Direct evidence for two-band superconductivity in MgB$_2$ single crystals from directional point-contact spectroscopy in magnetic fields

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We present the results of the first directional point-contact spectroscopy experiments in high-quality MgB$_2$ single crystals. Due to the directionality of the current injection into the samples, the application of a magnetic field allowed us to separate the contributions of the $\sigma$ and $\pi$ bands to the total conductance of our point contacts. By using this technique, we were able to obtain the temperature dependency of each gap independent of the other. The consequent, strong reduction of the error on the value of the gap amplitude as function of temperature allows a stricter test of the predictions of the two-band model for MgB$_2$.

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During the last year, the consensus has been growing within the scientific community on the fact that most of the features of MgB$_2$ discovered so far can be properly explained by admitting that two band systems are present in this new superconductor: quasi-2D $\sigma$ bands arising from hybrid $sp^2$ orbitals in the boron planes, and 3D $\pi$ bands that stem from the out-of-plane $p_z$ orbitals. The unusual consequence of this band structure is that two different energy gaps can be observed in clean limit: $\Delta_\sigma$ (the larger) and $\Delta_\pi$ (the smaller). Both gaps are expected to close at the same temperature $T_c$ because of an inter-band pair-scattering mechanism but, while $\Delta_\sigma(T_c)$ should approximately follow a BCS-like curve, a marked reduction of $\Delta_\sigma(T_c)$ with respect to a BCS-like behavior is expected at $T \geq 20$ K.

So far, one of the most convincing experimental supports of this model has been the observation of two gaps by tunneling and point-contact spectroscopy in polycrystal samples and films. However, a direct and accurate test of the predictions of the two-band model has been so far impossible due to the lack of high-quality single crystals large enough to be used for direction-controlled point-contact and tunnel spectroscopy.

In this Letter, we present the results of the first directional point-contact measurements in large single crystals of MgB$_2$. We injected current along the $ab$ plane or along the $c$ axis, and applied a magnetic field either parallel or perpendicular to the $ab$ planes. This allowed us to separate the partial contributions of the $\sigma$ and $\pi$ bands to the total conductance, and to fit them obtaining the temperature dependency of each gap with great accuracy. We will show that all the results of this technique confirm very well the predictions of the two-band model.

The high-quality MgB$_2$ single crystals used for our point-contact experiments were produced at ETH (Zürich) by starting from a mixture of Mg and B. This mixture was put into a BN container and the crystals were grown at a pressure of 30-35 kbar in a cubic anvil device. The thermal process includes a one-hour heating up to 1700-1800°C, a plateau of 1-3 hours, and a final cooling lasting 1-2 hours. MgB$_2$ plate-like crystals up to 200 $\mu$m in weight and $1.5 \times 0.9 \times 0.2$ mm$^3$ in size can be obtained by using this technique, even though the crystals used in our measurements were smaller ($0.6 \times 0.6 \times 0.04$ mm$^3$ at most). The crystals were etched with 1% HCl in dry ethanol to remove possible deteriorated surface layers. The critical temperature of the crystals, measured by AC susceptibility, is $T_c = 38.2$ K with $\Delta T_c \sim 0.2$ K.

Using Au or Pt tips to make point contacts did not ensure mechanical stability during thermal cycling and reproducibility of the conductance curves. Thus, we moved to a non-conventional technique that consists in using as a counterelectrode either a very small ($\leq 50 \mu$m) drop of Ag conductive paint, or a small piece of indium pressed on the surface of the sample. With this technique, a control of the contact characteristics is possible anyway, by applying short voltage pulses to the junction. The apparent contact area is much greater than that required to have ballistic current flow, but the effective electrical contact occurs in a much smaller region, due to the presence of parallel micro-bridges in the spot area. On the other hand, the resistance of all our contacts was in the range $10 \div 50 \Omega$. This, together with the estimated mean free path for the same crystals $\ell = 80$ nm, proves that our contacts are in the ballistic regime. The contacts were positioned on the crystal surfaces so as to inject the current along the $c$ axis or along the $ab$ planes. The directionality of current injection is ensured...
by the small roughness of the crystal surfaces even on a microscopic scale. Figures 1(a) and (b) report AFM measurements on the ab-plane surface, after removal of the In contact; the surfaces perpendicular to the ab plane are even smoother.

Figure 1(c) shows some examples of the low-temperature normalized conductances dI/dV of contacts with current injection either parallel or perpendicular to the ab plane. All the conductance curves shown in the present Letter were normalized by dividing the measured dI/dV data by the linear or quartic function that best fits them for |V|>30 meV. The ab-plane curves clearly show two peaks at V ≃ ±2.7 mV and V ≃ ±7.2 mV, while the c-axis curves only show a peak at V ≃ ±(2.8 ± 3.5) mV and a smooth shoulder at V ≃ ±7.2 mV. These features, marked by dashed lines in the figure, are clearly related to the two gaps Δσ and Δπ. Solid lines are the best-fitting curves calculated by using the BTK model [10] generalized to the case of two bands, in which the normalized conductance σ is given by: σ = wσσσσ + (1 − wπ)σπ. Here, σσ and σπ are the normalized conductances for the π and σ bands, respectively, and wπ is the weight of the π band, that depends on the angle ϕ between the direction of current injection and the boron planes [4]. The fit is almost perfect, especially at low voltage, but it must be said that there are 7 adjustable parameters: the gaps Δσ and Δπ, the broadening parameters Γσ and Γπ, the barrier height coefficients Zσ and Zπ, plus the weight factor wπ. The normalization may yield additional uncertainty on Γσ,π and Zσ,π but does not affect the gap values.

Figures 2(a) and 2(b) show the temperature dependency of the normalized conductance curves (circles) of Ag-paint and In point contacts, respectively. The current was mainly injected along the ab planes in (a), and parallel to the c axis in (b). At the increase of the temperature, the typical two-gap features shown in Fig. 1 merge in a broad maximum, which disappears at the Tc of the junction that fell in all cases between 34.1 and 37.6 K. Since neither the current direction nor the contact resistance depend on the temperature, in fitting the conductances at various temperatures we kept both wπ and the barrier parameters Zσ and Zπ equal to their low-T values, thus reducing the actual adjustable parameters to 4. The best-fit curves are shown in Fig. 2 as solid lines.

The inset of Fig. 2 reports the temperature dependency of the two gaps, obtained by fitting the conductance curves of various contacts. The relevant barrier parameters (independent of temperature) are Zσ = 0.5 ± 1.4 and Zπ = 0.3 ± 0.8 depending on the junction, while the broadening parameters Γσ,π increase with T always remaining in the range between 0.5 and 3 meV. An important result is that the average values of the σ-band weight factor resulting from the fits (wπ = 0.75 ± 0.03 for ab-plane current, and wπ = 0.980 ± 0.005 for c-axis current) are in very good agreement with the values predicted by the two-band model (wπ = 0.66 and wπ = 0.99, respectively [4]). The small mismatch that actually exists can be ascribed to the fact that, in our low-barrier contacts, the current is injected within a finite solid angle. The integration of the theoretical wπ(ϕ), taking into account the cos ϕ dependency of the electron injection probability, shows that our experimental values of wπ are compatible with cone apertures of about 26° and 60°, respectively.

The average low-temperature gap values Δσ = 7.1 ±
0.5 meV and $\Delta_\pi = 2.9\pm 0.3$ meV agree very well with the theoretical values predicted by the two-band model [2, 4]. Nevertheless, at $T/T_c \gtrsim 0.5$ the experimental uncertainty on the gap value increases so much that it becomes practically impossible to determine whether the $\Delta_\pi(T)$ and $\Delta_\sigma(T)$ curves strictly follow a BCS-like curve or not. Clearly, this problem is also present in all the previous point-contact or tunneling experiments in which the temperature dependency of the gaps was obtained.

A careful and reliable test of the predictions of the two-band model obviously requires a more accurate determination of the gaps and their temperature dependency. Only by reducing the number of free fitting parameters, e.g. by separating the contributions of the two bands to the total conductance, this goal might be obtained. On the basis of the recent results obtained by Szabó et al. [7] in polycrystalline samples exposed to magnetic fields, we developed a technique that combines the selective removal of one gap with the directional point-contact spectroscopy. By applying to each junction (at low temperature) magnetic fields of increasing intensity, either parallel to the $ab$-plane contact and normal to the $c$ axis (a), the peaks due to the large gap merge together at $B \gtrsim 4$ T giving rise to a broad maximum. In addition to this, if the field only slightly exceeds 1 T the conductance curves remain practically unchanged (see Fig. 3). These results demonstrate that: i) the $\pi$ band is quite isotropic and its critical field at 4.2 K is around 1 T; ii) the $\sigma$ band is anisotropic and, at 4.2 K, $\Delta_\sigma$ is unaffected by a field of 1 T parallel to the $ab$ plane; iii) the $\sigma$-band critical field parallel to $ab$ is rather high ($> 9$ T) at low temperature, in agreement with other results on similar samples [1].

In addition to this, some preliminary measurements we made in $c$-axis contacts with $B || ab$ at about 30 K have shown that $B^c_{\pi \parallel ab} \sim 3.5$ T. A detailed discussion of the temperature dependency of the critical fields determined by our Andreev reflection experiments will be given in a forthcoming paper [2]. Nevertheless, on the basis of the aforementioned results, we can be confident that a field of 1 T parallel to the $ab$ planes is too weak to affect seriously the large gap, even at temperatures close to $T_c$. This hypothesis will be confirmed by the $\Delta_\sigma(T)$ curve (see Fig. 5(c)) that shows a BCS-like behavior with no anomalous high-$T$ gap suppression due to the field.

As a consequence, we measured the conductance of a $\text{InMgB}_2$ $c$-axis contact at 4.6 K, with no field (see Figure 4, open circles) and with a field of 1 T parallel to the $ab$ planes (see Figure 4, light gray circles). When the magnetic field destroys the gap in the $\pi$ band, the normalized conductance becomes: $\sigma(B=1T) = w_\pi + (1-w_\pi)\sigma_\sigma$. This function contains only three free parameters: $\Delta_\pi$, $\Gamma_\sigma$ and $Z_\sigma$, whose best-fit values at $T = 4.6$ K are 7.1 meV, 1.7 meV and 0.6, respectively. In fact, we took $w_\pi = 0.98$, that is the value obtained from the fit of the total $c$-axis conductance at the same temperature. An independent determination of the small gap can be obtained by subtracting the conductance curve measured in the presence of the field from that measured without

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**FIG. 3:** (a) Some experimental normalized conductance curves of an $ab$-plane contact, in increasing magnetic fields parallel to the $c$ axis. Thick lines represent the curves measured at $B = 1$ T and $B = 4$ T. (b) Same as in (a) but for an $ab$-plane contact with $B \parallel ab$-plane. Thick lines represent the conductances at $B = 1$ T and $B = 9$ T.

**FIG. 4:** Open circles: normalized conductance of a $c$-axis contact, with no magnetic field. Light gray circles: conductance of the same contact with a field of 1 T applied parallel to the $ab$ plane. Dark gray circles: difference between the two previous curves (suitably shifted). Solid lines are the best-fit curves given by the appropriate BTK model (see text).
field. The resulting curve, vertically shifted by one unit, is reported in Fig. 4 (dark gray circles). The result of the subtraction can be expressed by the functional form \( \sigma(B=0) - \sigma(B=1T) = w_\pi (\sigma_\pi - 1) \). Fitting the experimental data to this function (again, with \( w_\pi = 0.98 \)) allows determining the three remaining free parameters \( \Delta_\pi, \Gamma_\pi \) and \( Z_\pi \), that assume at \( T = 4.6 \) K the values 2.8 meV, 2 meV and 0.6, respectively. Incidentally, the very good quality of the fits (solid lines in Fig. 4) further shows that the value of \( w_\pi \) is appropriate. Fig. 5 reports the temperature dependency of the curves already shown at 4.6 K in Fig. 4: the \( c \)-axis conductance in a field of 1 T parallel to the \( ab \) planes, \( \sigma(B=1T) \) (a) and the difference \( \sigma(B=0) - \sigma(B=1T) \) (b), with the relevant best-fitting curves (solid lines). Notice that the difference curves look particularly “clean” and noise-free since the subtraction also allows eliminating some experimental fluctuations that are present both in \( \sigma(B=0) \) and in \( \sigma(B=1T) \). The resulting fits are quite good at any temperature and in the whole voltage range. Finally, the temperature dependency of both the large and the small gap obtained from this fitting procedure is reported in Figure 5(c). A comparison with the inset of Fig. 2 clearly shows that the separate fitting of the partial conductances allows a strong reduction of the error bars (evaluated from the fitting procedure) and a consequent improvement of the accuracy. In particular, the error affecting \( \Delta_\pi \) is very small even at \( T \) close to \( T_c \), so that the deviation of the gap values from the BCS-like curve (dashed line) results to be much larger than the experimental uncertainty.

In conclusion, we have shown that a technique which combines directional point-contact spectroscopy with the selective removal of the \( \pi \)-band gap by a magnetic field not only proves the existence of two gaps in MgB\(_2\), but also allows a very accurate test of the predictions of the two-band model. In particular, by fitting the zero-field conductance curves of directional point contacts, we obtained the weights of the \( \sigma \) and \( \pi \) bands, which resulted in good agreement with those predicted theoretically both for \( c \)-axis and \( ab \)-plane current injection. Then, we separately analyzed the partial conductances \( \sigma_\sigma \) and \( \sigma_\pi \), getting the most accurate values of the gaps in MgB\(_2\) obtained so far: at low \( T \), \( \Delta_\pi = 7.1 \pm 0.1 \) meV and \( \Delta_\sigma = 2.80 \pm 0.05 \) meV. We also found that, while \( \Delta_\pi \) follows a BCS-like temperature evolution, \( \Delta_\sigma \) deviates from the BCS behavior at \( T > 25 \) K, in very good agreement with the two-band model. Due to the small error affecting the gap value, this deviation is here unquestionably determined for the first time.

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