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Measurement of the magnetic penetration depth of a superconducting MgB₂ thin film with a large intraband diffusivity

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We report the temperature dependent magnetic penetration depth $\lambda(T)$ and the superconducting critical field $H_{c2}(T)$ in a 500-nm MgB₂ film. Our analysis of the experimental results takes into account the two gap nature of the superconducting state and indicates larger intraband diffusivity in the three-dimensional (3D) π band compared to that in the two-dimensional (2D) σ band. Direct comparison of our results with those reported previously for single crystals indicates that larger intraband scattering in the 3D π band leads to an increase of λ . We calculated λ and the thermodynamic critical field $H_c \approx 2000$ Oe employing the gap equations for two-band superconductors. Good agreement between the measured and calculated λ value indicates the two independent measurements, such as magnetic force microscopy and transport, provide a venue for investigating superconducting properties in multi-band superconductors.

I. INTRODUCTION

During the last decade a significant effort has been made to understand the mechanism of two-band superconductivity in MgB₂.¹⁻⁴ MgB₂ has two s-wave gaps residing on four different disconnected Fermi surface (FS) sheets: two axial quasi two-dimensional (2D) σ -band sheets and two contorted three-dimensional (3D) π -band sheets. The σ band forms two concentric cylindrical sheets via in-plane sp^2 hybridization of the boron valence electrons. The π band results from the strongly coupled covalent bonding and antibonding of the boron P_z orbitals.⁵ Multiple bands allow for both inter- and intra-band scattering. It is thus possible to tune the upper critical field (H_{c2}) via doping, which has different effects on the inter- and intra-band scattering strengths.^{2,6-8} In MgB₂ the anisotropy of the temperature dependent penetration depth λ , $\gamma_\lambda(T) = \lambda_c(T)/\lambda_{ab}(T)$ shows remarkably different behavior compared to that of H_{c2} , $\gamma_{H_{c2}}(T) = H_{c2}^{ab}(T)/H_{c2}^c(T)$.^{9,10} This difference indicates that the two-band nature of superconductivity profoundly alters the superconducting properties compared to those in a single band material.¹¹ For example, the equations for critical fields and depairing current as a function of λ and ξ should be modified due to the inter/intra-band scattering. Knowledge of the absolute values of λ and ξ is also important for technological applications.¹² For example, the acceleration field in superconducting radio frequency (SRF) cavities could be enhanced by covering conventional superconducting Nb cavities with superconductor/insulator multilayers (such as MgB₂) with higher thermodynamic critical field (H_c).¹³

A number of measurements have been performed to determine the absolute value of λ in MgB₂.^{3,9,10} The reported values of λ range from 40 nm to 200 nm, indicating that λ is strongly affected by inter- and intra-band

scattering.¹⁴⁻²⁰ In this paper we present measurements of the absolute values of $\lambda(T)$, employing low temperature magnetic force microscopy (MFM), and of the angular-dependent $H_{c2}(T, \theta)$ performed via electrical transport, in a 500-nm thick MgB₂ film. Our MgB₂ film can be described by the dirty limit two-band Usadel equations. We analyze the measured values of H_{c2} and λ using a model developed for dirty superconductors,² which simplifies the analysis compared to that reported in Ref. 15. We investigate theoretically the influence of the inter/intra-band scattering on the superconducting properties. Using a two-band superconductor model with parameters obtained from a fit to $H_{c2}(T, \theta)$, we calculate λ and H_c which are consistent with the experimental values.

II. EXPERIMENT

A MgB₂ film was grown on a *r*-sapphire substrate by a reactive evaporation technique.^{21,22} The film is epitaxial and shows columnar growth morphology, with the *c* axis tilted by a few degrees from the normal direction of the substrate. For more details see Ref. 22. The sample has dimensions $L=4$ mm \times $W=5$ mm \times $t=500$ nm, and exhibits a full superconducting volume fraction based on measurements using a commercial SQUID magnetometer (Quantum Design magnetic property measurement system, MPMS). All MFM measurements described here were performed in a home-built low temperature MFM apparatus.²³ Temperature dependent vortex images were taken in the frequency-modulated mode after a small magnetic field was applied above T_c (field-cooled). We used high resolution SSS-QMFM cantilevers.²⁴ The magnetic field was always applied perpendicular to the film surface and parallel to the MFM tip. The absolute values of $\lambda(T)$ were determined by comparing the Meissner response curves with those for a reference sample at

4 K.^{25,26} The Meissner technique for the λ measurement was first proposed by Xu *et al.*²⁷ and demonstrated by Lu *et al.*²⁸ The film thickness of 500 nm is larger than $\lambda \approx 200$ nm, which makes corrections to λ due to the sample thickness insignificant. Conventional four-lead resistivity measurements used for determining $H_{c2}(T, \theta)$, where θ is an angle between the applied magnetic \mathbf{H} and the crystallographic c axis, were performed with a rotatable probe in a commercial Quantum Design physical property measurement system (PPMS), in magnetic fields between 0 T and 9 T. The superconducting critical temperature $T_c = 38.3$ K (zero resistance) and the transition width $\Delta T_c = 0.5$ K were determined from the transport measurements. Zero-field-cooling measurements at the MPMS with $H \approx 1$ Oe show $T_c = 38.0$ K. The small value of residual resistivity ratio ($\text{RRR} \approx 4$) indicates the presence of impurities, consistent with the dirty limit.

III. RESULTS AND DISCUSSION

A. MFM measurements in the MgB_2 film

Figure 1(a) presents a typical vortex image in the MgB_2 thin film. The well-formed vortices in the $6 \mu\text{m} \times 6 \mu\text{m}$ field of view were observed, which suggests the homogeneity of the sample on a micron scale. However, the irregular shape of individual vortices suggests the presence of inhomogeneity in the superfluid density on a sub-micron scale, which may be related to impurities. Figures 1(b) and (c) show MFM images of isolated vortices in MgB_2 at 4 K and 15 K, respectively. The features besides a single vortex represent a sub-micron scale inhomogeneity, indicating small variations of superfluid density. Figure 1(d) depicts a line profile taken along the dotted line in Figs. 1(b) and (c). The maximum force gradient [$\max(\partial f / \partial z)$] at the center of the vortex qualitatively indicates that the magnitude of λ at 15 K is larger than that at 4 K.^{29–31} In order to determine the absolute value of λ , we performed Meissner experiments. The force between the tip magnetic moment (a distance d above the sample) and the shielding currents induced by the tip field is equal to the force between the real tip and the image tip, with the mirror plane at a distance λ below the sample's surface.³² This force therefore is a function of $d + \lambda$ when $d \gg \lambda$. Direct comparison of the Meissner curves taken at 4 K for MgB_2 and a reference sample (Nb) with a known λ gives $\lambda(4 \text{ K}) = 200 \pm 30$ nm for MgB_2 .²⁵ Comparing Meissner curves for MgB_2 at 4 K and at a given temperature T yields $\delta\lambda(T)$. We obtain the absolute value of the temperature dependent $\lambda(T)$ by adding $\delta\lambda(T)$ to $\lambda(4 \text{ K})$. Figure 2(a) shows the Meissner force response as a function of the tip-sample distance at several temperatures. The systematic evolution of the Meissner response with respect to temperature reflects the change of λ with temperature. Figure 2(b) shows the normalized $\lambda(T)$ (black squares) obtained for MgB_2 using the procedure outlined above,

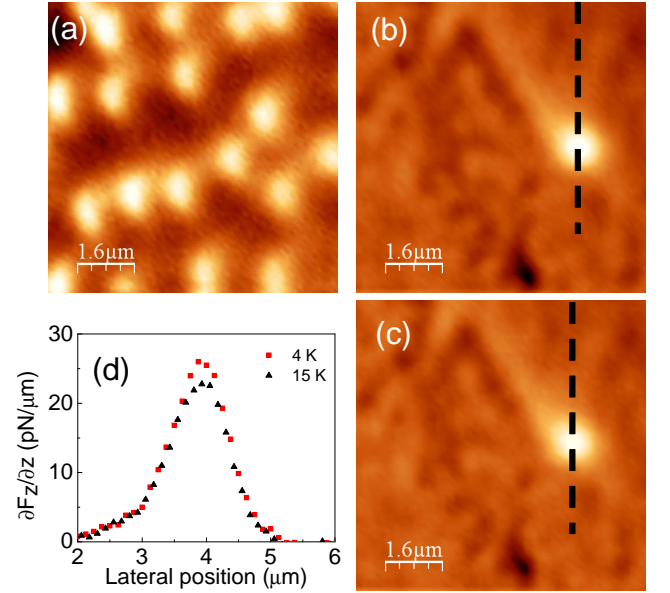


FIG. 1: (Color online) (a) A typical vortex image with a tip-lift height of 300 nm in the MgB_2 thin film. (b) and (c) Single vortex images with a tip-lift height of 300 nm, acquired at $T = 4$ K and $T = 15$ K, respectively. (d) The single vortex profile along the dotted lines in (b) and (c). Higher peak value corresponds to a smaller λ value.

deviating significantly from the BCS theory curve (the red dashed line), which is consistent with the previous studies shown as green solid circles.¹⁰ This discrepancy indicates a profound effect of two-band superconductivity in MgB_2 .¹⁰ The large λ in MgB_2 may be due to inclusion of impurities, such as C, N, and Al, which significantly affects the electron mean-free path in each band of MgB_2 .

B. H_{c2} measurements in the MgB_2 film

In order to investigate the nature of disorder, we performed temperature dependent H_{c2} measurements. Figure 3(a) shows $H_{c2}(T)$ with the field parallel to the c axis $H_{c2}^{\parallel c}(T)$ (black circles). The value of $H_{c2}(0)$ is considerably higher than that found in clean single crystals ($H_{c2}^{\parallel c}(0) \approx 3\text{--}5$ T)³³, which indicates that the film is in the dirty limit. The Gurevich model for two-band superconductors² considers inter- and intra-band scattering by non-magnetic impurities in the dirty limit. The high T_c in our film (which shows essentially no suppression compared to the clean crystals) is consistent with a small inter-band scattering, so we can use the equations obtained for $H_{c2}(T)$ neglecting the inter-band scattering:

$$a_2[\ln(t) + U(\eta h)] + a_1[\ln(t) + U(h)] + a_0[\ln(t) + U(h)][\ln(t) + U(\eta h)] = 0, \quad (1)$$

where $U(x) = \psi(1/2 + x) - \psi(1/2)$, $\psi(x)$ is the digamma function, $a_1 = 1 + L_-/L_0$, $a_2 = 1 - L_-/L_0$,

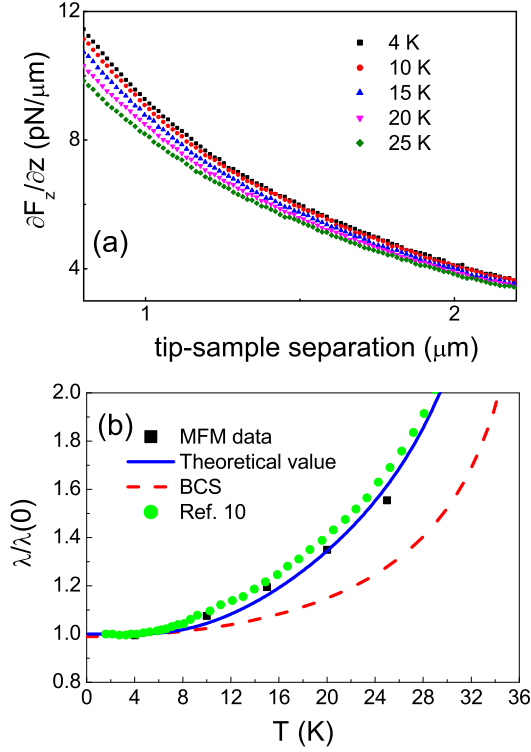


FIG. 2: (Color online) (a) Temperature dependence of the Meissner response in MgB₂. (b) $\lambda(T)$ marked by the black squares are inferred from the data shown in (a). The blue solid curve shows the calculated $\lambda(T)$ from the gap equations for two-band superconductors. The red dashed curve represents the conventional BCS model. The green circles are taken from tunnel diode resonator measurements (Ref. 10).

$a_0 = 2w/L_0$, $L_0 = \sqrt{(L_-^2 + 4L_{12}L_{21})}$, $L_{\pm} = L_{11} \pm L_{22}$, $w = L_{11}L_{22} - L_{12}L_{21}$, $t = T/T_c$, $\eta = D_2/D_1$, and $h = H_{c2}D_1/2\Phi_0T$. Φ_0 is a single magnetic flux quantum, and D_1 and D_2 are the intraband diffusivities. The angular-dependent diffusivities $D_1(\theta)$ and $D_2(\theta)$ for both bands are calculated using the following equation:

$$D_m(\theta) = \sqrt{D_m^{(a)2} \cos^2 \theta + D_m^{(a)} D_m^{(c)} \sin^2 \theta}. \quad (2)$$

From equations (1) and (2), we can obtain $H_{c2}(T, \theta)$.

The diffusivity D_1^c along the c axis is smaller than the in-plane diffusivity D_1^{ab} in MgB₂ due to the nearly 2D nature of the σ band. On the other hand, the values of D_2^c and D_2^{ab} do not differ substantially because of the isotropic 3D nature of the π band. The resulting relations among diffusivities are $D_1^c \ll D_1^{ab}$ and $D_2^c \approx D_2^{ab}$, which leads to the anomalous behavior of the anisotropy of $H_{c2}(T)$. The in-plane diffusivity ratio D_1^{ab}/D_2^{ab} is an important parameter in the equation (1).

We performed a numerical fit to three sets of transport data such as $H_{c2}(T)$ at $\theta = 0^\circ$, $H_{c2}(\theta)$ at $T = 22$ K, and $T = 32$ K using the equations (1) and (2), shown in

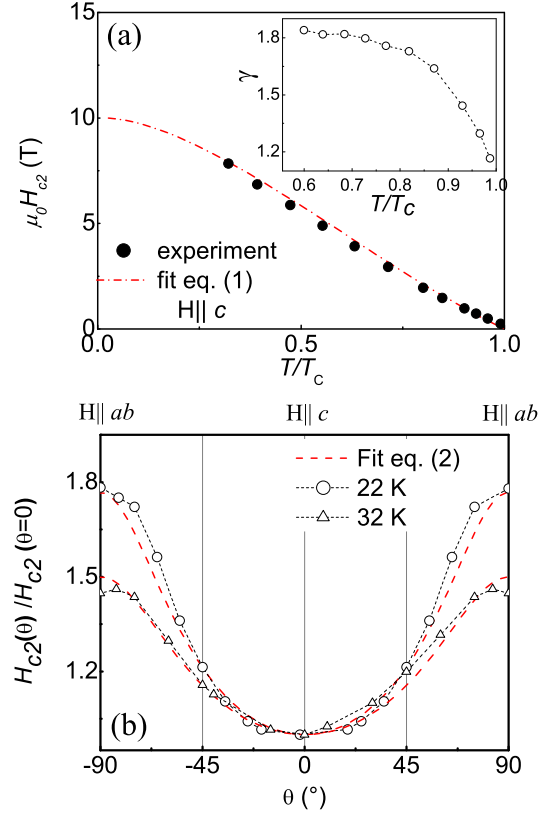


FIG. 3: (Color online) (a) Numerical fit to $H_{c2}(T)$ obtained from transport data. The inset shows the temperature dependence of the anisotropy of H_{c2} . (b) Numerical fit to $H_{c2}(\theta)$ at 22 K and 32 K with the same parameters used to fit $H_{c2}(T)$. From the fit, the diffusivity values of $D_1^{ab} = 2.36$ cm²/s and $D_2^{ab} = 19.7$ cm²/s were obtained; the coupling parameters obtained from the fit are $L_{\sigma\sigma} \approx 0.810$, $L_{\pi\pi} \approx 0.285$, $L_{\sigma\pi} \approx 0.25$, and $L_{\pi\sigma} \approx 0.18$, respectively, close to the values obtained from ab-initio calculations (Ref. 34). The uncertainty of the fit parameters is no more than 5%, which is smaller than our experimental errors of 10%.

Fig. 3. The relation between the best fit intraband diffusivities in the σ and π bands is $D_2^c = 8.5 \times D_1^c$. This large $\eta = 8.5$ is consistent with the absence of a sharp upward curvature in $H_{c2}^{\parallel c}(T)$ at low T (see Fig. 1 in Ref. [2]), frequently observed in C-doped MgB₂ with extremely high H_{c2} . The inset of Fig. 3(a) shows the anisotropy $\gamma_{H_{c2}}(T)$ as a function of T . Again, this behavior is qualitatively consistent with that expected for $\eta \gg 1$, see Fig. 3(c) in Ref. [2]. The superconducting critical field, $H_{c2}^{\parallel c}(0)$, for field applied parallel to the c axis, obtained from the fit, equals 10 T. This indicates the presence of strong multiple intraband scattering channels. The value of in-plane intraband diffusivity ratio $\eta = 8.5$ provides information about the type of the intraband scatterers. The larger value of η , (smaller value of D_1^c) indicates the weakening of the 2D σ band by certain types of impurities, such as C and N. These impurities affect the 2D landscape by

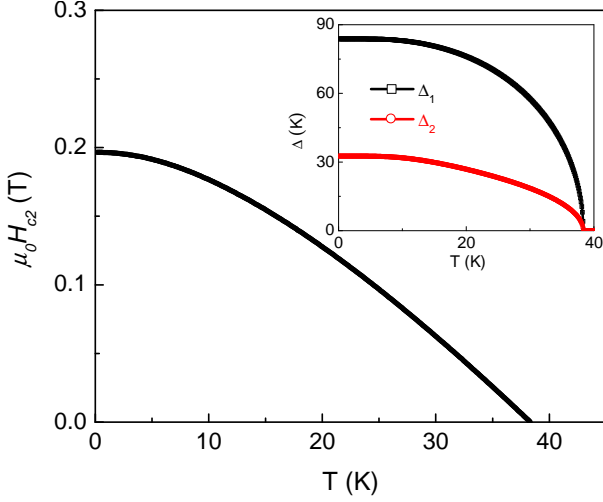


FIG. 4: (Color online) The calculated thermodynamic critical field H_c from the gap equations for two-band superconductors. The inset shows the calculated gap values from the two band model.

replacing p_{xy} orbitals of boron, and making the system more isotropic. The large value of D_2^a compared to D_1^a is also in good agreement with results from the α model,¹⁰ and is the result of a large contribution of the π band to the total density of states.

C. λ and H_c from the two-band model

We calculated λ using the parameters obtained from the $H_{c2}(T, \theta)$ fit and the band calculations. The London equation for a two-gap superconductor is given by $\nabla \times (\lambda_L^2 \nabla \times \mathbf{H}) + \mathbf{H} = 0$, where the London penetration depth is $\lambda_L^{-2}(T) = \pi e^2 \mu_0 (N_1 D_1^{ab} \Delta_1 \tanh \frac{\Delta_1}{2T} + N_2 D_2^{ab} \Delta_2 \tanh \frac{\Delta_2}{2T})$: Indices of 1 and 2 represent the σ band and the π band, respectively. N_1 and N_2 are the electron densities of states. Δ_1 and Δ_2 are the gap magnitudes. D_1^{ab} and D_2^{ab} are the intraband diffusivities.² Using $D_1^{ab} = 2.36 \text{ cm}^2/\text{s}$, $D_2^{ab} = 19.7 \text{ cm}^2/\text{s}$, obtained from the fit of $H_{c2}(T, \theta)$, $\Delta_1(0) = 84 \text{ K}$, $\Delta_2(0) = 33 \text{ K}$, obtained from the gap Eqs. (3), $N_1 = 0.3 \text{ states/a}^3\text{eV}$, and $N_2 = 0.41 \text{ states/a}^3\text{eV}$ with a unit cell volume of $a^3 = 87.2 \text{ \AA}^3$ ¹⁴ (obtained from the band calculations),³⁴ we obtain $\lambda_L(0) \approx 170 \text{ nm}$, consistent with the measured value of $\lambda_{ab}(0) = 200 \pm 30 \text{ nm}$. The calculated $\lambda_L(T)$ is shown as the blue curve in Fig. 2(b), consistent with the MFM experiment. This indicates that the two independent measurements of $\lambda(T)$ (MFM) and $H_{c2}(T)$ (transport) in MgB_2 are complementary for investigating superconducting properties.

The thermodynamic critical field (H_c) in MgB_2 is important for technological applications.¹² We evaluate H_c using the band coupling parameters, obtained from $H_{c2}(T, \theta)$, and the electron density of states obtained

from the band calculations.

The gap equations for two-band superconductors³⁵ are

$$\hat{g} \begin{pmatrix} \Delta_1 \\ \Delta_2 \end{pmatrix} - \begin{pmatrix} N_1(0)\Delta_1 Y(\Delta_1) \\ N_2(0)\Delta_2 Y(\Delta_2) \end{pmatrix} = 0, \quad (3)$$

with $Y(\Delta_j) = \int_0^{\omega_c} d\xi \frac{1}{\sqrt{(\xi^2 + |\Delta_j|^2)}} \tanh \left[\frac{\sqrt{\xi^2 + |\Delta_j|^2}}{2k_B T} \right]$, where \hat{g} is the superconducting coupling matrix with $g_{11} = N_1 L_{22}/w$, $g_{12} = g_{21} = N_1 L_{12}/w = N_2 L_{21}/w$, and $g_{22} = N_2 L_{11}/w$. ω_c is some unknown cutoff frequency obtained from Eqs. (3) using the T_c obtained from the transport data. Using the parameters obtained from the fit of $H_{c2}(T, \theta)$, we have the superconducting coupling matrix, $\hat{g} = \begin{pmatrix} 0.46 & -0.40 \\ -0.40 & 1.78 \end{pmatrix} / (a^3 \text{eV})$. The free energy is calculated³⁵ as $\mathcal{F} = \sum_{ij} (\Delta_i g_{ij} \Delta_j^*) - \frac{4}{\beta} \sum_i N_i \int_0^{\omega_c} d\xi \ln \left(\frac{\cosh(\frac{1}{2}\beta \sqrt{|\Delta_i|^2 + \xi^2})}{\cosh(\frac{1}{2}\beta \xi)} \right)$. Then H_c is given by $H_c^2/8\pi = -\mathcal{F}$. We calculate $\Delta_1(T)$ and $\Delta_2(T)$ as shown in Fig. 4 (inset). The calculated gap values at zero temperature are $\Delta_1(0) = 84 \text{ K}$ and $\Delta_2(0) = 33 \text{ K}$, which are slightly larger than reported values.¹⁰ The thermodynamic critical field at zero temperature, calculated from the two-band model, is approximately 2000 Oe. This value is smaller than those previously obtained in polycrystalline MgB_2 by specific heat measurements³⁶ and the values reported in clean single crystals.^{37,38}

As discussed earlier, the superconducting properties in multiband superconductors are affected by the interactions among the bands.³⁷ We obtain $\xi_{ab}(0) = 5.7 \text{ nm}$ using $H_{c2}(0) = \Phi_0/2\pi\xi^2(0)$ and our experimental value $H_{c2}^{\parallel c}(0) = 10 \text{ T}$. We can then use the Ginzburg-Landau theory to estimate the thermodynamic critical field in the film, $H_c = \Phi_0/2\sqrt{2}\pi\lambda(0)\xi(0) = 2100 \pm 300 \text{ Oe}$. This value is close to the calculated value of $H_c = 2000 \text{ Oe}$ from the two band model. This suggests that the strong intraband scattering in the 3D π band makes the system more isotropic, and thus the system shows single band characteristics.

IV. CONCLUSION

In conclusion, we have measured $\lambda_{ab}(T)$ and $H_{c2}(T, \theta)$ in a MgB_2 film. Our analysis of $H_{c2}(T, \theta)$ shows that the large value of the in-plane intra-band diffusivity in the 3D π band is due to the presence of non-magnetic impurities such as C and N, indicating the system is more isotropic, which is partly responsible for a large λ . We calculated λ and H_c employing the gap equations for the two-band superconductors using the parameters obtained from $H_{c2}(T, \theta)$ and derived from band calculations. The calculated $\lambda_L(0) \approx 170 \text{ nm}$ is close to the measured $\lambda(0) = 200 \pm 30 \text{ nm}$ in MgB_2 film, indicating that two independent measurements, such as MFM and transport, are complementary, and provides a venue for thoroughly investigating superconducting properties.

The determination of $H_c(T)$ in clean MgB_2 and in multi-band superconductors in general is a fascinating problem with both fundamental and technological relevance.

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