Two-band effect on the temperature and the angle dependences of the ratio of the surface to the bulk superconductivity in MgB$_2$

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Abstract. We investigated the ratio of the surface superconducting field ($H_{c3}$) to the bulk superconducting field ($H_{c2}$) in MgB$_2$ single crystals at different temperatures and at different angles between the field and the $c$-axis of the crystal by using electrical transport measurements. The temperature dependence of the ratio $H_{c3}/H_{c2}$ at different angles was qualitatively well described by a recent theory based on the two-gap nature of MgB$_2$. The angular dependence of the ratio shows that it can be both enhanced and reduced, which can be explained by the two-band theory. If slight deviations of $H_{c3}/H_{c2}$ from the value for the half-infinite geometry used in the theory are to be accommodated, effects due to the finite geometry and to the orientation of the applied field relative to the crystal plane need to be considered.
1. Introduction

In 1963, Saint-James and de Gennes predicted that in a decreasing magnetic field, the superconducting order parameter would nucleate at the sample surface at a field above the bulk superconducting field, \( H_{c2} \), when the applied magnetic field was parallel to the sample surface \([1]\). They assumed an infinite half-plane geometry and found that the surface superconducting field, \( H_{c3} \), is 1.695 times higher than \( H_{c2} \) (\( H_{c3} \approx 1.695H_{c2} \)), regardless of the temperature. A month later, two groups separately reported their experimental evidence proving the existence of surface superconductivity in thick films and foils \([2, 3]\). Since these pioneering works were confined to the half-plane and to simple boundary conditions under which no supercurrents could flow normal to the surface, subsequent studies have naturally focused on how a finitely curved geometry affects the surface nucleation of the superconducting order. From these efforts, the sample geometry was found to influence the surface nucleation \([4]–[6]\). For example, there have been results insisting that the ratio \( H_{c3}/H_{c2} \) for finite geometries is larger than that for the half-plane case (\( H_{c3}/H_{c2} > 1.695 \)) \([7]–[9]\). The most interesting results among the numerous calculations are the effects of the intrinsic anisotropy \([9]\) and of the two-band nature of a material \([10]\). Anisotropy in a single-gap superconductor causes the ratio \( H_{c3}/H_{c2} \) to be a function of the angle between the applied magnetic field and the crystal surface \([9]\), whereas anisotropic two-gap superconductivity causes the ratio \( H_{c3}/H_{c2} \) to have temperature dependence \([10]\).

Since the discovery of the two-gap nature \([11, 12]\) of the MgB\(_2\) superconductor \([13]\), intensive studies comparing its physical properties with those of single-gapped materials have been conducted. Various research efforts have shown that the anisotropy ratio of the bulk superconducting fields, \( H_{c3}^{ab}/H_{c2}^{c} \), differs from experiment to experiment even in the case of single crystals \([14]\), which might be attributed to the existence of surface superconductivity. As a result, the determination of \( H_{c3} \) has been an urgent problem, and \( H_{c3}/H_{c2} \) for MgB\(_2\) was found to be somewhat modulated from 1.695 \([14]–[17]\). Specifically, near the zero-field superconducting temperature \( T_c \), the ratio is about 1.6 for fields parallel to the \( c \)-axis and about 1.4 for fields parallel to the \( ab \)-plane \([14]–[16]\). However, detailed experiments addressing the temperature and the angular dependence of the \( H_{c3}/H_{c2} \) ratio for MgB\(_2\) single crystals, which might be affected by the finite geometry and by the anisotropic two-gap superconductivity, are still lacking. In addition, these intriguing variations in MgB\(_2\) still need to be explained theoretically. Beside this intriguing issue of MgB\(_2\), other interesting physics can be found elsewhere \([18]–[21]\).

In this research, we investigated not only the temperature dependence but also the angle dependence of \( H_{c3}/H_{c2} \) for MgB\(_2\) single crystals. The resistive transition and the critical
current ($I_c$) were measured at various applied magnetic fields ($H$) for different temperatures ($T$) and for different angles ($\theta$) between the field and the $c$-axis of the sample. The general features appearing in the temperature-dependent $H_{c3}/H_{c2}$, the ratio having a constant value for $H \parallel c$-axis and having a dip near $T_c$ for $H \parallel ab$-plane, are in good agreement with a numerical simulation based on the dirty two-gap Usadel equation [10, 22, 23]. However, subtle differences exist between the experimental and the theoretical [10] values of $H_{c3}/H_{c2}$ for $H \parallel c$-axis and for $H \parallel ab$-plane. When the field is directed along the $c$-axis, $H_{c3}/H_{c2}$ shows a somewhat larger value than the theory predicts. On the other hand, the ratio shows a smaller value than the theoretical one when the field lies in the $ab$-plane. Since the two-band theory considers only a half-plane, the enhanced ratio for $H \parallel c$-axis and the reduced ratio for $H \parallel ab$-plane near $T_c$ could be resolved, respectively, by the effects of the finite geometry and of the field’s orientation relative to the crystal plane.

2. Experiments

$\text{MgB}_2$ single crystals were obtained by using a high-pressure technique. A description of the process for crystal growth of $\text{MgB}_2$ and of the basic properties of $\text{MgB}_2$ can be found elsewhere [24]. To minimize the contact resistance for the electronic transport measurements, we patterned four metal leads on the clean surface of an as-grown rectangular-shaped $\text{MgB}_2$ single crystal (dimensions: $200 \times 40 \times 10 \mu\text{m}^3$) with $T_c \approx 35$ K and with a transition width of $\Delta T_c \approx 0.3$ K.

In order to investigate the temperature ($T$) and angle ($\theta$) dependences of the ratio $H_{c3}/H_{c2}$ for $\text{MgB}_2$ single crystals, we obtained $H_{c2}$ and $H_{c3}$ from the resistance and the $I$–$V$ curves with increasing $H$ at various $T$ and $\theta$, $\theta$ being the angle between the field and the $c$-axis of the crystal. To determine the critical current ($I_c$), we chose a voltage criterion in the $I$–$V$ curve so that the voltage did not exceed a value that allowed the vortex to cross the entire sample. In our sample, the estimated voltage was about $100 \text{nV}$. Hence, a $100 \text{nV}$ criterion was used in the $I$–$V$ characteristics to determine the $I_c$.

3. Results and discussion

Figure 1 shows the applied magnetic field dependence of the critical current $I_c(H)$ at different angles ($\theta = 0^\circ$ ($H \parallel c$-axis), $30^\circ$, $60^\circ$ and $90^\circ$ ($H \parallel ab$-plane)) and at $T = 20 \text{K}$ in $\text{MgB}_2$ single crystals. For $0^\circ$, $30^\circ$ and $60^\circ$, peak effects appear, and long tails are observed above the peak as the field is increased. Since the peak effect ends right before the bulk superconducting field $H_{c2}$, the tails indicate that a superconducting phase still exists at fields even above $H_{c2}$, which is reminiscent of the surface superconducting field $H_{c3}$. In those $I_c(H)$ curves, $H_{c2}$ and $H_{c3}$ can be chosen as indicated by arrows in the inset of figure 1. $H_{c2}$ was determined as the cross point where the dashed line along the steeply decreasing portion of the $I_c(H)$ curve after the peak intersects the dashed line along the long tail after $H_{c2}$, as shown in the inset of figure 1. $H_{c3}$ was determined to be the point where the critical current took on a value of the order of $10^{-4}$ A. However, for $90^\circ$, the peak and the tail are not observed due to the instrumental field limit. Therefore, we reinforced and cross-checked the values of $H_{c2}$ and $H_{c3}$ obtained at different $\theta$ and $T$ from $I_c(H)$ curves by comparing the values obtained from the field-dependent resistance, $R(H)$, at different $\theta$ and $T'$ for various magnitudes of the excitation currents.
Figure 1. Applied magnetic field dependence of the critical current for different $\theta$, which is the angle between the field and the $c$-axis, measured at 20 K in MgB$_2$ single crystals. The inset describes the method for estimating $H_{c2}$ and $H_{c3}$ for 0°. The details are in the text.

The representative data for $\theta = 0^\circ$ and $90^\circ$ at $T = 24$ K are shown in figures 2(a) and (b), respectively. In these current-dependent $R(H)$ curves, we determined $H_{c2}$ and $H_{c3}$ by using a method explained in early reports [2], [14]–[16], [25]. Because as the current increases, the portion of the current in the bulk area becomes larger and the resistive transition tends to occur at the bulk superconducting field, $H_{c2}$, we choose $H_{c2}$ at the position where, independent of the current, the resistance sharply drops. For instance, the $R(H)$ curves for 4 and 7 mA in figure 2(a) can be used for this process. The determined values of $H_{c2}$ are displayed with downward arrows in figure 2. $H_{c3}$ was determined from the common saturation point at which all the $R(H)$ curves for different excitation currents merge and is shown by the upward arrows in figures 2(a) and (b). The values of $H_{c2}$ and $H_{c3}$ obtained from the $I_c(H)$ and the $R(H)$ curves were consistent with each other within the error range. Also, values of $H_{c2}$ are in good agreement with those from different experimental techniques [14]–[16], [25]–[27].

Figure 3 presents the ratio $H_{c3}/H_{c2}$, which is our main interest, obtained from the $I_c(H)$ curves in figure 1 and the $R(H)$ curves in figure 2 as a function of the reduced temperature $T/T_c$ at different $\theta$. As figure 3 shows, the data set for $90^\circ$ is absent below 20 K because in this temperature range, $H_{c2}$ and $H_{c3}$ are estimated to be above our experimental limit, as shown in figure 1(b). The solid lines are guides for the eye. At $\theta = 0^\circ$, the ratio $H_{c3}/H_{c2}$ is constant in temperature, but at $\theta = 90^\circ$, the ratio shows temperature-dependent behavior with a dip. Those features of the ratio at $0^\circ$ and $90^\circ$ are consistent with the behavior corresponding to the two-gap effect of surface superconductivity, as suggested by Gorokhov [10]. Especially, the position of the dip for $\theta = 90^\circ$ has a value close to the theoretically predicted value of $T/T_c \approx 0.8$ [10]. Interestingly, the position of the dip at $\theta = 60^\circ$ is shifted away from $T/T_c = 1$, while the observation of the dip at $\theta = 30^\circ$ is not clear in this error range.
Figure 2. Resistance as a function of the applied magnetic field measured with different excitation currents (1, 4 and 7 mA) at $T = 24$ K for (a) $\theta = 0^\circ$ ($H \parallel c$-axis) and (b) $\theta = 90^\circ$ ($H \parallel ab$-plane). Criteria for determining $H_{c2}$ and $H_{c3}$ in (a) and (b), which are presented as arrows, are described in the text.

According to a theoretical calculation done by using the Ginzburg–Landau equations for one-gap superconductivity, the ratio $H_{c3}/H_{c2}$ is constant in $T$, independent of the field’s direction [1]. However, a theoretical work concerning two-gap superconductivity suggested that the ratio had different temperature dependences for two field directions, $\theta = 0^\circ$ and $90^\circ$ [10]. The ratio $H_{c3}/H_{c2}$ is independent of $T$ for $\theta = 0^\circ$, but as $\theta$ increases to $90^\circ$, $H_{c3}/H_{c2}$ comes to be a function of temperature because the contribution of the $\pi$ band becomes important [10]. The effect of the $\pi$ band on the ratio $H_{c3}/H_{c2}$ depends on the diffusivity ratio $\gamma_\alpha = \tilde{D}_{\alpha,x}/\tilde{D}_{\alpha,z}$, where $\alpha$ designates the $\pi$ or the $\sigma$ band. At $0^\circ$ ($H \parallel c$-axis), $\gamma_\alpha$ for each band equals 1, so the two operators governing the order parameters become equivalent, leading to the trivial case $H_{c3}/H_{c2} = 1.695$. However, at $90^\circ$ ($H \parallel ab$-plane), $\gamma_\sigma \gg \gamma_\pi$ so that one has to solve coupled equations for the order parameters. In a numerical simulation, a small dip in $H_{c3}/H_{c2}(T)$ appears near $T/T_c = 1$ [10]. From the above discussion, we can guess why the dip disappears as we lower an angle. The dip in the $H_{c3}/H_{c2}$ most clearly appears due to the difference between $\gamma_\sigma$ and $\gamma_\pi$ at $90^\circ$. If we slightly reduce an angle, the difference between $\gamma_\sigma$ and $\gamma_\pi$ becomes
Temperature dependence of the ratio \( H_{c3} / H_{c2} \) of MgB\(_2\) single crystals for fields directed along the \( c \)-axis (0°) and off the \( c \)-axis (30°, 60° and 90°).

Figure 4. Angle dependence of the ratio \( H_{c3} / H_{c2} \) obtained at \( T / T_c \approx 0.95 \).

smaller than it was at 90° and at the same temperature. In this case, the coupling between the two equations that we have to solve becomes weak and also the anomaly will be weaker.

On the other hand, Gorokhov’s theory [10], the two-gap theory for a half-plane, suggests that \( H_{c3} / H_{c2} \) near \( T_c \) can change with field direction: \( \sim 1.7 \) for \( \theta = 0^\circ \) and \( \sim 1.6 \) for \( \theta = 90^\circ \). However, for our MgB\(_2\) single crystals, the ratio becomes \( \sim 1.8 \) for \( \theta = 0^\circ \) and \( \sim 1.4 \) for \( \theta = 90^\circ \), as shown in figure 4, which are somewhat higher and lower than the values expected from theory. Furthermore, the position of the dip shifts to lower temperatures as the angle is decreased. In general, the deviations of \( H_{c3} / H_{c2} \) from 1.695, the value expected to appear in the isotropic single-gap superconductors, can be caused by several factors, such as an intrinsic anisotropy, the quality of the surface, the shape of the sample, and impurities or disorders that determine the clean and the dirty limits [4, 6, 9, 10]. First of all, among those factors listed above,
we cannot ignore the geometric effect on $H_{c3}/H_{c2}$ because we are dealing with rectangular-shaped MgB$_2$ single crystals having anisotropic two-gap and clean surface. Although no analytic mathematical expression is available for our situation (an angular-dependent ratio involving two-gap and finite-sample-geometry effects), we can obtain some physical insight about the angle dependence of the ratio $H_{c3}/H_{c2}$ from results of single-gapped materials because one-gap property is dominant near $T_c$. The values of $H_{c3}/H_{c2}$ near $T_c$ are shown in figure 4.

In the theory developed for an anisotropic one-gap superconductor by Kogan et al [9], corners and a finite cross section can enhance $H_{c3}/H_{c2}$ because superconducting order parameter tends to nucleate sample corners even above 1.695$H_{c2}$. In our case, for $\theta = 0^\circ$, the magnetic field is perpendicular to the rectangular cross section that has clear corners. So, the large value of $H_{c3}/H_{c2}$ for $\theta = 0^\circ$ compared to 1.695 can be explained by geometric effect. In addition, Kogan’s model also suggests small values of $H_{c3}/H_{c2}$ compared to 1.695, which result from the applied field being tilted from the principal axes of the crystal [9]. In our case, smaller values of $H_{c3}/H_{c2}$ were observed as the angle was increased. Even though the sample was perfectly perpendicular to the field at $\theta = 0^\circ$, a minor rotation around the c-axis will tilt the crystal surface away from the direction of the magnetic field when $\theta$ is increased. Finally, we conclude that the temperature and the angular dependence of the ratio $H_{c3}/H_{c2}$ in MgB$_2$ single crystal can be explained by using the two-gap and the finite-geometry effects.

4. Conclusion

We measured the bulk and the surface superconducting fields for MgB$_2$ single crystal by means of a resistive transition and a critical current for magnetic fields applied at different field directions and at different temperatures. The ratio $H_{c3}/H_{c2}$ was evaluated from those measurements as a function of the temperature and of the angle between the crystal c-axis and the magnetic field. The temperature-dependent behavior, that is the existence of a small dip near $T_c$ when the field is parallel to the $ab$-plane, is in good agreement with the previously developed theoretical prediction based on the two-gap effect. Moreover, the behavior of the dip when the field deviates from the $ab$-plane to the c-axis was well explained by considering the two-gap effect. As we moved the angle away from the c-axis, $H_{c3}/H_{c2}$ decreased from about 1.8 to 1.4, these values being both higher and lower than the result of Saint-James and de Genne ($H_{c3}/H_{c2} = 1.695$) appear to be due to both the two-gap nature and the finite geometry with anisotropy.

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