

## Angular dependence of the critical current in thin $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ films with unidirectional nanocracks

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We investigated the angular dependence of the critical current  $I_c$  in the  $a$ - $b$  plane of thin  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  films on (001)  $\text{NdGaO}_3$ . Films grown on this particular substrate relieve their stress by developing unidirectional cracks of 2–4 nm width. These unidirectional nanocracks (NC's) behave as Josephson junctions. We have measured  $I_c(\theta_{NC})$ , where  $\theta_{NC}$  is the angle to these nanocracks, in a set of microbridges that were patterned on the film at various angles  $\theta_{NC}$ . We fitted the results to a model of Andreev reflections in an anisotropic superconductor given by Tanaka and Kashiwaya [Phys. Rev. B **56**, 892 (1997)]. We found that an order parameter having a  $d+is$  symmetry fits our data best at high temperatures, while no satisfactory fit could be obtained at low temperatures using a pure  $d$ -wave,  $s$ -wave, or  $d+is$  order parameter.

Epitaxial  $c$ -axis oriented  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO) films on (100)  $\text{SrTiO}_3$  release their strain in the tetragonal to orthorhombic phase transition through bidirectional twinning along the (110) and  $(1\bar{1}0)$  directions. Recently, we found that the same type of film on (001)  $\text{NdGaO}_3$  (NDG) substrates grows unidirectionally twinned.<sup>1</sup> To release the strain in the other in-plane direction, these films develop a unidirectional array of parallel cracks, perpendicular to the twin planes. Transmission electron microscope (TEM) images reveal that the width of these cracks is only 2–4 nm, hence they can be termed nanocracks (NC's). These NC are the source of a strong in-plane anisotropy in the transport properties of the films,<sup>2</sup> such as the resistivity  $\rho$  and the critical current  $I_c$ . Along the NC (normal to the twin planes), these films have  $\rho$  and  $I_c$  values similar to those of YBCO films grown on the commonly used substrates such as  $\text{SrTiO}_3$ ,  $\text{LaAlO}_3$ , and  $\text{MgO}$ . However, along the direction normal to the NC, the values of  $\rho$  increase and the values of  $I_c$  decrease by about one and two orders of magnitude, respectively. The transport anisotropy therefore originates in the NC. Below  $T_c$ , the  $I$ - $V$  curves of the microbridges that cross these NC are characteristic of Josephson junctions of the weak-link type.<sup>2</sup> This seems to indicate that the vacuum gaps of the NC are probably shunted by a series of microshorts that govern the high transparency of the junctions. The angular dependence of the critical current in these films,  $I_c(\theta)$ , was measured at different temperatures in microbridges that were patterned at angles  $\theta_{NC}$  relative to the NC orientation. Initially, the results were interpreted in terms of a phenomenological model which is based on the fact that  $I_c$  of a junction is proportional to the Josephson coupling energy. Thus near  $T_c$  one has  $I_c(\theta) \propto \Delta_R(\theta)\Delta_L(\theta)A(\theta)$ , where  $\Delta_R(\theta)$  and  $\Delta_L(\theta)$  are the order parameters on the right- and left-hand sides of the NC junction, and  $A(\theta)$  is the cross-sectional area of the junction. In our analysis, we assumed that the quasiparticles cross the junctions parallel to the original direction of the current in the microbridges. Fits of our previous data to this

model at temperatures near  $T_c$  showed that the best fit is obtained when the order parameter has a  $d+is$ -wave symmetry.<sup>2</sup> We note here that the  $d$ -wave order parameter used in that paper had its positive  $d$  lobe aligned along the (110) NC direction ( $d_{xy}$ ), and not along the  $a$  or  $b$  axes ( $d_{x^2-y^2}$ ) as is commonly used. Although this simple model yielded very good fits, it was clear that a more quantitative approach based on a more detailed theoretical model is needed. In the present study, we extended the measurements of  $I_c(\theta)$  to lower temperatures, and used a more detailed model for the calculation of  $I_c(\theta)$ .<sup>3</sup> In our analysis of the data we keep the assumption that quasiparticles incident at the junction cross it parallel to the bridge. The model calculation fits the experimental results near  $T_c$  well with a  $d_{x^2-y^2}+is$  symmetry of the order parameter. At lower temperatures, however, no satisfactory fit to the data could be obtained.

Details of the films growth and patterning were given elsewhere;<sup>2</sup> here we shall mention briefly the main points. Epitaxial  $c$ -axis oriented YBCO films of various thicknesses ( $100 < d < 250$  nm) were deposited by dc sputtering onto (001) NDG substrates of  $10 \times 10$  mm<sup>2</sup> area. The films were patterned by deep UV photolithography into nine microbridges using a polymethyl methacrylate (PMMA) resist and ion-beam milling. The microbridges were 10  $\mu\text{m}$  wide, 100  $\mu\text{m}$  long, and at angles  $\theta_{NC}$  of  $-90^\circ$ ,  $-79^\circ$ ,  $-67^\circ$ ,  $-56^\circ$ ,  $-45^\circ$ ,  $-22^\circ$ ,  $0^\circ$ ,  $22^\circ$ , and  $45^\circ$  relative to the NC direction (see the inset to Fig. 1).  $\rho$  of the various bridges versus temperature is shown in Fig. 1 for a 200 nm thick film of YBCO on NDG. The maximum anisotropy ratio of the resistivities  $\rho(-90^\circ)/\rho(0^\circ)$  is about 10, similar to the value obtained before.<sup>2</sup> Representative  $I$ - $V$  curves of the various microbridges at angles  $\theta_{NC}$  and at 87 K are shown in Fig. 2. Since the NC offer little or no resistance to flux motion, the  $I$ - $V$  curves of all the microbridges appear rounded, with seemingly no critical current as seen in this figure. However, extrapolation of these curves to  $V=0$  from large bias shows that critical currents are definitely present. The  $I$ - $V$  curves

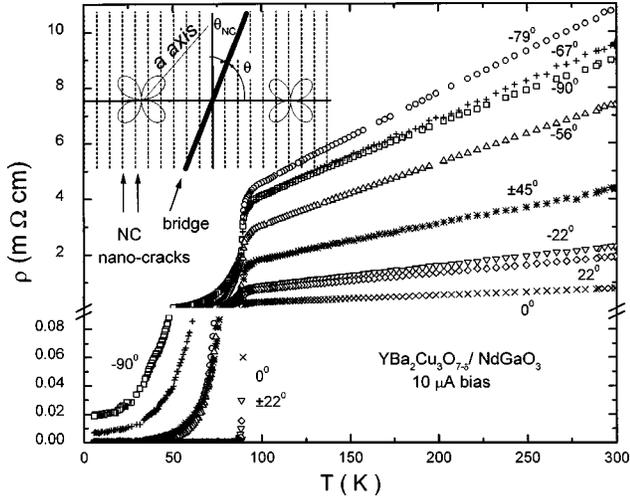


FIG. 1.  $\rho(T)$  of nine microbridges in an YBCO film on (001) NdGaO<sub>3</sub> at different angles  $\theta_{NC}$  to the NC orientation. The schematic drawing in the inset shows the relative orientations of a microbridge, the angles  $\theta$  and  $\theta_{NC}$ , the interfaces (NC), and the  $d_{x^2-y^2}$  symmetry.

appear rounded due to fluctuations of flux lines (noise), brought about by the total absence of pinning in the NC. In the presence of fluctuations, the correct procedure to determine  $I_c$  is to fit the  $I$ - $V$  curves to the Ambegaokar-Halperin model.<sup>2,4</sup> This model was used by Falco *et al.* to study the effect of thermal noise and extract the critical current  $I_c$  from the seemingly resistive  $I$ - $V$  data of their Josephson junctions.<sup>5</sup> This group also discussed the technique of using the Ambegaokar-Halperin model for extracting the junction parameters in great detail. Later on, Tinkham applied this method to the high-temperature superconductors to investigate the resistive transition below  $T_c$  in the flux flow regime,<sup>6</sup> and Gross *et al.* actually calculated the  $I$ - $V$  curves of grain-boundary junctions under thermally activated phase slippage conditions.<sup>7</sup> Figure 2 shows a typical result of a calculated  $I$ - $V$  curve of the  $\theta_{NC}=45^\circ$  bridge using the Ambegaokar-Halperin model. Clearly, the calculated curve fits the experimental data well, and similar quality fits were

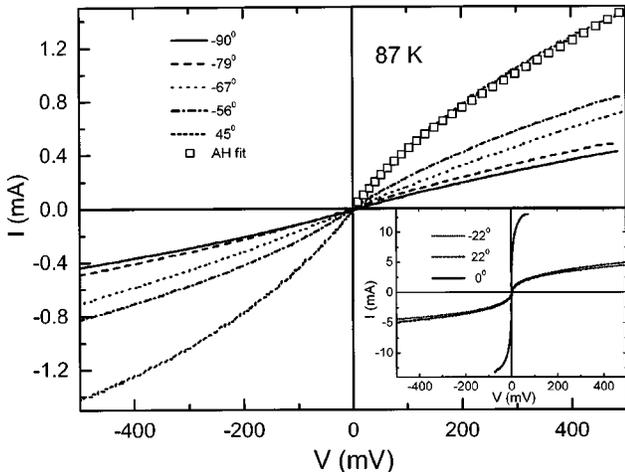


FIG. 2.  $I$ - $V$  curves of the various bridges with different  $\theta_{NC}$  angles. The open squares are calculated results using the Ambegaokar-Halperin model.

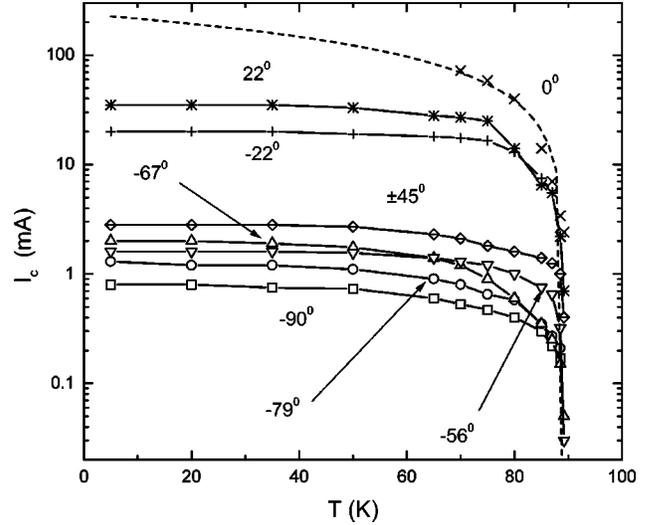


FIG. 3.  $I_c(T)$  of the microbridges in Fig. 1. The curves connect the experimental data points, except for the dashed one which is a power-law fit to the data of the  $0^\circ$  bridge.

obtained for all the other bridges.  $I_c$  was determined from the fit parameters using the detailed technique given by Falco *et al.*,<sup>5</sup> for all the bridges at any given temperature, and the results are shown in Fig. 3. In the  $\theta_{NC}=0^\circ$  bridge, the only bridge not cut by the NC, the critical current was determined by simple extrapolation of the  $I$ - $V$  curves from  $V \sim 1$  mV to  $V=0$ . In this case, it was not possible to reach much higher bias voltages on the microbridge without destroying it. Figure 3 shows that the anisotropy ratio of the critical currents  $I_c(0^\circ)/I_c(-90^\circ)$  is about 100 which is an order of magnitude higher than the corresponding inverted ratio of the resistivities.

Figures 4 and 5 show a typical angular dependence of  $I_c(\theta_{NC})$  deduced from the fits to the Ambegaokar-Halperin model, for the data taken from Fig. 3. Figure 4 shows the data near  $T_c$  at 87 K, and Fig. 5 shows the data at a low temperature of 5 K. To produce the theoretical curves for this data, we used a model by Tanaka and Kashiwaya (TK),<sup>3</sup>

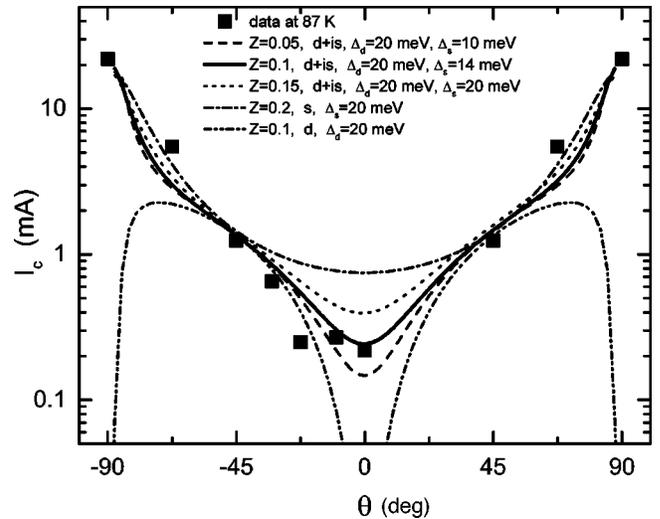


FIG. 4.  $I_c$  versus  $\theta$  at 87 K. The curves are fits of the data to Eqs. (1) and (3).

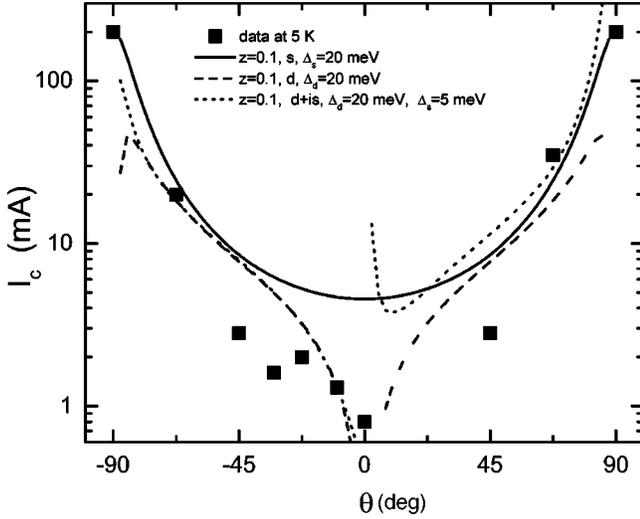


FIG. 5.  $I_c$  versus  $\theta$  at 5 K. The curves are fits of the data to Eqs. (1) and (3).

describing the critical current in junctions with  $d$ -wave superconductors. They assumed a perfectly smooth interface, and calculated the transport of quasiparticles through it by Andreev reflection and transmission processes, using the Bogolubov–de Gennes equations. They defined the angles between the normal to the interface and the crystallographic  $a$  axis on both sides of the junction as  $\alpha$  and  $\beta$ , and calculated the transport for any angle  $\theta$  between the direction of the impinging quasiparticles and the normal to the interface ( $\theta \equiv 90^\circ - \theta_{NC}$ , see the inset to Fig. 1). Then, in the calculation of  $I_c$ , they integrated over the angles  $\theta$ . For the present experiment we argue that the integration over  $\theta$  is not needed for the following reasons: For a low transparency junction with a high potential barrier (high  $Z$ ), the main contribution to  $I_c$  comes from angles around  $\theta \approx 0^\circ$  due to the exponential dependence of the tunneling on the barrier thickness, which leads to a perpendicular crossing of the barrier along the shortest route. For a high transparency barrier (low  $Z$ ), all incidence angles  $\theta$  contribute to  $I_c$  almost equally, if the superconductor is isotropic. Clearly, in the limit  $Z \rightarrow 0$  there is no change in the original direction of the current in the bridge near the junction. For these low  $Z$  junctions, we propose a scenario in which a very nearly uninterrupted crossing of the barrier occurs (parallel to the bridge). In this scenario, transport across the NC takes place via a set of nanoshorts which behave as pinhole junctions. This picture comes about from analogy to the situation which is believed to exist in grain-boundary junctions, where conduction occurs via a series of contacts, defects, or localized states in the barrier. In view of this picture, we shall model the transport of current across the NC, as analogous to the transmission of an optical beam through a diffraction grating. This means that we also assume almost equally spaced point contacts in the NC. The superconducting order parameter  $\psi$  is analogous to the electric field  $E$ , and the current ( $\propto \psi^* \nabla \psi$ ) to the light intensity ( $\propto E^2$ ). In the optical case, the zero-order diffraction peak of a beam passing through a diffraction grating is always in the same direction as that of the original beam and contains most of its intensity. Thus, by analogy, the current along the microbridge is diffracted by the pinholes grating mostly into the zero order which is also

in the direction of the bridge. This statement is equivalent to assuming *a priori* that the flux of quasiparticles is parallel to the microbridge. From the unidirectional twinning normal to the NC we estimate that the density of point contacts along a single NC is about equal to the twin density which is of the order of 30 per  $\mu\text{m}^2$ . Thus, if  $n_p$  is the total number of pinholes along a single NC, the analogy to the optical case also yields  $I_c \propto n_p^2$  ( $n_p$  large), which is the same as the intensity of the zero-order diffraction peak. Thus, selecting only the quasiparticles moving along the microbridge as those crossing the NC, allows us to use Eq. (57) of Ref. 3 for the calculation of  $I_c(\theta)$  without the integration step over  $\theta$ . This yields:

$$R_N I(\phi) = \frac{\pi \bar{R}_N k_B T}{e} \left\{ \sum_{\omega_n} \bar{F}(\theta, \omega_n, \phi) \sigma_N(\theta) \cos \theta \right\}, \quad (1)$$

where  $R_N$  is the normal resistance,  $I(\phi)$  is the supercurrent as a function of the phase difference  $\phi$  on the junction,  $\sigma_N$  is the conductivity in the normal state,  $\omega_n$  are the Matsubara frequencies, and the functions  $\bar{F}$  and  $\bar{R}_N$  are defined in Ref. 3.  $I_c$  is obviously the maximum of  $I(\phi)$  and for simplicity we take the normal conductivity as

$$\sigma_N(\theta) = \frac{\cos^2 \theta}{\cos^2 \theta + Z^2} \quad (2)$$

as used in Ref. 8. We note that since the unidirectional NC are formed in elongated twins *perpendicularly* to the (110) orientation of the twin planes, we have  $\alpha = \beta = 45^\circ$ . Because we do not integrate over  $\theta$ , the nodes of a pure  $d$ -wave order parameter are reflected in the calculations of  $I_c(\theta)$  as zeros at  $\theta = 0^\circ$  and  $\theta = \pm 90^\circ$ . An admixture of an imaginary  $s$ -wave component ( $is$ ) in the order parameter will prevent this situation at  $\theta = 0^\circ$ , but not at  $\theta = \pm 90^\circ$  due to the  $\sigma_N(\theta) \cos \theta$  terms in Eq. (1). Two more problems in the application of the TK model<sup>3</sup> are the assumption of a smooth and flat interface which is hard to realize in a real junction, and the breaking of pairs at the interface which changes the order parameter compared to its bulk value. The effect of pair breaking on  $I_c$  was recently calculated in a self-consistent way which showed a leveling off of  $I_c$  at low temperatures instead of the upturn found in the earlier analysis.<sup>3,8</sup> Nevertheless, we decided to use the analytical formula of the TK model [Eq. (1)] rather than the self-consistent model,<sup>8</sup> because of its relative simplicity. This should yield significant results at least at temperatures near  $T_c$  where the two models give similar results.<sup>3,8</sup>

In the calculations, we used a  $d+is$  order parameter having a maximum  $d$ -gap energy of  $\Delta_d = 20$  meV and  $\Delta_s$  gap values between 0 and 20 meV. For comparison, the calculations were done also for the case of a pure  $s$ -wave order parameter. The calculation of Eq. (1) yields the  $i_c r_N$  of a *single pinhole junction*. To apply this result to our *multijunction* system, we present a plausible picture for the structure of these junctions on a nanometer scale. From the atomic force microscope (AFM) and TEM micrographs of our YBCO films with nanocracks,<sup>2</sup> the fine details of the NC interfaces could not be resolved. This is due to the fact that the AFM measures only the surface morphology, and the required TEM resolution necessitates even thinner samples

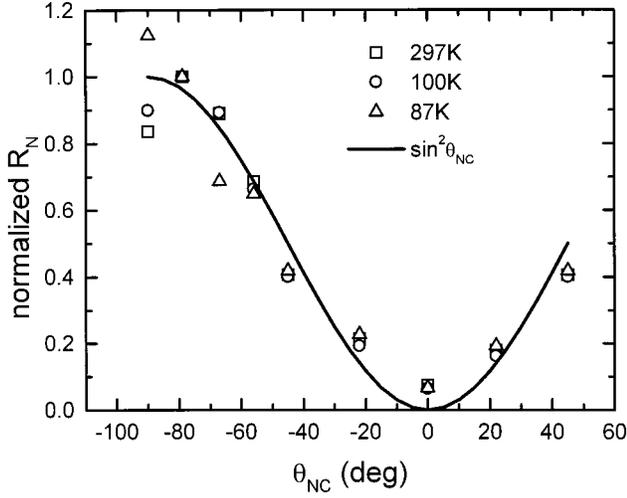


FIG. 6. Normal resistance  $R_N(\theta_{NC})$  at 87, 100, and 297 K, normalized to their values at  $\theta_{NC} = -79^\circ$ . The solid line is  $\sin^2(\theta_{NC})$ .

in which the original interfaces of the NC would be changed during the thinning process. We are thus left with less direct evidence as for the nanoscale structure of the NC interfaces and the transport of quasiparticles through them. Microbridges that were exposed to excessively high currents showed a typical catastrophic burn pattern which always appeared along a *single* NC as a straight transparent line of about  $0.5 \mu\text{m}$  width. This indicates that a *single* NC in a long microbridge with many NC is the weakest link that determines the critical current of the whole bridge. The quite uniform burn pattern suggests that conduction across the NC occurs via a set of nanoshorts or pinhole junctions that are spread almost evenly along the NC. An uneven distribution of these pin holes would have yielded a nonuniform burn pattern and this does not occur in reality. As was explained before, we use an optical picture to describe the transport of current in the microbridges. This current flows through a set of pinholes in the NC that cross each microbridge. Thus scattering of the current mostly into the zeroth-order diffraction yields  $I_c(\theta_{NC}) \propto I_{0c} n_p^2$ , where  $I_c(\theta_{NC})$  is the measured critical current,  $I_{0c}$  is the supercurrent at the NC plane, and  $n_p$  is the number of pinholes in a NC. Since  $I_{0c} = n_p i_c(\theta_{NC})$  where  $i_c$  is the supercurrent in a single pinhole junction, and since  $n_p \propto 1/\sin \theta_{NC}$ , we find that  $I_c(\theta_{NC}) \propto n_p i_c(\theta_{NC}) n_p^2 \propto i_c(\theta_{NC})/\sin^3 \theta_{NC}$ . As was mentioned above, Eq. (1) yields a calculated  $i_c(\theta)r_N(\theta)$  value of a *single* pinhole junction. To find the angular dependence of  $r_N(\theta)$ , we note that the normal resistance  $R_N(\theta_{NC})$  of a *whole* bridge is obtained by summing up the NC resistances in series and the pinhole resistances  $r_N(\theta)$  in parallel. This yields  $R_N(\theta_{NC}) = N(\theta_{NC})r_N(\theta_{NC})/n_p(\theta_{NC})$ , where  $N(\theta_{NC}) \propto \sin(\theta_{NC})$  is the number of NC in the bridge. Thus,  $R_N(\theta_{NC}) \propto \sin^2(\theta_{NC})r_N(\theta_{NC})$ . This result is compared to the data shown in Fig. 6 where we plot the *measured*  $R_N(\theta_{NC})$  at 87, 100, and 297 K normalized to their values at  $\theta_{NC} = -79^\circ$ , together with  $\sin^2(\theta_{NC})$ . In the superconducting state at 87 K,

the normal resistance values were obtained directly from the linear parts of the  $I$ - $V$  curves which were reached at about 300–500 mV. In the normal state at 100 K and 297 K the  $R_N(\theta_{NC})$  values were obtained from Fig. 1. One can see from Fig. 6 that  $R_N(\theta_{NC})$  is very nearly proportional to  $\sin^2(\theta_{NC})$ . We are thus led to the conclusion that  $r_N(\theta_{NC}) = r_N = \text{const}$ . This result may suggest that  $r_N$  originates in the constant quantum resistance  $\hbar/e^2$ , and the values that we measure of a few  $\text{k}\Omega$  seem to support this interpretation. Thus, using  $\theta = 90^\circ - \theta_{NC}$ , we obtain

$$I_c(\theta_{NC}) = \frac{i_c(\theta)r_N}{\sin^3 \theta_{NC} r_N}. \quad (3)$$

Equation (3) allows us to compare the measured  $I_c(\theta_{NC})$  of Figs. 4 and 5 with the calculated  $i_c(\theta)r_N$  according to Eq. (1).

The curves in Figs. 4 and 5 are the calculated angular dependences of  $I_c(\theta)$  using Eqs. (1) and (3). At 87 K, we see that the calculation that fits our data best is obtained when  $Z$  is low (0.1) and the order parameter has a  $d+is$  symmetry with  $\Delta_d = 20 \text{ meV}$  and  $\Delta_s = 14 \text{ meV}$ . We note here that mixing of a  $d_{xy}$  wave instead of the  $s$  wave in this fit also yields a good fit to our data. The pure  $s$ -wave symmetry does not fit the data around  $\theta = 0^\circ$ , while the pure  $d$ -wave misfits both around  $\theta = 0^\circ$  and around  $\theta = \pm 90^\circ$ . At 5 K, we could not find any satisfactory fit to our data using the present model (see the curves in Fig. 5). This seems to result from the fact that the present model,<sup>3</sup> even with a small  $s$ -wave component, develops an asymmetry in  $\pm \theta$  that prevents a good fit near  $\theta = 0^\circ$ . Qualitatively though, the  $d+is$  fit in Fig. 5 seems to indicate the presence of a smaller  $s$ -wave component. Theoretically, the fact that no good fits could be obtained is consistent with the previous finding of significant deviations of  $I_c$  at low temperatures, from the prediction of the present model.<sup>3,8</sup> These deviations of  $I_c$  were found when the order parameter was allowed to change self-consistently near the interface of the junction because of pair breaking.<sup>8</sup> Further theoretical work is needed in order to have better fits to our data at low temperatures.

In summary, the in-plane angular dependence of the critical current in unidirectionally cracked thin films of YBCO was both measured and compared with a model calculation. The NC were assumed to consist of a series of pinhole junctions in which the transport of current was described in terms of an optical model. The calculated dependencies of  $I_c(\theta)$  reproduced the experimental data only at temperatures near  $T_c$ , and good fits were obtained with an order parameter having a  $d+is$  symmetry, with  $\Delta_d = 20 \text{ meV}$  and  $\Delta_s = 14 \text{ meV}$ . At low temperatures, the theory will have to be improved by allowing for pair breaking at the interface of the junctions, which changes the order parameter from its bulk symmetry (pure  $d$  wave?).

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