observed the decrease of T_c (onset temperature) from 39 to 5 K by neutron irradiation. This behaviour is very similar to the T_c decrease of A-15 compounds and ternary superconductors due to radiation damage, which has been explained by the weak localization effect [23]. Now we check whether or not MgB_2 data satisfy the Testardi correlation and the Mooij rule.

Figure 3 shows T_c/T_{c0} against the RRR for MgB_2 (points/samples 1,3,4 and 5) and Mg^{10}B_2 (point/sample 2). The points/samples lie within the correlation band of A-15 compounds, although sample 4 shows some deviation, presumably due to intergrain resistivities [29]. Note that this correlation is universal for phonon-mediated superconductors. For MgB_2 T_{c0} was assumed to be 39.4 K, corresponding to the T_c of MgB_2 wire [4], whereas the T_{c0} of Mg^{10}B_2 was chosen to be 40.2 K [30]. The data are from Canfield *et al* [4] (sample 1), Finnemore *et al* [30] (sample 2), and Jung *et al* [31], and Karkin *et al* [29] (samples 4 and 5). Since samples 4 and 5 show a broad transition width ($\sim 8 \text{ K}$), a criterion of a 50% drop of the resistivity was used to determine T_c for samples 4 and 5. The measured values of RRR are: sample 1, 25.3 [4]; sample 2, 19.7 [30]; sample 3, 3 [31]; sample 4, ~ 1.30 [29]; and sample 5,

~ 1.076 [29]. For sample 3 the disorder of the sample may be due to the high-pressure sintering at high temperature [31]. Overall, the Mooij rule is also satisfied, approximately.

It seems that the preliminary data support the phonon mechanism in MgB_2 . It is highly desirable to perform irradiation experiments using a better quality sample to confirm this result. The Mooij rule can also be confirmed separately, although the Testardi correlation would invariably lead to the Mooij rule.

4. Conclusions

We introduce a new method of probing the phonon mechanism in superconductors, including MgB_2 . Weak localization decreases both λ in superconductivity and λ_{tr} in the phonon-limited resistivity at the same rate, as manifested by the Testardi correlation of the T_c and the resistance ratio. Above T_c the Mooij rule follows accordingly. Preliminary data of MgB_2 show the Testardi correlation and thereby support the phonon mechanism in this newly discovered superconductor. More thorough experimental investigations are required using better samples to clarify the details of the pairing mechanism.

Acknowledgments

We are grateful to Professors Ceyhun Bulutay, B Tanatar, and A E Karkin for discussions. MP thanks NSF-EPSCOR (grant No. EPS9874782) for financial support.

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