## Anisotropic Magnetic Field Dependence of the Zero-Bias Anomaly on In-Plane Oriented [100] $Y_1Ba_2Cu_3O_{7-x}/In$ Tunnel Junctions

## R. Krupke\* and G. Deutscher

School of Physics and Astronomy, Raymond and Beverly Sackler Faculty of Exact Science, Tel Aviv University, 69978 Tel Aviv, Israel (Received 4 March 1999)

Tunneling spectroscopy was performed on in-plane oriented [100]  $Y_1Ba_2Cu_3O_{7-x}/In$  tunnel junctions. Splitting of the zero-bias conductance peak at zero magnetic field was observed below 6 K. The splitting increases initially at the rate of 1.3 mV/T when the field is applied along the [001] direction, and at a rate of 0.09 mV/T and 0.11 mV/T when applied parallel to the [010] and [100] directions, respectively. In the [001] direction, at 4.2 K, a sharp transition is seen at a field of about 0.8 T, from a rapid to a slow increase of the splitting. In that orientation, the splitting shows a large magnetic hysteresis.

PACS numbers: 74.50.+r, 74.72.Bk

Tunneling spectroscopy on  $Y_1Ba_2Cu_3O_{7-x}$  (YBCO) is a complex issue. The short coherence length of the material, its anisotropy, and its sensitivity to defects and impurities make it difficult to obtain reproducible conductance spectra, and to distinguish between intrinsic and extrinsic contributions to the tunneling conductance. Nevertheless, a robust feature, observed by many authors, mainly by planar tunneling along the [100], [110], or [103] directions, is the zero-bias conductance peak (ZBCP) [1-3]. Two possible origins of the ZBCP are the presence of magnetic impurities [4] in the vicinity of the barrier and d-wave pairing [5]. In the last case, for low transparency contacts, it is due to the formation of Andreev surface bound states at the [110] surface [6]. It has been shown that microscopic interface roughness gives rise to ZBCPs with similar spectral weight for both [110] and [100] planar tunnel junctions [7].

The subject of this paper is the occasionally observed splitting of the conductance peak, in zero magnetic field at low temperatures, and its field dependence [1-3]. Assuming that the ZBCP is due to Andreev bound states, Fogelström et al. [7] have shown that it should split in the presence of a magnetic field perpendicular to the CuO2 planes (parallel to the c axis). This splitting is due to the field induced in-plane screening currents, which shift the energy of the surface bound states by an amount of the order  $\mathbf{v}_S \cdot \mathbf{p}$ , where  $\mathbf{v}_S$  is the superfluid velocity and **p** is the momentum of the excitation. No such shift should occur when the field is applied parallel to the CuO<sub>2</sub> planes. We show here, for the first time, precisely this anisotropy of the field splitting. This rules out the possibility that, in our experiment, the ZBCP is primarily of magnetic origin.

The YBCO thin films were grown by planar sputtering, using a technique similar to that described by Fuchs *et al.* [8]. A  $Pr_1Ba_2Cu_3O_7$  template layer first deposited on tetragonal [100] LaSrGaO<sub>4</sub> substrates enables one to grow a-axis oriented YBCO films at growth conditions for c-axis orientation. X-ray diffraction analysis revealed a

pure a-axis (100) orientation of the multilayer with only a small amount of PrBaO<sub>4</sub> secondary phase. Because of the tetragonal structure of the substrate, in-plane orientation of the b axis [010] and the c axis [001] is achieved. As a result, the electrical anisotropy is large in terms of the absolute values of the sheet resistance  $R_{\square}$ , as well as of its temperature dependence (Fig. 1). In this respect, such YBCO films are similar to untwinned single crystals. The dimensions of the samples are  $7 \times 7 \text{ mm}^2$ . The thickness of the YBCO layer is estimated to be 3000 Å.

After the film growth, an indium (In) pad was pressed onto the YBCO surface. The YBCO/In interface area measures approximately 3 mm<sup>2</sup>. The sample was electrically wired using silver paint and hooked on a sample holder with the major crystallographic axis carefully aligned with respect to the magnetic field. In order to measure different magnetic field orientations, the sample had to go through several thermal cyclings, because the sample holder did not allow one to change the orientation at low temperature. To ensure reproducibility, each experiment was double-checked. It should be mentioned

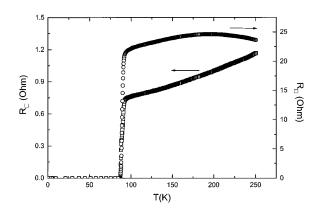


FIG. 1. Sheet resistance  $R_{\square}$  vs temperature T. The four-lead measurement was done in square geometry with the current driven along the c axis (circles) and along the b axis (squares).  $T_{C_0}=86~\mathrm{K}$ .

that the reproducibility concerning sample preparation and measurement was high. We therefore believe that by using YBCO films, with the same structural and electrical properties, our measurements should be easily reproduced.

The current-voltage characteristics of the YBCO/In junction were recorded by a four-point measurement using a current source and a voltmeter. At positive voltage, electrons are injected into the YBCO. The differential conductance dI/dV-V was calculated using a computer program.

The overall conductance spectrum is characterized by an asymmetric increasing background at high bias and a gap feature at about 17 meV (Fig. 2). A conductance peak is observed at zero bias with a conductance minimum about 6 meV. Below T=6 K, the conductance peak splits. The spontaneous zero magnetic field splitting  $\delta_S$  is in the range of 1–1.5 meV, depending on the specific sample and its history. This is similar to the zero field splitting seen by Geerk *et al.* [1], also on YBCO/In junctions.

We focus now on the conductance spectrum in the low energy range between  $V=\pm 8$  meV at T=4.2 K. In Fig. 3(a) the evolution of the conductance spectrum is followed as a function of the magnetic field B. The field B is aligned parallel to the c axis  $(B \parallel c)$ . As B increases, the zero-bias conductance (ZBC) diminishes, and the peak position  $\delta$  moves to higher energies. When reducing B, the ZBC increases and  $\delta$  is reduced, however hysteretically. In Fig. 3(b) the field evolution is shown for B parallel to the b axis  $(B \parallel b)$ . In that configuration the ZBC decreases and  $\delta$  increases with increasing B, but with weaker field dependence, and no hysteresis is observed. The field dependence for B parallel to the a axis  $(B \parallel a)$  (not shown) is qualitatively the same as for  $B \parallel b$ .

The detailed field dependence of  $\delta$  is shown in Fig. 4. We first describe the case  $B \parallel c$ , symbolized by solid

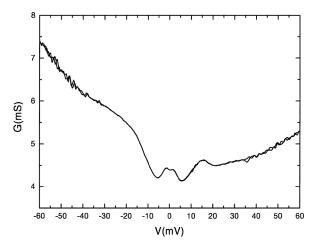


FIG. 2. Dynamical conductance taken at 4.2 K and zero magnetic field.

circles. For *B* increasing from 0 to 6 T, one can distinguish three regions: i) for  $B \le 0.1$  T, the peak position is field independent within the experimental error and  $\delta = \delta_S$ ; ii) for 0.1 < B < 0.8 T,  $\delta$  is strongly field dependent and increases with a slope of 1.3 mV/T; iii) for  $B \ge 0.8$  T,  $\delta$  is weakly field dependent and increases at a rate of 0.1 mV/T. For *B* decreasing from 6 to 0 T, roughly two regions can be distinguished: a) for 5.5 T  $< B \le 6$  T,  $\delta$  is strongly field dependent and decreases with an initial slope of 1.1 mV/T. b) for  $B \le 5.5$  T,  $\delta$  is weakly field dependent and changes with a slope of 0.14 mV/T. In the case of  $B \parallel b$ , symbolized by open squares, the hysteresis is absent, and  $\delta$  is weakly field dependent with a slope of 0.09 mV/T over the entire field range. The slope for the case  $B \parallel a$  is 0.11 mV/T.

In order to get more information about the hysteretic behavior in the case of  $B \parallel c$ , the magnetic field was cycled between different starting and end points. First, a field of 6 T was applied, which was then reduced to 3 T. Second, the field dependence was measured between 3 and 5 T, as depicted in Fig. 4 by open circles. Again, a strong field dependence shows up with an initial slope of 1.4 mV/T, which flattens above 3.5 T to 0.12 mV/T. Finally, the field was swept between 5 and 4.8 T. The field dependence in that range is strong, with a slope of 1.9 mV/T, and not hysteretic.

The field dependence can be summarized as follows. In the case of  $B \parallel c$ , there exist two distinct regimes of field sensitivity, which are characterized by a strong and weak field dependence of the ZBCP splitting, with slopes of  $\sim$ 1.3 and  $\sim$ 0.1 mV/T, respectively. If the magnetic field variation is small,  $\Delta H < \Delta H^*$ , the field dependence

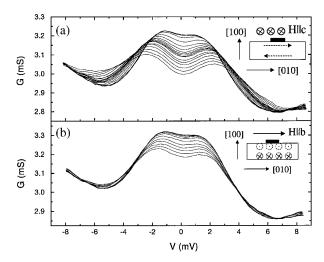


FIG. 3. Dynamical conductance G = dI/dV vs potential V. (a)  $B \parallel c$ , B = 0, 0.05, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, 1, 1.5, 2, 3, 4, 5, and 6 T. (b)  $B \parallel b$ , B = 0, 0.1, 0.2, 0.3, 0.4, 0.5, 1, 2, 3, 4, 5, and 6 T. The insets illustrate the a-b plane cross section of the sample (solid rectangle: In pad). Included are the field orientation and the screening currents (dashed lines).

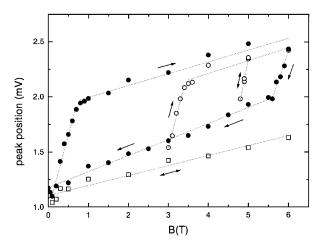


FIG. 4. Peak position  $\delta$  vs magnetic field B. Open and solid circles:  $B \parallel c$ ; open squares:  $B \parallel b$ . The arrows indicate the sweep direction of the magnetic field as explained in the text.

is strong and the ZBCP splitting is reversible and nonhysteretic. If  $\Delta H > \Delta H^*$ , the splitting successively follows two slopes, and is hysteretic. In the cases of  $B \parallel b$  and  $B \parallel a$ , the field dependence is nonhysteretic and weak with slopes of  $\sim 0.09$  and  $\sim 0.11$  mV/T, respectively.

The field dependence  $\delta(H)$  that we observe for  $H \parallel c$  is similar to that reported by Covington  $et\ al.$  [2]. However, we note some difference. First, Covington  $et\ al.$  [2] did not observe any anisotropy of  $\delta(H)$ , maybe because their films were not in-plane oriented. Second, our more detailed data show a sharp change in slope at 0.8 T, which does not quite fit the theoretical predictions of Fogelström  $et\ al.$  [7]. Third, we definitely observe a finite slope  $(d\delta/dH)$  at large field, while Covington  $et\ al.$  fitted their data to a theory predicting a saturation of  $\delta$  at strong fields.

We now propose an interpretation of our observed field dependence. First, we make use of the ZBCP splitting theory of Fogelström *et al.* [7] to explain the large initial slope in the case  $B \parallel c$ . We will then explain the hysteretic behavior in the framework of the Bean-Livingston (BL) barrier of type-II superconductors [9]. Furthermore, we will argue that the small slope comes from the Pauli-field contribution.

As mentioned above, a d-wave symmetry of the order parameter leads intrinsically to pair breaking at the [110] surface and gives rise to surface bound states which are measured by tunneling as ZBCP [6]. The field splitting of the ZBCP can be explained by electromagnetic coupling of these surface bound states to the magnetic field via the screening currents in the superconductor. Excitations, which have a momentum component parallel or coparallel to the superflow, are shifted to higher or lower energies, respectively. Therefore the current shifts the Andreev bound states spectrum. Those excitations, which are Andreev reflected at glancing angle, experience the largest energy shift. The maximum energy shift,  $\varepsilon^{\text{max}}$ , should be

of the order of the gap  $\Delta \approx 17$  meV. It is limited by the surface currents, reaching the pair breaking current at the thermodynamical critical field  $H_C = \phi_0/\sqrt{8 \pi \lambda_0 \xi_0}$ . Excitations, which are Andreev reflected at a larger angle, are shifted by  $\varepsilon_p = \Delta(H/H_C) \sin \phi_p$ , where with  $\phi_p$  is the angle between incidence and [110] surface normal. Which excitations can be detected by tunneling depend on the barrier thickness. If the barrier is thin, the tunneling cone is large, and excitations with a large energy shift can be measured. However, if the barrier is thick, as is the case here, and the tunneling cone is small, only excitations with a small energy shift can be detected. It follows that the maximum shift observable in tunneling is  $\delta(H) = \Delta(H/H_C) \sin \phi_C$ , where  $\phi_C$  is the angle of the acceptance cone measured from the [110] surface normal. From that argumentation, the field splitting due to surface currents has a slope  $(\Delta/H_C)\sin\phi_C$ , which reproduces the expression of Fogelström et al. (see page 284 in Ref. [7]) in the low field limit. With  $H_C \approx 0.8$  T and  $\Delta \approx 17 \text{ meV}, \, \phi_C \text{ has to be equal to } 3.5^{\circ} \text{ in order to fit}$ the value of the large slope of the field splitting. These values are consistent with those used by Covington et al. [2] to fit their low field data. It should be emphasized that an important condition to observe the splitting is that the tunnel junction has a low transmission. Only then is the surface barrier strong and are the Andreev bound states well defined. A small value for  $\phi_C$  is therefore expected. We want to mention that we also prepared junctions with Ag counterelectrodes, which reproducibly showed a ZBCP, however, without zero field splitting.

A very strong argument in favor of the described scenario being at the origin of the large field splitting is the fact that the field splitting is anisotropic with respect to the field orientation. It should be recalled that the sample is in-plane oriented and that all [110] facets, which are present due to surface roughness, are parallel to the [001] direction. Excitations are shifted only to higher energies, if surface currents are flowing in the a-b plane. For currents flowing perpendicular to the a-b plane, no energy shift occurs since the additional momentum, given by the supercurrent, is then perpendicular to the momentum of the Andreev reflected excitations. Indeed, a strong field dependence is observed only when  $B \parallel c$ , not when  $B \parallel b$ . In the case  $B \parallel a$ , where the field is perpendicular to the sample surface and the supercurrents are flowing at the sample edges, the field dependence is also weak.

The field splitting crosses over from a strong field dependence to a weak field dependence, if the field variation is larger than  $\Delta H^* \sim 0.8$  T. This is of the order of the BL barrier field, observed in  $Y_1Ba_2Cu_4O_8$  [10]. Once the vortices can enter the superconductor, a further field increase does not modify the surface current and a further splitting of the ZBCP does not occur. In decreasing fields, vortices do not exit if the field is reduced by less than  $\Delta H^*$ . In that range the surface current decreases, and so does the splitting. What is, however, somewhat

surprising is the sharp transition from the low field to the high field behavior. Another observation that does not fit the predictions of Fogelström et al. is that the field splitting does not saturate at high field, but rather shows a weak field dependence with a slope of approximately 0.1 meV/T. According to Fogelström et al. [7], a saturation should occur at  $H \sim 3H_C$ . Instead, the slope is close to  $\delta(H)/H = g\mu_B = 0.1158 \text{ mV/T}$ , predicted for the Zeeman splitting of electrons with  $g = g_{\text{electron}} = 2$ . We therefore conclude that the weak field dependence seen at strong applied fields is caused mainly by Zeeman splitting. This statement is reinforced by the fact that similar slopes are measured with  $B \parallel a$  and  $B \parallel b$ , where no strong field dependence is observed and field splitting due to supercurrents does not occur, and is not expected. We want to point out that the high field data of Cu/YBCO and Pb/YBCO junctions [2] can be linearly fitted with a similar slope, which is consistent with our interpretation of a dominant Zeeman splitting.

We now turn to the splitting of the ZBCP in zero field. If the very same physics, which determines the strong field dependence, is causing the zero field splitting, then the conclusion must be that spontaneous surface currents do exist at zero applied magnetic field (spontaneous time reversal symmetry breaking). The value of the spontaneous currents can be estimated from the value of the zero field splitting  $\delta_S = 1.1$  meV and the large slope of the field dependence 1.3 meV/T. The implication therefore is that the spontaneous surface currents are of the order of the pair breaking critical current. However, we note that the value of  $\delta_S$ , and therefore that of the corresponding spontaneous currents, depends on the type of junction studied. In the junctions used by Covington et al. [2],  $\delta_S$  appears only below 4.2 K, while in the case of our YBCO/In junctions, it already appears below 6 K. In the case of YBCO/Ag contacts it is not observed down to 1.5 K.

One parameter that varies with that kind of contact is the level of oxygen doping [11]. If the zero field splitting is attributed to a subdominant s-wave or  $d_{xy}$ -wave order

parameter, it must be concluded that its strength is doping dependent. This has not been predicted theoretically.

In conclusion, a strongly field dependent splitting of the ZBCP has been observed in in-plane aligned a-axis YBCO films, only when the field is applied perpendicular to the CuO<sub>2</sub> planes. This observation is consistent with a  $(d_{x^2-y^2}+is)$  surface order parameter, with the s component being enhanced by the Doppler shift of the Andreev bound state energy. But it may also be due to a  $(d_{x^2-y^2}+id_{xy})$  bulk order parameter [12–14]. Further work is required to clarify this point.

This research was supported in part by the Heinrich Hertz Minerva-Center for High Temperature Superconductivity and the Oren Family Chair of Experimental Solid State Physics. The authors are thankful to C. C. Tsuei for helpful remarks concerning the possibility of  $d_{x^2-y^2}+id_{xy}$  ordering, and Y. Dagan, E. Farber, and A. Kohen for useful discussions.

- \*Email address: krupke@post.tau.ac.il
- [1] J. Geerk, X. X. Xi, and G. Linker, Z. Phys. 73, 329 (1988).
- [2] M. Covington et al., Phys. Rev. Lett. 79, 277 (1997).
- [3] J. Lesueur, L.H. Greene, W.L. Feldmann, and A. Inam, Physica (Amsterdam) 191C, 325 (1992).
- [4] J. A. Appelbaum, Phys. Rev. **154**, 633 (1967).
- [5] Y. Tanaka and S. Kashiwaya, Phys. Rev. Lett. 74, 3451 (1995).
- [6] C.-R. Hu, Phys. Rev. Lett. 72, 1526 (1994).
- [7] M. Fogelström, D. Rainer, and J. A. Sauls, Phys. Rev. Lett. 79, 281 (1997).
- [8] D. Fuchs et al., Physica (Amsterdam) 280C, 167 (1997).
- [9] C. P. Bean et al., Phys. Rev. Lett. 12, 14 (1964).
- [10] M. Xu, Phys. Rev. B 48, 10630 (1993).
- [11] D. Racah and G. Deutscher, Physica (Amsterdam) 263C, 218 (1996).
- [12] K. Krishana et al., Science 277, 83 (1997).
- [13] R. B. Laughlin, Phys. Rev. Lett. 80, 5188 (1998).
- [14] M. Franz and Z. Tešanović, Phys. Rev. Lett. 80, 4763 (1998).