

## Fully Band-Resolved Scattering Rate in MgB<sub>2</sub> Revealed by the Nonlinear Hall Effect and Magnetoresistance Measurements

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We have measured the normal state temperature dependence of the Hall effect and magnetoresistance in epitaxial MgB<sub>2</sub> thin films with variable disorders characterized by the residual resistance ratio  $RRR$  ranging from 4.0 to 33.3. A strong nonlinearity of the Hall effect and magnetoresistance have been found in clean samples, and they decrease gradually with the increase of disorders or temperature. By fitting the data to the theoretical model based on the Boltzmann equation and *ab initio* calculations for a four-band system, for the first time, we derived the scattering rates of these four bands at different temperatures and magnitude of disorders. Our method provides a unique way to derive these important parameters in multiband systems.

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The multiband character of MgB<sub>2</sub> [1] dominates its properties in both the superconducting [2–6] and normal states [7–10]. In MgB<sub>2</sub>, there are two holelike quasi-two-dimensional  $\sigma$  bands (bonding  $\sigma_1$  and antibonding  $\sigma_2$ ), an “electronlike” antibonding ( $\pi_1$ ), and a “holelike” bonding ( $\pi_2$ ) three-dimensional  $\pi$  band [11,12]. The electron scattering rates in each band and between different bands are the most critical parameters dictating all aspects of the properties of MgB<sub>2</sub> [8–10,13]. The temperature dependence of the electron scattering rates arises from electron-phonon ( $e$ -ph) coupling, and the strong  $e$ -ph coupling between the  $E_{2g}$  phonon mode and the  $\sigma$  bands are responsible for the high  $T_c$  in MgB<sub>2</sub> [14]. Consequently, measuring the intraband and interband scattering rates in MgB<sub>2</sub>, with a goal to further manipulate them in order to reveal new physics and achieve desirable properties, has been central to many research studies. The properties used to extract the band-resolved scattering rates include electrical resistivity [10], magnetoresistance (MR) [8,9,15,16], far-infrared spectroscopy [17], and upper critical field  $H_{c2}$  [13]. For example, Monni *et al.* was able to derive the temperature-dependent relaxation times,  $\tau_\sigma$  and  $\tau_\pi$ , for the generalized  $\sigma$  and  $\pi$  bands, respectively, from the MR measurement and *ab initio* calculations [15]. However, although the two  $\sigma$  bands (or the two  $\pi$  bands) have similar properties, they are distinct from each other. For example, the two  $\pi$  bands have different types of carriers. Without the knowledge of scattering rates in all the *four* different bands, the understanding of the multiband nature of MgB<sub>2</sub> is incomplete. To our best knowledge, information from MR alone is not sufficient to derive the scattering rates in four different bands. In this Letter, we report results of

strong nonlinear Hall effect (NLHE) and large MR in pure epitaxial MgB<sub>2</sub> films. This made it possible to derive the scattering rates and their dependencies on temperature and disorder in each of the four bands, a significant advancement of the knowledge concerning the multiband character of MgB<sub>2</sub>.

The MgB<sub>2</sub> films used in this work were grown by the hybrid physical-chemical vapor deposition (HPCVD) technique [18] on (0001) 6H-SiC substrates. They were epitaxial with a  $c$ -axis orientation, and the thickness was between 100 and 200 nm. Films with different magnitudes of disorder, thus different residual resistivity, were grown at slightly different temperatures, pressures, and growth rates. Nevertheless, all the films had similar  $T_c$  around 40 K and narrow x-ray diffraction rocking curves. The longitudinal and the transverse resistivity were measured with sweeping magnetic field at a fixed temperature or sweeping temperature at a fixed field. All the measurements were performed with magnetic field applied perpendicular to the  $ab$  plane of the film. For the seven films investigated here, the residual resistance ratios  $RRR \equiv \rho(300\text{ K})/\rho(41\text{ K})$  are 33.3, 24.5, 20.9, 14.4, 6.88, 6.4, and 4.0, and we mark them as  $RRR_{33.3}$ ,  $RRR_{24.5}$ ,  $RRR_{20.9}$ ,  $RRR_{14.4}$ ,  $RRR_{9.9}$ ,  $RRR_{6.4}$ , and  $RRR_{4.0}$ , respectively. Their corresponding residual resistivities  $\rho_n$  are 0.293, 0.347, 0.411, 0.740, 1.32, 2.55, and 4.66  $\mu\Omega\text{ cm}$ , respectively.

In Fig. 1, we show the field dependence of  $\Delta\rho_{xx}(B)/\rho_{xx}(0)$  and  $R_H(B)/R_H(0)$  for all the seven samples at  $T = 41\text{ K}$ . Here,  $R_H(B) = \rho_{xy}/B$  is the Hall coefficient at the magnetic field  $B$ ;  $\rho_{xx}$ , and  $\rho_{xy}$  are the longitudinal and transverse resistivity, and  $\Delta\rho_{xx}(B) = \rho_{xx}(B) - \rho_{xx}(0)$ . The dashed lines show the cases for zero MR and linear

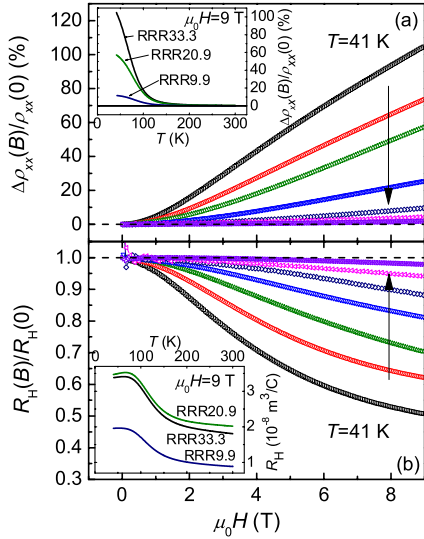


FIG. 1 (color online). Field dependence of  $\Delta\rho_{xx}(B)/\rho_{xx}(0)$  (a), and  $R_H(B)/R_H(0)$  (b), at  $T = 41$  K for seven samples with  $RRR$  values from 33.3 to 4.0 (direction of the arrows). Insets in (a) and (b) show the temperature dependence of the normalized MR and  $R_H$  at 9 T for sample  $RRR33.3$ ,  $RRR20.9$ , and  $RRR9.9$ . One can see that the MR and  $R_H$  decrease rapidly when the temperature is increased.

Hall effect, as one can expect in single band metals with the spherical or the columned Fermi surface. MR and NLHE are observed in all the samples, but the MR is larger and the NLHE is stronger in cleaner samples. In sample  $RRR33.3$  at 9 T and 41 K, a large MR of more than 100% is observed, and the Hall coefficient decreases to about half of the zero-field value. The magnitude of MR is similar to an earlier report in a clean  $MgB_2$  film due to the multiband effect [9]. For the dirty samples with low  $RRR$ , because the interband scattering is very strong, the multiband natures, such as the MR and the NLHE, are weakened, and the Kohler's rule is satisfied [19], which is not the case in clean  $MgB_2$  films [9]. The insets in Figs. 1(a) and 1(b) present the temperature dependence of the normalized MR and NLHE at 9 T of the films  $RRR33.3$ ,  $RRR20.9$ , and  $RRR9.9$ . Both MR and the NLHE decrease rapidly with increasing  $T$ . In a single band metal with anisotropic Fermi surface,  $R_H$  should be very weakly  $T$  dependent. However, as shown in the inset of Fig. 1(b), the Hall coefficient has a strong  $T$  dependence, especially for the clean samples. It should be noted that the value of  $R_H$  is about an order of magnitude larger than those reported by Eltsev *et al.* in single crystals [20], which is not necessarily an indication of a smaller density of charge carriers in this clean film. As discussed below, due to the existence of multiple bands, the Hall coefficient can no longer be written simply as  $1/ne$  as in single band materials.

The large MR and NLHE originate directly from the multiband character of  $MgB_2$ , from which we can extract information on the electron scattering rates in the different

bands. To do this, we first need to determine the contribution of each band to the (Hall) conductivity at the given electron scattering rate. The complex band structure of  $MgB_2$  renders the simple formulae presented in textbooks invalid. We thus employ the semiclassical Boltzmann theory with the relaxation time approximation [21]. In this approximation, the electron state is assumed to have a finite lifetime, which is induced by all possible electron scattering processes, including the intraband and interband scatterings. The conductivity tensor  $\sigma$  for the  $n$ th band reads:

$$\sigma^{(n)} = e^2 \int \frac{d\mathbf{k}}{4\pi^3} \tau_n \mathbf{v}_n(\mathbf{k}) \bar{\mathbf{v}}_n(\mathbf{k}) \left( -\frac{\partial f}{\partial \varepsilon} \right)_{\varepsilon=\varepsilon_n(\mathbf{k})}, \quad (1)$$

where  $\mathbf{v}_n(\mathbf{k}) = \partial \varepsilon_n / \partial \hbar \mathbf{k}$  is the group velocity at the wave vector  $\mathbf{k}$ . Under a magnetic field, the wave vector of electron evolves by  $\hbar \dot{\mathbf{k}}_n = -(e/c) \mathbf{v}_n(\mathbf{k}) \times \mathbf{B}$ ; and  $\bar{\mathbf{v}}_n(\mathbf{k}) = \int_{-\infty}^0 dt e^{t/\tau_n} \mathbf{v}_n(\mathbf{k}) / \tau_n$ , is a weighted average of the velocity over the past history of the electron orbit passing through  $\mathbf{k}$ ;  $f$  is the Fermi distribution function. Here, we have assumed that  $\tau_n$  is independent on the wave vector  $\mathbf{k}$ . The band structure necessary for the evaluation is determined from the *ab initio* electronic structure calculations. Here, we employed the ultrasoft pseudopotential plane-wave method [22] with generalized-gradient approximation [23] for the exchange and correlation potential. For the case of  $B = 0$ , we obtain the electronic density of state at the Fermi level and plasma frequency  $\omega_{xx}$  and  $\omega_{zz}$  for each band in very good agreement with those given by Liu *et al.* [10]. For  $B \neq 0$ ,  $\bar{\mathbf{v}}_n$  is obtained by self-adaptive Runge-Kutta integration. The calculated Hall conductivity  $\sigma_{xy}^{(n)}/\tau_n$ , as a function of  $B\tau_n$ , is shown in Fig. 2. The calculation results for both  $\sigma_{xx}$  and  $\sigma_{xy}$  can be well interpolated by the Padé series:

$$\begin{aligned} \sigma_{xx}^{(n)}/\tau_n &= \frac{a_1^{(n)} + a_2^{(n)}(B\tau_n)^2}{1 + b_1^{(n)}(B\tau_n)^2 + b_2^{(n)}(B\tau_n)^4}, \\ \sigma_{xy}^{(n)}/\tau_n &= \frac{c_1^{(n)}(B\tau_n) + c_2^{(n)}(B\tau_n)^3}{1 + d_1^{(n)}(B\tau_n)^2 + d_2^{(n)}(B\tau_n)^4}, \end{aligned} \quad (2)$$

with the coefficients shown in Table I. The total conductivity is the summation of the four bands. The *ab initio* calculation reveals some unexpected behaviors: (i) The “electronlike”  $\pi_1$  band behaves as a hole band when the magnetic field is normal to the  $ab$  plane; (ii) The “holelike” band  $\pi_2$  has a sign change from positive to negative with increasing  $B\tau$ . The latter can be understood as a result of the complex structure of the  $\pi_2$  Fermi surface, which is composed of electronlike belly and holelike hills, as shown in the left inset of Fig. 2. At small  $B\tau$ , the conduction is dominated by the small orbits around the hills, showing the holelike behavior. The opposite happens when  $B\tau$  is large. These unusual behaviors clearly indicate the inapplicabil-

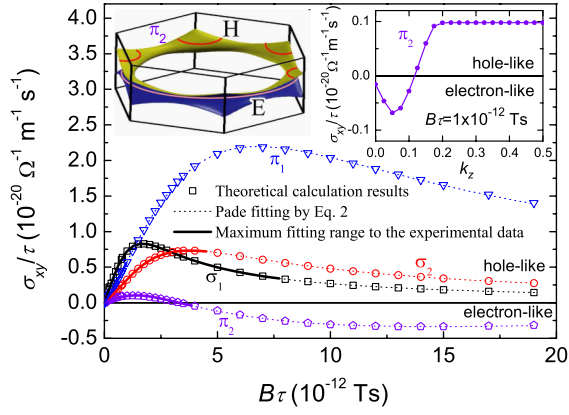


FIG. 2 (color online).  $B\tau$  dependence of  $\sigma_{xy}/\tau$  for the four bands. The left inset shows the Fermi surface of  $\pi_2$  band, the electronlike orbit (“E”) around the belly and the holelike orbit (“H”) where the hills are indicated as the arc lines. The right inset shows the partial Hall conductivity  $\sigma_{xy}^{(n)}(k_z)/\tau = \frac{e^2}{4\pi^3} \times \int dk_x dk_y \int_{-k_z}^{k_z} dk_z' v_x^{(n)}(\mathbf{k}) v_y^{(n)}(\mathbf{k}) (-\frac{\partial f}{\partial \epsilon})_{\epsilon=\epsilon_n(\mathbf{k})}$  contributed by orbits between  $-k_z$  to  $k_z$  of  $\pi_2$  band as a function of  $k_z$ . It demonstrates that the contribution from the belly region is negative while the hill region is positive.

ity of the simple formula in describing the magnetoresistance of  $\text{MgB}_2$  system [8].

By using the least-squares fit to multifunctions [24], we then fit the experimental data  $\sigma_{xx}(B)$  and  $\sigma_{xy}(B)$  (derived from the resistivity by  $\sigma_{xx} \equiv \rho_{xx}/(\rho_{xx}^2 + \rho_{xy}^2)$  and  $\sigma_{xy} \equiv \rho_{xy}/(\rho_{xx}^2 + \rho_{xy}^2)$ ) at each temperature to Eq. (2) by adjusting the four electron scattering times. The maximum range of  $B\tau$  in the fitting are plotted by the thick solid lines as shown in Fig. 2. Figure 3 shows the temperature dependence of four scattering times for samples  $RRR33.3$  and  $RRR20.9$ . Note that the fitting becomes less reliable at high temperature ( $T > 100$  K), as the nonlinearity of the magneto-resistivity becomes weaker.

To extend our fitting to higher temperature, we adopt a global fitting approach: Instead of fitting an individual curve at a time, we simultaneously fit all the curves for

TABLE I. Fitting parameters of Fig. 2 using Eq. (2) (All parameters are in SI unit).

$\sigma_{xx}$	$a_1$ ( $10^{-20}$ )	$a_2$ ( $10^4$ )	$b_1$ ( $10^{24}$ )	$b_2$ ( $10^{48}$ )
$\sigma_1$	1.66990	10.94632	6.76549	2.29927
$\sigma_2$	1.69337	0.29735	0.27378	0.01295
$\pi_1$	4.95528	2.20045	0.51383	0.00998
$\pi_2$	1.88788	0.32211	0.26752	0.00269
$\sigma_{xy}$	$c_1$ ( $10^{-8}$ )	$c_2$ ( $10^{16}$ )	$d_1$ ( $10^{24}$ )	$d_2$ ( $10^{48}$ )
$\sigma_1$	0.92146	2.59031	2.90378	0.93769
$\sigma_2$	0.28948	0.41532	1.08441	0.07650
$\pi_1$	0.45035	1.46465	2.26895	0.04918
$\pi_2$	0.12735	-0.01163	0.20542	0.00127

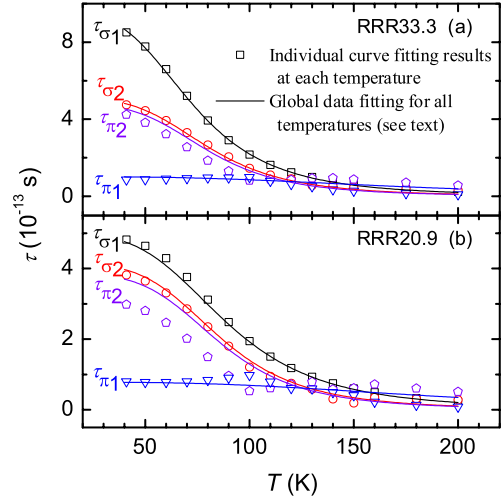


FIG. 3 (color online). Temperature dependence of calculated transport scattering times of the four bands for  $\text{MgB}_2$  films  $RRR33.3$  (a), and  $RRR20.9$  (b).

all temperatures. To do that, we assume  $1/\tau^{(n)} = 1/\tau_{\text{imp}}^{(n)} + 1/\tau_{e\text{-ph}}^{(n)}(T)$ , where  $\tau_{\text{imp}}^{(n)}$  is the scattering time from impurities and is  $T$ -independent, and  $\tau_{e\text{-ph}}^{(n)}(T)$  is due to the  $e$ -ph coupling, and can be modeled as  $1/\tau_{e\text{-ph}}^{(n)}(T) = \alpha_n T^{\beta_n}$ . The results are shown by the solid lines in Fig. 3. We find that both approaches coincide well.

We can separate the  $T$ -independent impurity contribution with the  $T$ -dependent  $e$ -ph contribution. In Fig. 4, we plot the temperature dependence of  $1/\tau_{e\text{-ph}}$  of three clean samples  $RRR33.3$ ,  $RRR24.9$ , and  $RRR20.9$ . The values from all the three samples are close to each other, indicating the  $e$ -ph coupling are not strongly affected by the impurities. The exponents  $\beta_n$  for the temperature dependence of  $1/\tau_{e\text{-ph}}$  are found to be 3.80 ( $\sigma_1$ ), 4.15 ( $\sigma_2$ ), 4.22

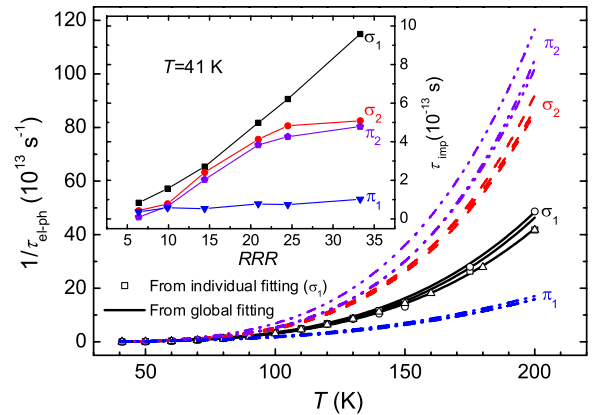


FIG. 4 (color online). Temperature dependence of  $1/\tau_{e\text{-ph}}$  of four bands for three different samples  $RRR33.3$ ,  $RRR24.5$ , and  $RRR20.9$ . The inset shows the correlation between the scattering time from the impurities and  $RRR$  for seven different samples.

( $\pi_1$ ), and 3.12 ( $\pi_2$ ), respectively. This is in accordance with the expectation of theory [25]. The results clearly show that the  $\sigma$  bands have stronger  $e$ -ph coupling than the  $\pi_1$  band, in agreement with the theoretical calculation [15]. Surprisingly, the  $e$ -ph coupling in the  $\pi_2$  band is also very strong. The difference between the  $e$ -ph coupling in the two  $\pi$  bands can be understood by our *ab initio* calculation: in the fitting range shown in Fig. 2, the magnetotransport of  $\pi_2$  band is dominated by the hills of its Fermi surface, while that of the  $\pi_1$  band is dominated by the belly (see insets of Fig. 2). Momentum changes to backscatter an electron in the hills of  $\pi_2$  band, as well as in the two sigma ones, are much smaller than that in the  $\pi_1$  band. As a result, phonon is much more effective in scattering electrons in  $\pi_2$  and two  $\sigma$  bands than in  $\pi_1$ . The intriguing result cannot be revealed without our fully band-resolved method.

The inset to Fig. 4 shows the dependence of the impurity scattering rate on RRR. It shows that disorder affects the impurity scattering much more strongly in the  $\sigma$  bands and  $\pi_2$  band than in the  $\pi_1$  band. The calculation results show that for the dirty sample RRR6.4, the scattering times for the four bands are comparable.

In conclusion, we have observed large magnetoresistance and nonlinear Hall effect in clean MgB<sub>2</sub> films, from which we have calculated the scattering times of each one of the four bands, in the aid of theory based on the Boltzmann equation and *ab initio* calculations. Surprisingly, the  $\pi_2$  band seems to be similar as the two  $\sigma$  bands, i.e., has a large scattering time in pure samples at low temperature, and also has a strong  $e$ -ph coupling. And the electron-phonon scattering is much weaker in the  $\pi_1$  band than other three bands, making the  $\pi_1$  band the least scattered at high temperatures. This may be caused by the different structure of the Fermi surface of this band. Disorder modifies the multiband electron scattering much in the same way as the phonon, reducing the scattering times of the two  $\sigma$  bands and  $\pi_2$  band to be comparable to the  $\pi_1$  band. This detailed knowledge on the band-resolved scattering rates allow to have deeper insight on the properties of MgB<sub>2</sub> than using the two-band approximation.

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- [1] P. C. Canfield and G. W. Crabtree, Phys. Today **56**, No. 3, 34 (2003).
- [2] A. Rydh *et al.*, Phys. Rev. B **70**, 132503 (2004); A. E. Koshelev and A. A. Golubov, Phys. Rev. Lett. **92**, 107008 (2004); A. A. Golubov and A. E. Koshelev, Phys. Rev. B **68**, 104503 (2003).
- [3] H. J. Choi *et al.*, Nature (London) **418**, 758 (2002).
- [4] H. Yang *et al.*, Phys. Rev. B **76**, 134513 (2007).
- [5] R. S. Gonnelli *et al.*, Phys. Rev. Lett. **89**, 247004 (2002).
- [6] M. R. Eskildsen *et al.*, Phys. Rev. Lett. **89**, 187003 (2002).
- [7] P. de la Mora, M. Castro, and G. Tavizon, J. Phys. Condens. Matter **17**, 965 (2005).
- [8] I. Pallecchi *et al.*, Phys. Rev. B **72**, 184512 (2005); I. Pallecchi *et al.*, Eur. Phys. J. B **52**, 171 (2006).
- [9] Q. Li *et al.*, Phys. Rev. Lett. **96**, 167003 (2006).
- [10] A. Y. Liu, I. I. Mazin, and J. Kortus, Phys. Rev. Lett. **87**, 087005 (2001); I. I. Mazin *et al.*, *ibid.* **89**, 107002 (2002).
- [11] J. Kortus *et al.*, Phys. Rev. Lett. **86**, 4656 (2001).
- [12] E. A. Yelland *et al.*, Phys. Rev. Lett. **88**, 217002 (2002).
- [13] A. Gurevich, Phys. Rev. B **67**, 184515 (2003).
- [14] S. L. Bud'ko *et al.*, Phys. Rev. Lett. **86**, 1877 (2001); D. G. Hinks, H. Claus, and J. D. Jorgensen, Nature (London) **411**, 457 (2001).
- [15] M. Monni *et al.*, Europhys. Lett. **81**, 67006 (2008).
- [16] V. Ferrando *et al.*, J. Appl. Phys. **101**, 043903 (2007).
- [17] M. Ortolani *et al.*, Phys. Rev. B **71**, 172508 (2005).
- [18] X. H. Zeng *et al.*, Nature Mater. **1**, 35 (2002); C. G. Zhuang *et al.*, J. Appl. Phys. **104**, 013924 (2008).
- [19] S. L. Bud'ko *et al.*, Phys. Rev. B **63**, 220503(R) (2001).
- [20] Yu. Eltsev *et al.*, Phys. Rev. B **66**, 180504(R) (2002).
- [21] N. W. Ashcroft and N. D. Mermin, *Solid State Physics* (Harcourt, Orlando, 1976).
- [22] Z. Fang and K. Terakura, J. Phys. Condens. Matter **14**, 3001 (2002).
- [23] J. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. **77**, 3865 (1996).
- [24] P. R. Bevington and D. K. Robinson, *Data Reduction and Error Analysis for the Physical Sciences* (McGraw-Hill Inc., New York, 2003).
- [25] For the simple metals, the low temperature  $e$ -ph transport scattering rate follows the so-called "Bloch  $T^5$  law." However, for systems with complex Fermi surfaces or small Fermi pockets, the backscattering of electron may not require large momentum transfer, and the transport scattering rate will be proportional to  $T^3$ . In general, the exponent should be in the range of 3–5. See Ref. [21].