Distribution of superconducting energy gaps in $GdBa_2Cu_3O_7$ obtained from point-contact spectroscopy

A. I. Akimenko,* G. Goll, and H. v. Löhneysen
Physikalisches Institut, Universität Karlsruhe, D-7500 Karlsruhe, Federal Republic of Germany

V. A. Gudimenko
Institute for Low Temperature Physics and Engineering, Academy of Sciences of the Ukrainian SSR, Kharkov, 310164, Ukraine
(Received 6 January 1992)

Point-contact spectra of $GdBa_2Cu_3O_7$ (with Ag as counterelectrode) exhibit reproducible wide minima in the differential resistance dV/dI vs voltage V, which are attributed to the superconducting energy gap. The energy positions and widths of these gap features correspond to the gap distribution extracted earlier from $YBa_2Cu_3O_7$ point-contact spectra (with narrow gap minima). A comparison with the Blonder-Tinkham-Klapwijk model suggests that the distribution of wide gaps in $GdBa_2Cu_3O_7$ results from orificelike point contacts. The form of the spectra does not change when cooling below the antiferromagnetic transition at $T_N = 2.24$ K.

I. INTRODUCTION

One of the approaches to better understand the high-T_c superconductors is to vary the stoichiometry in these compounds and look for ensuing changes in the superconducting and normal-state properties. In $YBa_2Cu_3O_7$ (YBCO hereafter) a replacement of copper and barium by other atoms or a decrease of the oxygen concentration as a rule yield a decrease of the transition temperature T_c.[1] On the other hand, T_c does not change significantly when yttrium is replaced by trivalent rare-earth ions (with the exception of Pr) although they have a magnetic moment. At present, after many such studies and also other investigations there is general agreement that the CuO_2 layers are mainly responsible for the superconductivity,[2] while the yttrium layers (for YBCO) play a secondary role (they only help to yield the characteristic crystal structure and isolate adjacent CuO_2 layers), and the CuO chains act as a charge-carrier reservoir.

In line with these assumptions a two-layer model was proposed for YBCO (Ref. 3) based on the existence of pairs of CuO_2 layers within the crystallographic unit cell. As a result two bands with two different energy gaps appear in this model. For the more common model which assumes equal interaction between any adjacent CuO_2 layers a gap anisotropy was predicted.[4] In the present paper, we explore a possible gap anisotropy with an investigation of $GdBa_2Cu_3O_7$ (GBCO hereafter). A brief account of this work has already been given elsewhere.[5]

II. ANALYSIS OF POINT-CONTACT SPECTRA

Previous point-contact measurements[6] of YBCO polycrystals (measured against silver as a counterelectrode) have shown a strong dependence of energy position, shape, and even of the number of the gap features in the point-contact spectra (i.e., differential resistance dV/dI versus voltage V) on mutual electrode position (for experimental details, see Ref. 6). Figure 1 shows an additional set of point-contact spectra of YBCO. These spectra resemble the earlier ones which were interpreted in terms of gap anisotropy.[6] It is well known that Andreev reflection determines the I-V (or dV/dI-V) characteristics of N-c-S point contacts (c: constriction) as long as there is only a weak barrier at the interface between normal metal and superconductor. Andreev reflection leads to a minimum in the dV/dI versus V (voltage) curves.[7]

If there is an "ideal" point contact which one can visualize as a small-size orifice in a very thin insulating layer separating the two electrodes, with thickness small compared to the orifice diameter, Andreev reflection is possible for almost all electron trajectories. Thus even if there is a gap anisotropy, we will obtain in this case point-contact spectra which contain information of the energy-gap distribution which arises from averaging the anisotropic gaps over all possible angles of the electron trajectories. However, in reality there is a large probability to obtain a point contact which looks more like a channel connecting the two electrodes, since a typical point-contact diameter ($D \approx 100 \div 1000$ Å) and the thickness of an insulating layer on the sample surface are approximately of the same size. In this case, for the ballistic regime, the point-contact spectra will yield information about gaps only for directions almost parallel to the channel axis. D is so large that quantization of transverse momentum in the constriction is expected to play a minor role. Therefore, if there is a gap anisotropy one is likely to obtain different point-contact spectra for different point contacts.

In Fig. 1 we compare the experimental results of YBCO with the theoretical prediction for a usual superconductor in the single-gap case[7,8] (dotted curves). For curves 3 and 4 the shape of the gap minima (marked by arrows) does not differ largely from the theoretical prediction although the experimental minima are less pronounced than the theoretical ones. On the other hand,

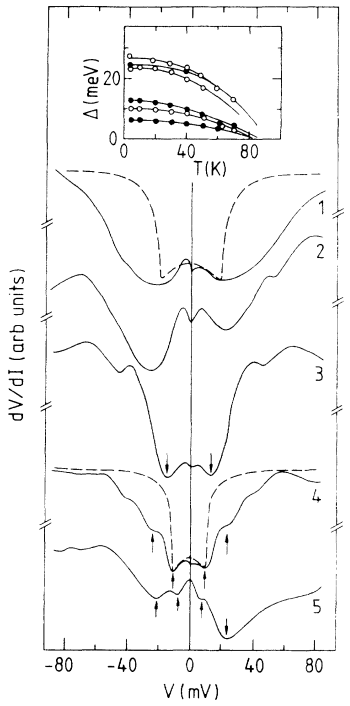

FIG. 1. Different types of the $YBa_2Cu_3O_7$ point-contact spectra (differential resistance dV/dI vs applied voltage V) at $T=4.2$ K. The arrows indicate the minima of the gap-related structure. Dotted curves were calculated from BTK theory (Ref. 7) for a single gap with adjusted barrier value to yield the best agreement with the experimental spectra. Inset: temperature dependence of the narrow gap-related structure Δ for $YBa_2Cu_3O_7$.

curves 1 and 2 exhibit very broad minima which cannot at all be described by theory. While most spectra are symmetrical, some asymmetry is occasionally observed (cf. curve 5 in Fig. 1). In the light of the above interpretation, we suggest that the spectra with narrow minima (curves 3 and 4) correspond to channel-like point contacts and those with wide minima (curves 1 and 2) to orificelike contacts. If there are two (or more) crystallites in a point-contact region (or, possibly, several conducting channels), one can obtain spectra with several gap minima (curves 4 and 5).

Our previous YBCO point-contact spectra have yielded different gap energies associated with the minima in dV/dI versus V.[6] Besides the above-mentioned interpretation in terms of anisotropy there are many other possible reasons for gap-value changes which might arise from the presence of surface effects on superconductivity or local changes in the point-contact region (for example, a local variation in oxygen concentration or a large local strain arising from the pressure exerted by the counterelectrode). However, these latter possibilities all lead to changes of the transition temperature. It is therefore important to point out that in our experiments all observed gap structures in the point-contact spectra arise in a superconducting region with the transition temperature of the bulk material, T_c^{bulk}. This is inferred from the temperature dependence of the point-contact resistance which shows a sharp drop at $T \approx T_c^{\text{bulk}}$ and also from the temperature dependence of the voltage at which the minima occur. These voltages, corresponding to gap-related structures Δ/e, all go to zero at approximately the same temperature T_c^{bulk} (see inset of Fig. 1). Hence we believe that different gap energies (for a narrow gap structure) correspond to different directions in k space. Hence, a wide gap structure (curve 1 in particular) is a result of the gap integration over a wide range of angles. By investigating many point contacts on the same sample, we have previously obtained the gap distribution for YBCO which is reproduced in the inset of Fig. 2(a). This histogram

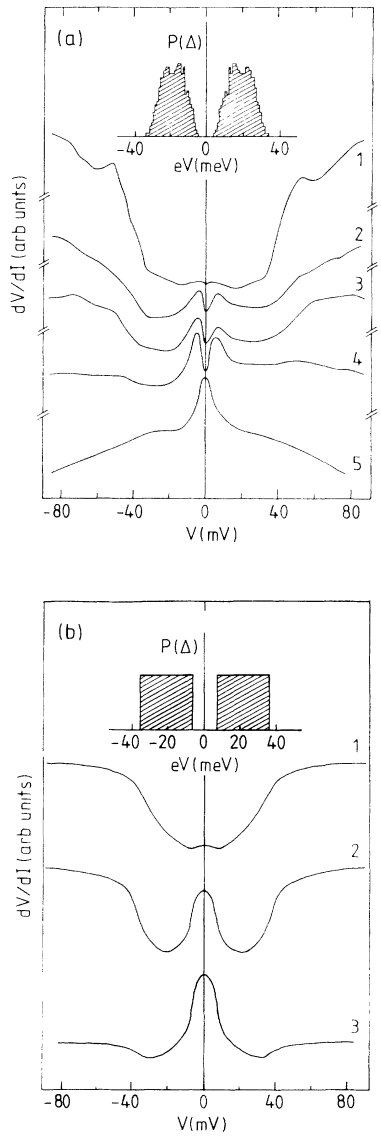

FIG. 2. (a) Point-contact spectra of $GdBa_2Cu_3O_7$ at $T=4.2$ K. Point-contact resistances at $V \approx 0$ are 2.5 Ohm (curve 1), 3 (2), 2 (3), 6 (4), 55 (5). Inset: energy-gap distribution extracted from $YBa_2Cu_3O_7$ point-contact spectra (Ref. 6) (for explanation see Ref. 6). (b) Calculated point-contact spectra for barrier values $z=0.2$ (curve 1), 0.5 (2), 1 (3). Inset: energy gap distribution function used for the calculation.

was obtained by choosing the halfwidth of the minimum as a single event.[6]

III. RESULTS AND DISCUSSION

Figure 2(a) shows our $GdBa_2Cu_3O_7$ point-contact spectra taken at 4.2 K which were obtained in different places (points) on the polycrystal surface. As previously, a Ag counterelectrode was used. As a rule, only one wide minimum at a position ranging between $\approx\pm 10$ and $\pm 35 \div 40$ meV was observed (besides an additional zero-bias minimum). With rising temperature this minimum goes toward smaller bias and disappears near $T \approx T_c^{\text{bulk}}$. This is shown in Fig. 3 for one particular point contact. The inset of Fig. 3 shows the temperature dependence of the point-contact resistance of this contact (measured at $V \approx 0$) and of the bulk susceptibility near T_c^{bulk} of the same sample. The very good coincidence in the local and bulk superconducting transitions indicates that we measure the bulk properties in this case, too. In particular, an appreciable suppression of T_c in the point-contact region by proximity-effect coupling[10] can be ruled out. The observation of spectra exhibiting only wide minima suggests that the GBCO point contacts are mostly orificelike. The reliability of the point-contact spectra allows a comparison of the experimental curves with the Blonder-Tinkham-Klapwijk (BTK) theoretical prediction for N-c-S contacts with different barrier values z.[7] Such a barrier in an interface region between normal metal and superconductor (due to an insulating layer or impurities) has a strong influence on the general shape of the point-contact spectra but the voltage position of the gap

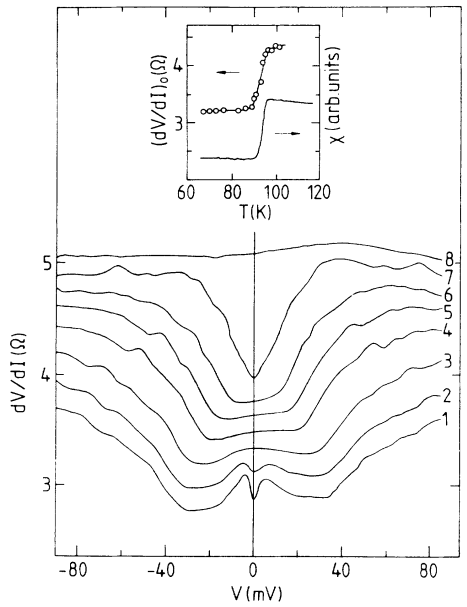

FIG. 3. Point-contact spectra of $GdBa_2Cu_3O_7$ at various temperatures $T=6$ K (curve 1), 19 (2), 40 (3), 62 (4), 71 (5), 81 (6), 91 (7), 98 (8). Note: curves are shifted vertically for clarity. Inset: temperature dependence of resistance $(dV/dI)_0$ at $V \approx 0$ for this point contact (solid circles) and of magnetic susceptibility χ for bulk material (solid line, after Ref. 9).

minimum in the dV/dI versus V curves does not change much (for $k_B T \ll \Delta$).[7] Figure 2(b) shows the results of a calculation for N-c-S contacts (according to BTK theory[7,8]) obtained with the simple rectangular gap distribution between 7 and 35 meV as shown in the inset of Fig. 2(b). Here we have summed over 29 BTK-like curves for different gaps corresponding to the rectangular gap distribution. For these calculations, different barrier heights z were assumed with z increasing from top to bottom.

The important point of the present investigation is that the experimental dV/dI versus V curves of GBCO can be modeled by the BTK theory if one assumes that there is a simple rectangular gap distribution which resembles the experimental distribution in the inset of Fig. 2(a). (As a matter of fact, assumption of a distribution mimicking the small indentation at the center of the experimentally determined distribution gave nearly identical results.) One may notice that for both series of curves there is a gentle shift of the deepest gap minimum to larger bias with rising z. In our opinion this comparison between experimental and calculated spectra clearly shows that there is indeed a gap distribution in the 1:2:3 superconductors. The maximum and minimum gaps correspond to $2\Delta/k_B T_c = 9$ and 1.8, respectively, with an average of 5.4. These numbers are in close agreement to our previous YBCO results.[6] It has been suggested that some fine structure within the superconducting energy gap arises from contributions of different planes parallel to the ab plane, i.e., within the layered structure of YBCO, which might be observable by PCS or tunneling spectroscopy.[11] How this modifies the gap anisotropy remains to be explored.

As mentioned above, the GBCO spectra almost always resemble the case of a presumably "ideal" orificelike point contact (curve 1 in Fig. 1) while for YBCO this was only occasionally observed. It is, however, very difficult to imagine that there were always orificelike point contacts for GBCO since there were no essential differences of the surface conditions with respect to YBCO. The typical crystallite size was about several micrometers (from electron-microscope measurements) and the insulating-layer thickness on the sample surface was approximately the same (≈ 100 Å from Auger spectroscopy depth profiling) for both compounds. This point deserves further study.

Turning to the PCS temperature dependence (Fig. 3), we note that the gap-related feature of GBCO shifts gradually toward zero voltage with increasing T, similar to what has been observed before for PCS of YBCO (see, e.g., inset of Fig. 1 and Ref. 6), and for tunneling spectra of $Bi_2Sr_2CaCu_2O_x$.[12] In these (and other) cases, the observed T dependence is in rough agreement with the BCS prediction. However, quite often features have been observed in tunneling or PCS which do not shift with T but simply disappear for $T \rightarrow T_c$.[13] The origin of these latter features and their relation to the BCS-like features is not known. We note, however, that features showing BCS-like T dependences are also regularly observed for YBCO single crystals with varying oxygen stoichiometry.[14] We should mention that quite often point contacts do not al-

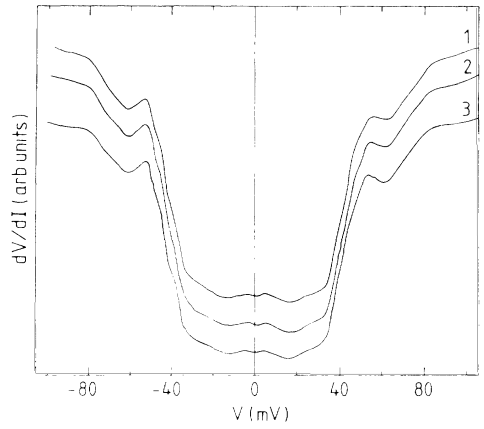

FIG. 4. Point-contact spectra of $GdBa_2Cu_3O_7$ at low temperatures $T = 1.7$ K (curve 1), 3.2 (2), and 4.2 (3).

low measurements up to $T > T_c$. However, where this has been possible a decrease of the position of the gap-related structure with T was always observed in our measurements.

In order to look for a possible effect of antiferromagnetic ordering of the Gd moments below $T_N = 2.24$ K,[15] we measured the point-contact spectra in the temperature range 1.7 to 4.2 K (see Fig. 4). No significant differences in the spectra were observed below and above T_N. Hence it is unlikely that fluctuating Gd magnetic moments give rise to the observed "gap-integrating" effect. However, we cannot exclude fluctuating Gd ions even below T_N in a point-contact region because it is difficult to make a small N-S interface without disturbance of the crystal structure.

In summary, we have obtained convincing evidence of a gap distribution in the 1:2:3 compounds from reproducible point-contact spectra of $GdBa_2Cu_3O_7$. These spectra could be successfully modeled within the standard BTK model of N-c-S point contacts. The spectra for GBCO appear to arise mainly from orificelike point contacts, in contrast to earlier YBCO spectra. The origin of this "gap integration" (over directions) remains to be established.

ACKNOWLEDGMENTS

The authors wish to thank G. Czjzek and H. J. Bornemann for allowing us to use their GBCO samples, and C. Sürgers for the Auger analysis of the sample surface. One of us (A.I.A.) thanks the Deutsche Forschungsgemeinschaft for the support of his stay at the Universität Karlsruhe. This work was supported by the Bundesminister für Forschung und Technologie, Project No. 13N5486 4.

*Permanent address: Institute for Low Temperature Physics and Engineering, Academy of Sciences of the Ukrainian SSR, Kharkov, Ukraine.

[1]A. V. Narlikar, C. V. Narasimha Rao, and S. K. Agarwal, in *Studies of High Temperature Superconductors,* edited by A. Narlikar (Nova Science, New York, 1989), Vol. 1, p. 341.

[2]A. R. Bishop, R. L. Martin, K. A. Müller, and Z. Tesanovic, Z. Phys. B **76**, 17 (1989).

[3]U. Hofmann, J. Keller, K. Renk, J. Schützmann, and W. Ose, Solid State Commun. **70**, 325 (1989).

[4]T. Schneider, H. De Raedt, and M. Frick, Z. Phys. B **76**, 3 (1989).

[5]A. I. Akimenko, G. Goll, H. v. Löhneysen, and V. A. Gudimenko (unpublished).

[6]A. I. Akimenko, G. Goll, I. K. Yanson, H. v. Löhneysen, R. Ahrens, T. Wolf, and H. Wühl, Z. Phys. B **82**, 5 (1991).

[7]G. E. Blonder, M. Tinkham, and T. M. Klapwijk, Phys. Rev. B **25**, 4515 (1982).

[8]A. V. Zaitsev, Zh. Eksp. Teor. Fiz. **86**, 1742 (1984) [Sov. Phys. JETP **59**, 1015 (1984)].

[9]H. J. Bornemann, Thesis, Universität Karlsruhe (1990).

[10]M. A. M. Gijs, D. Scholten, T. van Rooy, and A. M. Gerrits, Appl. Phys. Lett. **57**, 2600 (1990).

[11]M. Tachiki, S. Takahashi, F. Steglich, and H. Adrian, Z. Phys. B **80**, 161 (1990).

[12]M. Lee, D. B. Mitzi, A. Kapitulnik, and M. R. Beasley, Phys. Rev. B **39**, 801 (1989).

[13]For a review, see M. R. Beasley, Physica C **185-189**, 227 (1991).

[14]G. Goll, K. Seemann, G. Bräuchle, H. v. Löhneysen, A. Erb, G. Müller-Vogt, A. I. Akimenko, and I. K. Yanson, Fiz. Niz. Temp. **18**, 593 (1992).

[15]J. O. Willis, Z. Fisk, J. D. Thompson, S.-W. Cheong, R. M. Aikin, J. L. Smith, and E. Zirngiebl, J. Magn. Magn. Mater. **67**, L139 (1987).