Electrical and magnetic transport properties of laser-deposited \( \text{Ba}_{1-x}\text{K}_x\text{BiO}_3 \) thin films

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We report measurements of the electrical and magnetic transport properties of high-quality superconducting \( \text{Ba}_{1-x}\text{K}_x\text{BiO}_3 \) (BKBO) thin films grown by \textit{in situ} pulsed laser deposition. The best films exhibit sharp resistance and magnetic transitions with zero resistance above 28 K and widths of 0.3 K, and nearly linear normal state resistance extending from the transition to above room temperature. Critical current densities obtained by transport in patterned BKBO lines are greater than \( 3 \times 10^6 \, \text{A/cm}^2 \) at 4 K. The transverse magnetoconductance exhibits predominantly BCS-like behavior with an upper critical field slope of \(-0.78 \, \text{T/K near} \ T_c \), giving \( H_{c2}(0) = 15.2 \) T and a BCS coherence length \( \xi = 46 \, \text{Å} \). We also observe substantial field broadening suggestive of flux flow dissipation.

The observation of nearly ideal BCS (Bardeen-Cooper-Schrieffer) quasiparticle tunneling\(^1\) and Josephson tunneling\(^2\) into the medium-temperature superconductor \( \text{Ba}_{1-x}\text{K}_x\text{BiO}_3 \) (BKBO) has triggered efforts to develop a superconductor device technology based on BKBO thin films and tunnel junctions. At the same time, there is renewed interest in the mechanism of superconductivity in BKBO. Many of the tunneling properties\(^3\) and the presence of a strong isotope effect\(^4\) suggest that BKBO is an ordinary electron-phonon-coupled BCS superconductor. Like the classic superconductors, BKBO is a cubic material with a moderately long coherence length (\( \sim 50 \, \text{Å} \)), and exhibits no evidence for the antiferromagnetic correlations that may dominate the normal state behavior in the cuprates. However, it has been reported that BKBO may share many characteristics with the cuprates such as a predominately linear electrical resistivity,\(^5\) non-Drude behavior in the optical reflectivity and dielectric loss function,\(^6\) anomalous penetration depth measured by muon spin relaxation,\(^7\) and linear tunneling conductance above the energy gap.\(^8\) An understanding of the pairing in BKBO may serve as a bridge to understand the mechanism for superconductivity in the high-temperature oxides.

Recently, it has become possible to fabricate high-quality thin films of BKBO by sputtering,\(^9\) molecular beam evaporation,\(^10\) thermal coevaporation,\(^11\) and pulsed laser ablation.\(^12\) In this letter, we report measurements of the electronic and magnetic transport properties of BKBO films grown by pulsed laser deposition. In zero field, the BKBO films exhibit low metallic normal state resistivities and excellent superconducting state properties, with the sharpest transitions and high current densities required for electronic device applications. In an applied magnetic field, the resistive transition shifts in accordance with BCS theory, and there is substantial broadening of the magnetoresistance indicating that flux flow may be important in the BKBO films. The critical current density is also BCS-like in its temperature and magnetic field dependence, showing no evidence for weak link coupling.

The BKBO films were grown by \textit{in situ} pulsed laser deposition from a \( \text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3 \) target using an ArF excimer laser; details of the growth process have been previously reported.\(^13\) The films were deposited at 580–620 °C in pure Ar at a pressure of 1 Torr, and oxygenated in a 12 min soak in 1 atm \( \text{O}_2 \) introduced only after the films were cooled to 410 °C. All of the films reported here were grown on (100)-\( \text{MgO} \) substrates and have a nominal thickness of 1500 Å. The films exhibit a single-crystalline phase with (100) orientation, in-plane epitaxy, and composition close to the starting pellet stoichiometry.\(^14\) Films typically have smooth surfaces and exhibit very little microcracking. For transport measurements, the BKBO films were patterned into 25 and 100 µm wide lines by standard optical lithography (AZ1350J) and dry etching by Ar ion milling, keeping the film temperature below 50 °C at all times in the processing. In good films, we find no evidence that the film properties are degraded by the lithography process. Films are stable when stored in a dry environment at room temperature; films stored for six months have shown no appreciable change in their superconducting properties.

Our BKBO films exhibit sharp magnetic and resistive transitions. The magnetic transition of a good BKBO film, measured by a two coil inductive screening method, is shown in Fig. 1(a). The film exhibits an onset at about 28 K and a width (10%–90%) of less than 0.5 K. Our highest observed transition onset temperature in a film is about 29 K, although single crystals of BKBO can exhibit transition temperatures as high as 31 K.\(^15\) The SQUID susceptibility of the BKBO ablation target used to grow this film has roughly the same onset as the film but a substantially broadened width (\( \sim 5 \, \text{K} \)). The resistive transition of a line patterned from the same film occurs at a higher temperature, as shown in Fig. 1(a). The film exhibits zero resistivity at about 28 K (corresponding to the onset of the magnetic screening) and a transition width of 0.3 K. In the normal state, the best films are metallic with a nearly linear resistivity above \( T_c \) that extends to well above room temperature, as in Fig. 1(b). Just above the transition, the resistivity flattens out—this temperature-dependent region is more pronounced in films with lower \( T_c \). All films exhibit a large residual resistivity; the resistivity at the transition for the sample shown is 250 µΩ cm and is increased...
FIG. 1. (a) Comparison of the magnetic screening and resistivity transitions for a BKBO thin film. The onset of the magnetic transition and the zero resistance temperature each occur at 28 K, with transition widths less than 0.5 K. (b) Resistivity vs temperature for a patterned BKBO film. The resistivity is nearly linear from the transition to above room temperature, with residual resistance ratio $\rho(300)/\rho(T_c) \approx 2$.

FIG. 2. (a) Superconducting critical current density vs temperature at zero magnetic field measured by transport in a 25 $\mu$m wide line patterned from the same BKBO film as in Fig. 1, attaining $3 \times 10^6$ A/cm$^2$ at low temperature. (b) Critical current density vs temperature in transverse magnetic fields up to 5 T applied normal to the film surface.

FIG. 3. (a) Resistivity vs temperature in magnetic fields up to 6 T for the patterned film in Fig. 2. The transition shifts to lower temperature and broadens as the field is increased. (b) Upper critical magnetic field vs the onset transition temperature—the critical field slope of $-0.78$ T/K yields a critical field $H_c(0)$ of about 15 T. (c) Field broadening of the resistive transition demonstrated by shifting the magnetoresistance at each field by the superconducting onset temperature. The fractional broadening $(\Delta T/T_c)$ is comparable to that in the cuprates at the same magnetic fields.
for a perpendicular field. We see only a small variation in the resistance (≤20%) as the film plane is rotated relative to the field. Since BKBO is cubic, we attribute this anisotropy to the film geometry. The magnetoresistance transition shows characteristics intermediate between classical (low $T_c$) and the cuprate (high $T_c$) superconductors. The shift in the onset temperature is characteristic of a classic BCS superconductor, but there is also substantial broadening of the transition with increasing field. From the field dependence of the onset temperature, determined by extrapolating the magnetoresistance to the normal state value (at $T_c$), we determine $dH_{c2}/dT$ from Fig. 3(b). The critical field slope $(-0.78 T/K)$ and zero field transition temperature (28.5 K) implies an upper critical field $H_{c2}(0)$ of 15.2 T, assuming the Werthamer–Helfand–Hohenberg formula $H_{c2}(0) = \frac{\Phi_0}{2\pi a^2}$. We deduce a coherence length of 46 Å. All of these values are in reasonable agreement with results obtained from magnetization measurements in bulk composites and single crystals of BKBO. We demonstrate the field broadening of the resistivity by shifting the curve at each field by the corresponding onset temperature; these curves are plotted in Fig. 3(c). Although some field broadening is observed in all type II superconductors, the effect in BKBO is very significant. At 6 T, the width of the transition (10%–90%) is about 15% of the zero field $T_c$; the fractional broadening at the same field in the cuprates (in the c-axis direction) is comparable. However, the transition width in our BKBO films is roughly linear in the applied field, in contrast to the $H^{2/3}$ dependence reported in the cuprates.

The broadening of the magnetoresistance and rapid suppression of the critical current density with field suggest that flux flow and flux creep may be important in these materials. We do find evidence for thermally activated behavior in the resistivity in finite field at low temperatures, with a field-dependent activation energy ranging from 0.67 eV at 1 T to 0.04 eV at 6 T. Vortex barrier energies of comparable magnitude have been deduced from magnetic flux relaxation measurements in BKBO single crystals.

In conclusion, we have fabricated high-quality epitaxial thin films of BKBO that exhibit sharp magnetic and resistive superconducting transitions above 28 K and current densities above $10^6$ A/cm$^2$. For the most part, the BKBO films generally behave like conventional BCS superconductors in their transport and tunneling properties. However, the linear resistivity, field broadening of the resistive transition, and strong suppression of the critical current with field show similarities with the cuprates and warrant further study. Prospects for development of a BKBO device technology are encouraging with the improvement in film quality we have attained by the pulsed laser deposition growth procedure.

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