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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. Because of the directionality of the current injection into the samples, the application of a magnetic field allowed us to separate the contributions of the σ and π 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 a function of temperature allows a stricter test of the predictions of the two-band model for MgB_2.

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During the past year, a 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 σ bands arising from hybrid sp^2 orbitals in the boron planes, and 3D π bands that stem from the out-of-plane p_z orbitals [1,2]. The unusual consequence of this band structure is that two different energy gaps can be observed in the clean limit [2–4]: Δ_σ (the larger) and Δ_π (the smaller). Both gaps are expected to close at the same temperature T_c because of an interband pair-scattering mechanism [5] but, while $\Delta_\sigma(T)$ should approximately follow a BCS-like curve, a marked reduction of $\Delta_\pi(T)$ with respect to a BCS-like behavior is expected at $T \gtrsim 20$ K [3,4].

So far, one of the most convincing experimental supports of this model has been the observation of two gaps by tunneling [6] and point-contact spectroscopy [7] 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 σ and π bands to the total conductance and to fit them obtaining the temperature dependency of each gap with great accuracy. We 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 the Swiss Federal Institute of Technology (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 1 h of heating up to 1700–1800 °C, a plateau of 1–3 h, and a final cooling lasting 1–2 h. MgB_2 platelike crystals up to 200 μg 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 nonconventional technique that consists in using as a counterelectrode either a very small ($\lesssim 50$ μm) 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 [8], but the *effective* electrical contact occurs in a much smaller region, due to the presence of parallel microbridges in the spot area. On the other hand, the resistance of all our contacts was in the range 10–50 Ω. This, together with the estimated mean free path for the same crystals $\ell = 80$ nm [9], proves that our contacts are in the ballistic regime. The contacts were

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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 1(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 $V \simeq \pm 2.7$ mV and $V \simeq \pm 7.2$ mV, while the c-axis curves show only a peak at $V \simeq \pm(2.8 \div 3.5)$ mV and a smooth shoulder at $V \simeq \pm 7.2$ mV. These features, marked by dashed lines in the figure, are clearly related to the two gaps Δ_π and Δ_σ. Solid lines are the best-fitting curves calculated by using the Blonder-Tinkham-Klapwijk (BTK) model [10] generalized to the case of two bands, in which the normalized conductance σ is given by $\sigma = w_\pi \sigma_\pi + (1 - w_\pi)\sigma_\sigma$. Here, σ_π and σ_σ are the normalized conductances for the π and σ bands, respectively, and w_π is the weight of the π band, which 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 seven adjustable parameters: the gaps Δ_σ and Δ_π, the broadening parameters Γ_σ and Γ_π, the barrier height coefficients Z_σ and Z_π, plus the weight factor w_π. The normalization may yield additional uncertainty on $\Gamma_{\sigma,\pi}$ and $Z_{\sigma,\pi}$ 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 T_c of the junction that fell in all cases between 34.1 and 37.6 K. Since neither the current direction nor the contact resistance depends on the temperature, in fitting the conductances at various temperatures we kept both w_π and the barrier parameters Z_σ and Z_π 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 two ab-plane contacts (solid symbols) and of two c-axis contacts (open symbols). For clarity, error bars are shown only for a data set in the ab-plane-current case, but they are of the same order of magnitude in the c-axis cases. The relevant barrier parameters (independent of temperature) are $Z_\sigma = 0.5$–1.4 and $Z_\pi = 0.3$–0.8 depending on the junction, while the broadening parameters $\Gamma_{\sigma,\pi}$ increase with T always remaining in the range between 0.5 and 3 meV. An important result is that the average values of the π-band weight factor resulting from the fits ($w_\pi = 0.75 \pm 0.03$ for ab-plane current, and $w_\pi = 0.980 \pm 0.005$ for c-axis current) are in very good agreement with the values predicted by the two-band model ($w_\pi = 0.66$ and $w_\pi = 0.99$, 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 $w_\pi(\varphi)$, taking into account the angular variation of the electron injection probability, shows that our experimental values of w_π are compatible with cone apertures of about 26° and 60°, respectively.

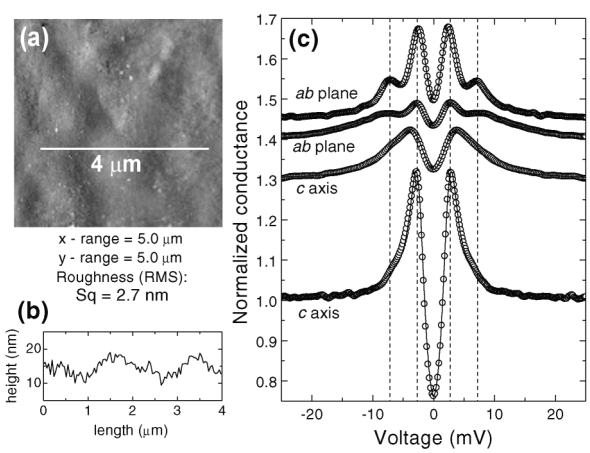

FIG. 1. (a) AFM image of the ab-plane crystal surface in the contact region, after removal of the In electrode. (b) Profile curve along the white line in (a). (c) Some *normalized* experimental conductance curves measured at low temperature (4.2–4.6 K) with ab-plane and c-axis current injections. The curves are vertically displaced for clarity and solid lines are the best-fitting curves (see text).

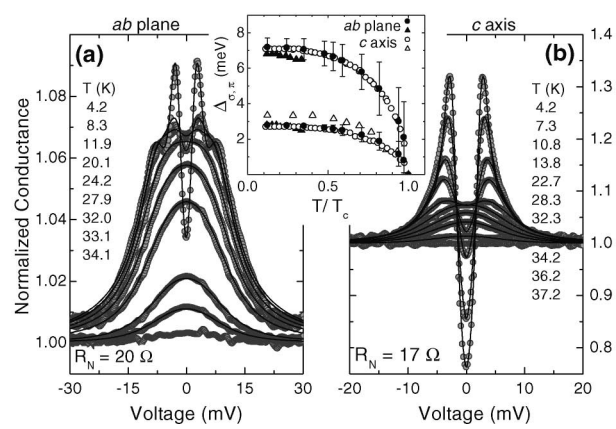

FIG. 2. Temperature dependency of the normalized conductance in a Ag-paste contact with current parallel to the ab planes (a) and in a In-spot contact with current along the c axis (b). Solid lines are the BTK best-fitting curves. Inset: temperature dependency of the gaps obtained from the fit of the conductance curves of various contacts.

The average low-temperature gap values $\Delta_\sigma = 7.1 \pm 0.5$ 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 some point-contact 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 c axis or to the ab planes, we observed in both cases the complete vanishing of the small-gap features in the conductance when $B \simeq 1$ T. This effect is clearly visible in Fig. 3, which shows the magnetic field dependency at 4.2 K of the conductance of ab-plane contacts in a field parallel to the c axis (a) and parallel to the ab plane (b). The crucial point here is to show that a field of 1 T is enough to remove superconductivity in the π band without modifying the conductance of the σ band up to a temperature close to T_c. Actually, the effect of the field on the large gap depends on the field direction. Figure 3(b) shows that, when $\mathbf{B} \parallel ab$ plane, the large-gap features remain clearly distinguishable up to 9 T, with only some marks of the gap closing. Instead, when $\mathbf{B} \parallel c$ axis (a), the peaks due to the large gap merge together at $B \geq 4$ 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 π band is quite isotropic and its critical field at 4.2 K is around 1 T, (ii) the σ band is anisotropic and, at 4.2 K, Δ_σ is unaffected by a field of 1 T parallel to the ab plane, and (iii) the σ-band critical field parallel to ab is rather high ($>$ 9 T) at low temperature, in agreement with other results on similar samples [11]. In addition to this, some preliminary measurements we made in c-axis contacts with $\mathbf{B} \parallel ab$ at about 30 K have shown that $\mathbf{B}^{\sigma}_{c2\|ab} \sim 3.5$ 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 [12]. Anyway, based on the present 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 In-MgB_2 c-axis contact at 4.6 K, with no field (see Fig. 4, open circles) and with a field of 1 T parallel to the ab planes (see Fig. 4, light gray circles). When the magnetic field destroys the gap in the π band, the *normalized* conductance becomes $\sigma(B = 1\ \mathrm{T}) = w_\pi + (1 - w_\pi)\sigma_\sigma$. This function contains only three free parameters: Δ_σ, Γ_σ, and Z_σ, whose best-fit values at $T = 4.6$ K are 7.1 meV, 1.7 meV, and 0.6, respectively. In fact, we took $w_\pi = 0.98$, which 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 field. The

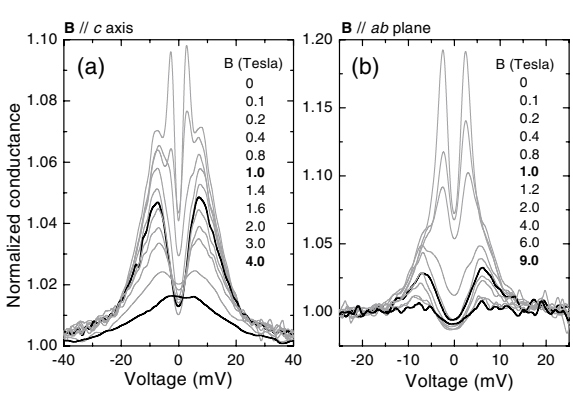

FIG. 3. (a) Some experimental normalized conductance curves of an ab-plane contact, in increasing magnetic fields parallel to the c axis. Thick lines represent the curves measured at $B = 1$ T and $B = 4$ T. (b) Same as in (a) but for an ab-plane contact with $\mathbf{B} \parallel ab$ plane. Thick lines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

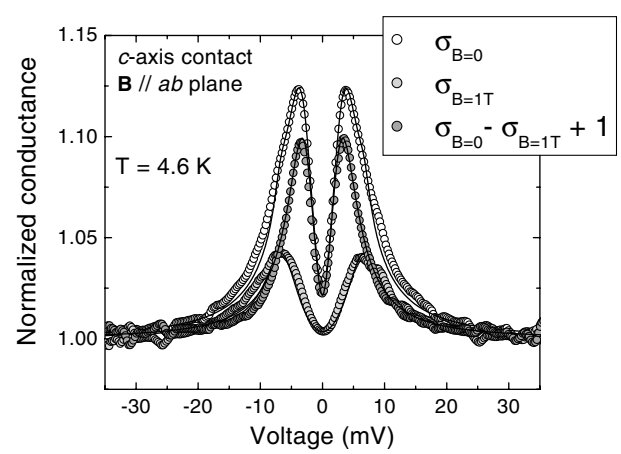

FIG. 4. Open circles: normalized conductance of a c-axis contact, with no magnetic field. Light gray circles: conductance of the same contact with a field of 1 T applied parallel to the ab plane. Dark gray circles: difference between the two previous curves (suitably shifted). Solid lines are the best-fit curves given by the appropriate BTK model (see text).

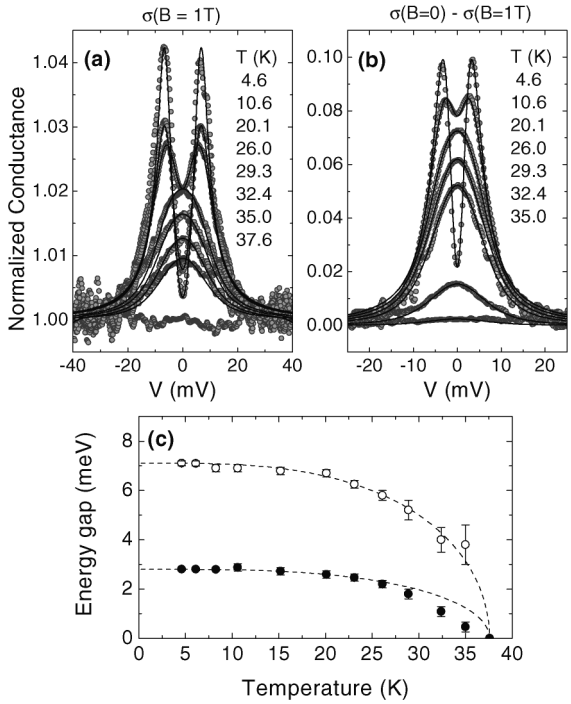

FIG. 5. (a) Temperature dependency of the normalized conductance of a c-axis junction, in a field $B_{\parallel ab} = 1$ T (symbols). (b) Temperature dependency of the difference between the conductance in zero field (see Fig. 4) and the conductance in a field of 1 T (previous panel). In both (a) and (b) solid lines are the BTK best-fitting curves with three parameters (see text). (c) $\Delta_\sigma(T)$ (open circles) and $\Delta_\pi(T)$ (solid circles) obtained from the fit of the curves in (a) and in (b), respectively. Dashed lines are the corresponding BCS-like curves.

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 $\sigma(B = 0) - \sigma(B = 1\ \mathrm{T}) = w_\pi(\sigma_\pi - 1)$. Fitting the experimental data to this function (again, with $w_\pi = 0.98$) allows determining the three remaining free parameters Δ_π, Γ_π, and Z_π, which 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_π is appropriate. Figure 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, $\sigma(B = 1\ \mathrm{T})$ (a) and the difference $\sigma(B = 0) - \sigma(B = 1\ \mathrm{T})$ (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 in both $\sigma(B = 0)$ and $\sigma(B = 1\ \mathrm{T})$. 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 Fig. 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 Δ_π 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 π-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 σ and π bands, which resulted in good agreement with those predicted theoretically both for c-axis and ab-plane current injections. Then, we separately analyzed the partial conductances σ_σ and σ_π, getting the most accurate values of the gaps in MgB_2 obtained so far: at low T, $\Delta_\sigma = 7.1 \pm 0.1$ meV and $\Delta_\pi = 2.80 \pm 0.05$ meV. We also found that, while Δ_σ follows a BCS-like temperature evolution, Δ_π deviates from the BCS behavior at $T > 25$ K, in very good agreement with the two-band model. Because of the small error affecting the gap value, this deviation is here unquestionably determined for the first time.

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[1] J. Kortus *et al.*, Phys. Rev. Lett. **86**, 4656 (2001); S.V. Shulga *et al.*, cond-mat/0103154; J.M. An and W.E. Pickett, Phys. Rev. Lett. **86**, 4366 (2001); Y. Kong, O.V. Dolgov, O. Jepsen, and O.K. Andersen, Phys. Rev. B **64**, 020501(R) (2001).
[2] H.J. Choi *et al.*, Nature (London) **418**, 758 (2002).
[3] A.Y. Liu, I.I. Mazin, and J. Kortus, Phys. Rev. Lett. **87**, 087005 (2001).
[4] A. Brinkman *et al.*, Phys. Rev. B **65**, 180517(R) (2002).
[5] H. Suhl, B.T. Matthias, and L.R. Walker, Phys. Rev. Lett. **3**, 552 (1959).
[6] F. Giubileo *et al.*, Phys. Rev. Lett. **87**, 177008 (2001); H. Schmidt *et al.*, Phys. Rev. Lett. **88**, 127002 (2002); M.H. Badr *et al.*, Phys. Rev. B **65**, 184516 (2002); M. Iavarone *et al.*, Phys. Rev. Lett. **89**, 187002 (2002).
[7] P. Szabó *et al.*, Phys. Rev. Lett. **87**, 137005 (2001); Yu. G. Naidyuk *et al.*, JETP Lett. **75**, 238 (2002); Y. Bugoslavsky *et al.*, Supercond. Sci. Tech. **15**, 526 (2002).
[8] A.M. Duif, A.G.M. Jansen, and P. Wyder, J. Phys. Condens. Matter **1**, 3157 (1989).
[9] A.V. Sologubenko *et al.*, Phys. Rev. B **66**, 014504 (2002).
[10] G.E. Blonder, M. Tinkham, and T.M. Klapwijk, Phys. Rev. B **25**, 4515 (1982).
[11] M. Angst *et al.*, Phys. Rev. Lett. **88**, 167004 (2002); A.V. Sologubenko *et al.*, Phys. Rev. B **65**, 180505(R) (2002).
[12] R.S. Gonnelli *et al.*, cond-mat/0209516.