Quasiparticle-tunneling properties of $Ba_{1-x}K_xBiO_3/BaBi_2O_y/Ba_{1-x}K_xBiO_3$ sandwich junctions

S. Martin, E. S. Hellman, A. Kussmaul, and E. H. Hartford, Jr.
AT&T Bell Laboratories, Murray Hill, New Jersey 07974
(Received 5 January 1993; revised manuscript received 1 March 1993)

Quasiparticle-tunneling measurements on sandwich junctions patterned from molecular-beam-epitaxy-grown $Ba_{1-x}K_xBiO_3/BaBi_2O_y/Ba_{1-x}K_xBiO_3$ trilayers are presented, where we find evidence for superconductor–insulator–normal-metal tunneling with a 2-meV gap. The subgap conductance is found to vary from a linear voltage dependence to a BCS-like voltage dependence. The temperature dependence of the zero-bias conductance and the thermal broadening of the conductance peak are studied.

Studies of transport behavior in high-T_c heterostructures composed of superconductor-insulator-superconductor layers are of current interest both from a fundamental and from a technological point of view. Tunneling measurements provide important information about quasiparticle states, and can elucidate carrier transport mechanisms in the superconductor and the nature of superconductivity itself. Measurements on sandwich junctions are significant for probing the properties of the superconductor/insulator interface which are crucial for characterizing the behavior of Josephson junctions. The bismuthate $Ba_{1-x}K_xBiO_3$ (BKBO) is especially suitable for fabrication of sandwich Josephson junctions due to its relatively large coherence length and its isotropic transport properties as compared to the high-T_c cuprates. Moreover, the bulk $T_c \simeq 30$ K of BKBO is promising for the viable operation of circuits using closed cycle refrigeration instead of the present operation of conventional superconductors at helium temperatures.

Evidence for a clean gap in BKBO was found by several groups in BKBO/Au tunnel junctions,[1–3] and by Pargellis *et al.*[4] in BKBO-thin-film/BKBO-single-crystal point-contact junctions. Fink *et al.* reported on hysteretic Josephson junctions made from $YBCO/SrTiO_3/BKBO$ trilayer films, where YBCO$=YBa_2Cu_3O_7$ and recently in $BKBO/KNbO_3/BKBO$ trilayers.[5] Transport measurements in BKBO/I/BKBO sandwich junctions were recently reported by Hellman *et al.*[6] Here, we present studies of quasiparticle tunneling properties in junctions patterned from molecular-beam-epitaxy-grown (MBE-grown) $BKBO/BaBi_2O_y/BKBO$ trilayers on (100) MgO substrates. We find superconductor–insulator–normal-metal (S-I-N) behavior with the gaps and T_c's in agreement with weak coupling BCS. The maximum junction conductances in the range 0.2–1 Ω^{-1} are two orders of magnitude higher than those typically found in point contact BKBO junctions. However, the specific conductances of $2–9\times10^3$ $\Omega^{-1}\,cm^{-2}$ are comparable or even higher than those of point-contact junctions. The temperature dependence of the zero-bias conductance and the conductance peak at the gap energy are in quantitative agreement with the form expected from thermal broadening of the density of states.

The films were grown by molecular-beam epitaxy on (100) MgO substrates with a 1700 Å bottom BKBO layer, an $\simeq 50$ Å $BaBi_2O_y$ barrier layer, and an 850 Å top BKBO layer. In order to improve the growth of the BKBO layers, the trilayers are grown on substrates with buffer layers made up of five alternating $BaBi_2O_y$ and BKBO layers. The trilayer films are processed into 80×140 μm^2 junctions using conventional photolithography. The patterns are defined by Ar-ion milling and the samples are annealed in an oxygen atmosphere to obtain superconducting BKBO layers. The T_c's of the films after annealing are around 20 K, lower than bulk T_c values, presumably due to the difficulty in controlling the potassium concentration. The details of growth, structural properties and junction preparation are described elsewhere.[7] The transport measurements were made with four probes using Ag contacts to BKBO. We measured both dc IV characteristics using a constant current source and the conductance directly using an ac modulation technique and found consistent results.

Figure 1 shows IV characteristics and corresponding conductance data taken at 4.2 K for three junctions. All have a finite zero-bias conductance of $G_0 \simeq 0.04$ Ω^{-1}, but the maximum conductance is sample dependent. The conductance peak is pronounced in samples 2 and 3 but broadened in sample 1. In all samples an asymmetry of the conductance with respect to zero bias is evident, as reported earlier for BKBO/I/metal junctions.[1] In those measurements the conductance was larger with the BKBO positively biased. In our data the conductance is larger when the bottom BKBO is at a positive bias corresponding to a negative bias in the measurement. The results suggest that the top and bottom BKBO layers are not equivalent and because the nucleation of the BKBO layer on the barrier is likely to suppress the T_c within a few coherence lengths of the interface we assume that the bottom BKBO layer has the higher T_c and the top layer is showing the metallic behavior.

In Fig. 2(a) the conductance data taken at several temperatures are plotted for sample 1. The data suggest an S-I-N behavior with a superconducting gap of $\Delta = 2.2$ meV and $T_c = 13$ K, resulting in the gap parameter $2\Delta/kT_c = 3.93\pm0.4$, within the range expected for weak coupling BCS superconductors and in agreement with previous reports on BKBO.[1–4] Note a sharp onset of os-

0163-1829/93/47(21)/14510(4)/$06.00

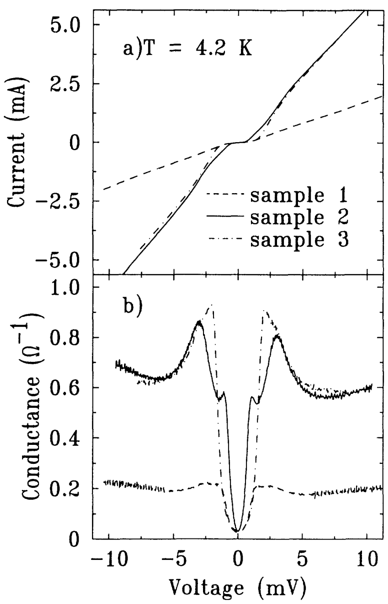

FIG. 1. Current-voltage characteristics and corresponding conductance-voltage data for three junctions measured at 4.2 K.

cillations in the low-temperature conductance data above $\simeq 5.7$ mV, which is found to be quite reproducible. As shown in the inset of Fig. 2(a) the "oscillations" have more than twice the amplitude of the typical experimental noise, and we find that the period of the oscillations is about 5–10 times larger than the voltage resolution. Fourier analysis of the data yield an oscillation period of 125 μV for $T=4.2$ K and 67 μV for $T=5.45$ K. A possible mechanism responsible for the oscillations is that the barrier together with the suppressed T_c portions of the BKBO layers is acting as a metal layer sandwiched between two superconducting layers (S-N-I-N-S structure). Due to Andreev reflection at the S/N interfaces, interference of quasiparticle wave functions can occur giving rise to periodic conductance oscillations above 2Δ.[8,9] Although we measure a gap of 2.2 meV at the barrier/BKBO interface, the BKBO layers farther from the barrier would have a larger gap consistent with the observation of the oscillations above 5.7 mV. The oscillation period depends on the Fermi velocity v_F of the carriers and the thickness d of the "metal" layer and is given by $\Delta V = \pi\hbar v_F/2ed$. Taking the thickness to be between $d=100$ (possible N layer) and 1000 Å (possible N-I-N layer) and $\Delta V = 125$ μV we obtain $v_F \simeq 10^5 - 10^6$ cm/s. However, this value is about one to two orders of magnitude smaller than the Fermi velocity estimated from band-structure calculations. Another possible scenario involves phonon slab modes that could give rise to conductance oscillations with periods of 95 μV (LA phonons) and 69 μV (TA phonons)[10] for a slab thickness of 1000 Å. Presently there is no clear experimental evidence for either of these scenarios.

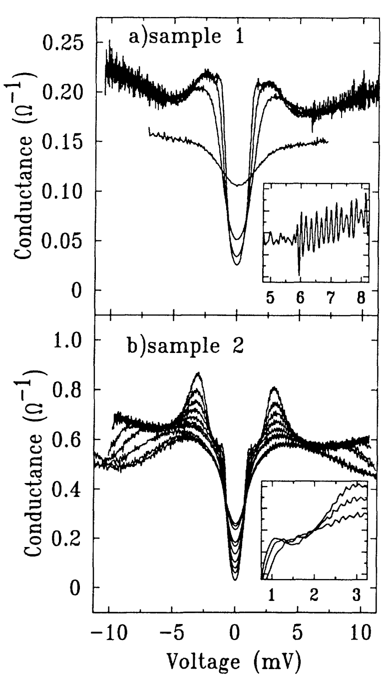

FIG. 2. (a) Conductance vs voltage measured in sample 1 at 4.2, 5.45, 7.8, and 13.4 K (in order of decreasing conductance at the gap). The inset is a blowup of the 4.2 K data showing onset of oscillations above 5.7 mV. (b) Conductance vs voltage measured in sample 2 at 4.2, 4.93, 6, 7, 8.2, 9, 9.7, 11.1, 11.6, and 11.8 K (in order of decreasing conductance at the gap). The inset is an expanded view of a selected region for the first three curves illustrating the disappearance of the 1 mV peak.

The temperature dependence of the tunneling conductance in sample 2 is shown in Fig. 2(b) where a gaplike feature at 3 mV is pronounced at low temperatures, broadens with increasing temperature and finally disappears at $\simeq 13$ K. A second gaplike feature at 1 mV can be resolved for temperatures up to $\simeq 6$ K, which disappears by moving to higher voltages [see inset of Fig. 2(b)]. Although this could be considered evidence for S-I-S' behavior, note that even the larger peak disappears by moving towards higher voltages. Assuming that the two features represent peaks of quasiparticle tunneling at the sum and difference of two distinct gaps, we obtain $2\Delta/kT_c = 3.57$ for a 2 meV gap and 3.87 for a 1 meV gap, in good agreement with weak coupling BCS.

The data in Fig. 3(a) for sample 3 indicate a more obvious BCS-like S-I-N junction behavior with suppressed subgap conductance. Only one gap is resolved here at 2.2 mV which persists up to $\simeq 14$ K. This measurement, similar to sample 1, indicates an S-I-N behavior with the gaplike feature due to the bottom layer having a $T_c = 14$ K and a suppressed T_c top layer. We then obtain a gap parameter of $2\Delta/kT_c = 3.65$, consistent with the other samples. Note in Fig. 3(a) a conductance peak at zero bias for the curves taken at 15.52 and 17.14 K. This feature seems to be due to the overlap of the conductance gaps for the case when the gap is small compared to the thermal broadening. In Fig. 3(b) fits to the data of the normalized conductance are plotted using the BCS form for the density of states: $N(E) = E/(E^2 - \Delta^2)^{1/2}$ and taking into account temperature smearing and finite quasiparticle lifetime effects by substituting $E' + i\delta E$ for E,[1] with δE as a fitting parameter. The data fit well to the BCS expression for temperatures up to about 9 K. The

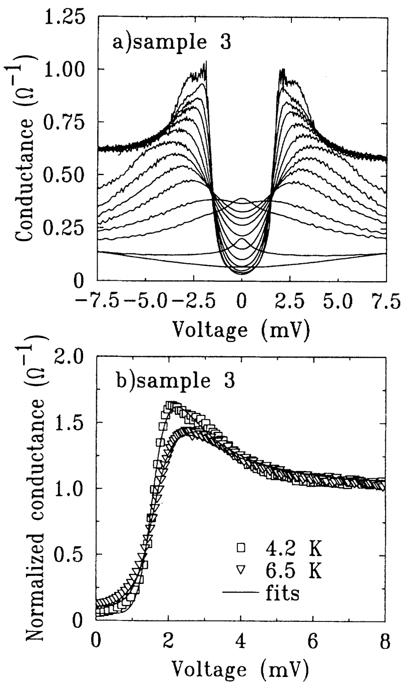

FIG. 3. (a) Conductance vs voltage measured in sample 3 at 2, 4.2, 5.4, 6.45, 7.87, 9.36, 10.36, 12.24, 13.07, 14.02, 14.67, 15.52, 17.14, and 18.46 K (in order of decreasing conductance at the gap). (b) BCS fits to the normalized conductance measured in sample 3 at 4.2 and 6.45 K. Fit parameters are $\Delta=1.4$ meV (4.2 and 6.45 K) and $\delta E=0.7$ meV (4.2 K) and 0.9 meV (6.45 K).

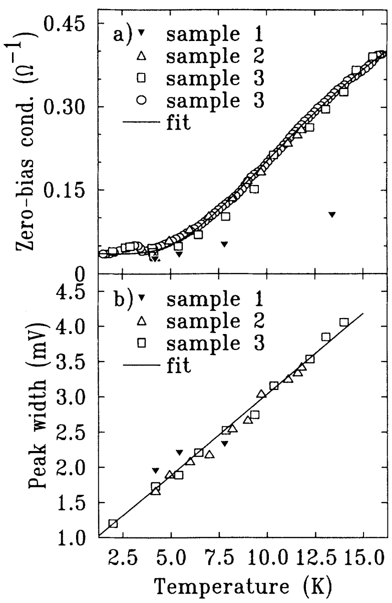

FIG. 4. (a) Temperature dependence of the zero-bias conductance for all three samples. Due to the onset of lateral resistance in the BKBO layers the conductance could not be measured reliably above $\simeq 16$ K. For sample 3 the circles denote data obtained from a direct conductance measurement while the sample was cooled. The other symbols show data obtained from the conductance curves in Figs. 2 and 3(a). The solid line is a fit to the data ($\bigcirc$) from 3 to 16 K using an expression derived from the thermal broadening of the density of states. (b) Temperature dependence of the conductance peak width for all three samples. The slope of the linear fit suggests that the thermal smearing of the peak is due to tunneling into a superconducting density of states rather than inelastic tunneling.

fits are less reliable for higher temperatures owing to substantial broadening and large zero-bias conductance. The gaps derived from the fits are typically 65% and the energy smearing about 30% of the values obtained from the peak positions. Since the energy smearing is large, the peak positions rather than the fitted gap values reflect the intrinsic quasiparticle excitation spectrum.

The characteristic features of tunneling data in all high-T_c junctions have been the presence of finite zero-bias conductance and the broad conductance peak near the gap, and the temperature dependence of these quantities. The temperature dependence of the zero-bias conductance $G_0(T)$ of all three samples is plotted in Fig. 4(a). Except for sample 1 the other sets of data show a decrease of G_0 by an order of magnitude on cooling from 15 to 2 K. Above 15 K the BKBO layers become normal and give rise to a lateral resistance, so that the conductance of the sandwich junction cannot be measured reliably. Below 4 K we find in sample 3 an anomalous discontinuity of the conductance which is quite reproducible and is not presently understood. Note that $G_0(T)$ saturates to a background conductance of $\simeq 0.04\ \Omega^{-1}$ below 5 K, rather than decrease to zero for $T \rightarrow 0$. The finite temperature smearing of the density of states results in a tail extending to zero bias and can therefore be used to relate the zero-bias conductance to the gap.[11] In the limit $kT \ll \Delta$ an S-I-N tunneling yields:

$$G_0(T)=G_n(2\pi\Delta/kT)^{1/2}\exp(-\Delta/kT)\ ,$$

where G_n is a normalizing factor and the exponential term is due to the Fermi function describing the density of states. Figure 4(a) shows that the data from samples 2 and 3 give an excellent fit to the above expression after the saturation conductance value is subtracted, although the above expression is only valid in the low-temperature regime ($T<10$ K). The fit yields a gap of $\Delta=2.3$ meV in good agreement with the mean position of the conductance peaks for the lowest temperatures in Fig. 1(b). The excellent agreement of the $G_0(T)$ data with predictions for conventional BCS superconductors is in contrast to the linear temperature dependence reported for high-T_c cuprate tunnel junctions.[12] In the latter case the $G_0(T)$ behavior has been correlated to the anomalous linear temperature dependence of the normal-state resistivity in the CuO planes. The present measurements show, however, that the $G_0(T)$ could arise from conventional thermal broadening of the density of states.

The origin of the gaplike feature can be either inelastic tunneling or tunneling from a low-T_c metal into a superconducting density of states.[13] The former case involves the phonon density of states of the barrier and the adjacent electrodes. The thermal broadening in either case is different. The inelastic tunneling effect gives rise to a broadening $\propto 5.4kT$, whereas the superconducting density of states effect results in a broadening $\propto 3.5kT$. In Fig. 4(b) the full width at half maximum of the conductance peak is plotted as a function of temperature for the three samples. The data show a linear temperature dependence with a slope of $2.7kT$. The small value of the slope can be considered evidence for the observed thermal broadening of the conductance peak being due to the density of states effect.

Interestingly, the tunneling data contain features, such as temperature dependent zero-bias conductance and voltage dependent subgap conductance, which are similar to those measured first in single crystal YBCO/Au S-I-N tunnel junctions[12] and have been originally interpreted as a result of non-BCS-like superconductivity in YBCO. However, recent reports on 60 and 90 K YBCO based tunnel junctions have shown evidence for BCS-like behavior.[14] Similarly, present measurements and previous reports on BKBO/Au S-I-N tunnel junctions clearly provide evidence that the superconducting properties of bulk BKBO can be very well accounted for by weak coupling BCS theory. The non-BCS-like features of tunneling in BKBO trilayers are likely to be dependent on properties of the barrier and the barrier/BKBO interface. Similar behavior in high-T_c tunnel junctions has been recently accounted for by the marginal Fermi liquid model of high-T_c superconductivity.[15] The differences in the tunneling data have been ascribed to the probing of a smaller or larger section of the Fermi surface, e.g., as a result of variations in the barrier thickness. It is shown that the tunneling matrix elements differ depending on the admixture of states near or far from k_F, leading to a flat background conductance or a linear voltage dependent conductance, respectively. The linear background conductance is associated with probing a small portion of the Fermi surface, so that the subgap conductance is expected to deviate from the conventional BCS-like form. Comparing the different forms of the conductance curves in Figs. 2–3 we note that sample 1 has the most pronounced and sample 3 the least pronounced linear background conductance above the gap. As expected from the above argument, sample 1 has indeed the most non-BCS-like and sample 3 the most BCS-like tendency. Variations in the barrier thickness and/or scattering of quasiparticles in the barrier could be responsible for this behavior.

In conclusion we have shown that transport measurements in BKBO/I/BKBO trilayers reveal important information on quasiparticle tunneling properties of patterned sandwich junctions. We found that the BKBO layers adjacent to the barrier within a few coherence lengths have suppressed T_c's as compared to the bulk films. Although the conductance curves resemble those found in high-T_c cuprate junctions, the temperature dependence of the zero-bias conductance and the thermal broadening of the gaplike feature can be well accounted for by weak coupling BCS theory.

We acknowledge many useful discussions with B. Batlogg, A. F. Hebard, and P. B. Littlewood. One of us (A.K.) acknowledges partial support by the Consortium for Superconducting Electronics.

[1]F. Sharifi *et al.*, Phys. Rev. B **44**, 12 521 (1991).

[2]H. Sato, H. Tagaki, and S. Uchida, Physica C **169**, 391 (1990).

[3]Q. Huang *et al.*, Appl. Phys. Lett. **57**, 2356 (1990).

[4]A. N. Pargellis *et al.*, Appl. Phys. Lett. **58**, 95 (1991).

[5]R. L. Fink *et al.*, Appl. Phys. Lett. **61**, 595 (1992); R. L. Fink *et al.* (unpublished).

[6]E. S. Hellman *et al.*, Physica C **201**, 166 (1992).

[7]E. S. Hellman *et al.*, in *Advances in Superconductivity IV,* Proceedings of 4th International Symposium on Superconductivity (ISS '91), edited by H. Hayakawa and N. Koshizuka (Springer, Tokyo, 1992), p. 855.

[8]W. J. Tomasch, Phys. Rev. Lett. **15**, 672 (1965); J. M. Rowell and W. L. McMillan, *ibid.* **16**, 453 (1966).

[9]M. Holcomb, J. P. Collman, and W. A. Little, Physica C **185-189**, 1747 (1991).

[10]A. V. Belushkin *et al.*, Physica C **199**, 103 (1992).

[11]M. Tinkham, *Introduction to Superconductivity* (Robert E. Krieger, Malabar, 1980), p. 47.

[12]M. Gurvitch *et al.*, Phys. Rev. Lett. **63**, 1008 (1989).

[13]J. Geerk, X. X. Xi, and G. Linker, Z. Phys. B **73**, 329 (1988).

[14]A. M. Cucolo, C. Noce, and A. Romano, Phys. Rev. B **46**, 5864 (1992).

[15]P. B. Littlewood and C. M. Varma, Phys. Rev. B **45**, 12 636 (1992).