Upper critical field in $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$: Magnetotransport vs. magnetotunneling

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(Received 4 July 1997; accepted in final form 27 November 1997)

PACS. 74.60Ec – Mixed state, critical fields, and surface sheath.
PACS. 74.25Dw – Superconductivity phase diagrams.
PACS. 74.50+r – Proximity effects, weak links, tunneling phenomena, and Josephson effects.

Abstract. – Elastic tunneling is used as a powerful direct tool to determine the upper critical field $H_{c2}(T)$ in the high-$T_c$ oxide $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$. The temperature dependence of $H_{c2}$ inferred from the tunneling follows the Werthamer-Helfand-Hohenberg prediction for type-II superconductors. A comparison will be made with resistively determined critical-field data.

The upper critical field $H_{c2}$ of high-$T_c$ superconductors remains a contradictory issue. In classical type-II superconductors this quantity has been unequivocally determined from the magnetotransport measurement and its temperature dependence can, in most cases, be well described by the Werthamer-Helfand-Hohenberg (WHH) theory [1]. However, for the high-$T_c$ superconductors, $H_{c2}$ extracted from magnetotransport data reveals an unusual increase with decreasing temperatures without any saturation down to low temperatures. This upward curvature is observed in a pronounced way in the superconducting cuprates $\text{Sm}_{2-x}\text{Ce}_x\text{CuO}_4$ [2], $\text{Tl}_2\text{Ba}_2\text{CuO}_{6+δ}$ [3], $\text{Bi}_2\text{Sr}_2\text{CuO}_y$ [4], and $\text{YBa}_2(\text{Cu}_{1-x}\text{Zn}_x)\text{O}_{7-δ}$ [5]. However, the effect is also found in the fully three-dimensional and non-magnetic $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$ [6]. Among others, a bipolaron scenario [7], an unconventional normal state [8], a strong electron-phonon coupling [9], the presence of inhomogeneities and magnetic impurities [10] have been put forward for an explanation of the anomalous $H_{c2}(T)$ dependence. Because depinned vortices, either in the liquid or solid state, cause a finite dissipative resistance before reaching the full transition to the normal state, the complexity of the $H$-$T$ phase diagram in high-$T_c$’s [11] undermines any direct determination of the upper critical field from magnetotransport data. There are
Fig. 1. – Magnetoresistance of the Ba$_{1-x}$K$_x$BiO$_3$ single crystal at different temperatures. Arrows: $H_{c2}^t$ obtained from tunneling.

Fig. 2. – Differential conductance of the Ba$_{1-x}$K$_x$BiO$_3$-Ag tunnel junction measured at different temperatures. The inset shows the temperature dependence of the superconducting energy gap obtained from the tunneling conductances at zero magnetic field and 2 T together with the BCS prediction (full lines).

indications that, also in the fully 3D system of Ba$_{1-x}$K$_x$BiO$_3$, fluctuations can lead to a melting of the vortex-glass state [12]. This could be a reason complicating the determination of $H_{c2}$ from a dissipative measurement as magnetoresistance.

Avoiding the dissipative mechanisms which could obscure the determination of $H_{c2}$ from transport measurements, we show that tunneling measurements can be used as an effective and direct method for the determination of the upper critical field. The temperature dependence, obtained from tunneling, of $H_{c2}$ in Ba$_{1-x}$K$_x$BiO$_3$ follows the WHH model revealing a saturation at low temperatures. At the tunneling $H_{c2}$ values the resistance is very close to the full resistive transition into the normal state as measured on the same sample. This result was obtained repeatedly on several samples.

The single-crystalline Ba$_{1-x}$K$_x$BiO$_3$ samples grown by electrochemical crystallisation were dark-blue crystals of a cubic shape with a size of about 0.6 mm. The superconducting transition of our crystals was single stepped and sharp with $T_c \simeq 23$ K as determined by susceptibility measurements. The low-temperature resistivity was about 100 $\mu$\Omega cm with the metallic temperature dependence. The tunnel junctions were prepared by painting a silver spot of about 0.1 to 0.2 mm diameter on the surface of the crystal. The interface between the silver and the Ba$_{1-x}$K$_x$BiO$_3$ counter electrodes served as a natural barrier forming a planar normal-insulator-superconductor (N-I-S) tunnel junction. Low-resistance electrical contacts were prepared for the four-probe measurements of the current-voltage ($I$-$V$) and differential conductance ($dI/dV$) characteristics of the tunnel junction. The tunneling measurements were performed in magnetic fields up to 30 T perpendicular to the planar junction enabling the formation of the vortex state in the junction area. On the same samples the magnetoresistance was measured using a four-probe measurement at low frequencies.

Figure 1 shows the magnetoresistance $R(H)$ of the Ba$_{1-x}$K$_x$BiO$_3$ single crystal up to 26 T at temperatures from 1.5 K to $T_c \simeq 23$ K. The resistive transitions are shifted and broadened towards lower temperatures as the magnetic field is increased, although the broadening is much smaller than in the case of the cuprates. A simple evaluation of the transition field defined as
Fig. 3. – Normalized tunneling conductances in magnetic fields from zero up to 30 T in steps of 2 T (if not otherwise specified) at the indicated temperatures.

$H^*(T)$, where $R/R_n$ equals, for instance, 0.1, 0.5, or 0.9 ($R_n$ is the normal-state resistance) leads to a positive curvature of $H^*(T)$ down to the lowest temperatures. If $H^*(T)$ is defined for $R$ even closer to $R_n$, the curvature of $H^*(T)$ changes at the lowest temperatures which makes that the dependence $H^*(T)$ depends on the chosen criterion.

In the following the normalized tunneling conductances of our junctions are presented, where the normal-state conduction for the normalization is taken above $H_{c2}(T)$ for the temperature $T$ under investigation. Figure 2 shows a quality certificate of our junction. The spectra can be perfectly described by the Dynes formula [13] of the quasi-particle density of states $\rho(\epsilon)$ smeared by the finite temperature at which the N-I-S junction has been measured. $\rho(\epsilon) = \text{Re}[\epsilon'/[(\epsilon'^2 - \Delta^2)^{1/2}]]$ contains an isotropic superconducting energy gap $\Delta$ and a complex energy $\epsilon' = \epsilon - i\Gamma$ which takes account of some additional smearing $\Gamma$. The $\Gamma$ smearing of the spectrum is case-dependent with a tendency $\Gamma/\Delta \to 0$ in the best junctions. As mentioned already in the original paper of Dynes et al., such an “intrinsic” width of the spectrum can be the consequence of anisotropy effects, noise, or concentration fluctuations. The presence of microphases due to fluctuations in the potassium and oxygen concentration seems to be a general problem in a substitution system like Ba$_{1-x}$K$_x$BiO$_3$ [6]. The Dynes formula fits our tunnel spectra at 1.5 K and zero magnetic field with $\Delta = 3.9 \pm 0.1$ meV and $\Gamma = 0.4 \pm 0.1$ meV, yielding $2\Delta/kT_c = 3.9 \pm 0.1$ and indicating that Ba$_{1-x}$K$_x$BiO$_3$ is a BCS-like superconductor with a medium coupling strength [14]. In the inset the temperature dependence of the superconducting gap is shown for the data from fig. 2 and also for data taken at $B = 2$ T.

The broadening parameter $\Gamma$ is found to be independent of temperature.

At high magnetic fields the normalized tunneling conductances of the Ba$_{1-x}$K$_x$BiO$_3$-Ag junction are displayed in fig. 3 for different constant temperatures. With increasing magnetic fields an increasing smearing of the superconducting features in the tunneling spectra is observed. At a certain field strength no structure from superconductivity can be found anymore and the transition from a S-I-N to a N-I-N junction is accomplished. Similar tunneling data have been obtained on a thin Ba$_{1-x}$K$_x$BiO$_3$ film in a parallel field up to 7 T at 0.45 K [15]. Unlike a parallel-field configuration, our experiment on a cubic single crystal involves the occurrence of a mixed state in a strong magnetic field.
In the mixed state the tunneling conductance will probe an average of the local densities of states. For an isolated vortex, the superconducting order parameter is zero at the center, increases linearly up to a coherence length distance $\xi$ away from the center where it saturates to the zero-field value. The local quasi-particle density of states (DOS) equals the normal-state DOS at the vortex core, but is broadened near the vortex due to the pair-breaking effect of the local magnetic field (as described by the Abrikosov-Gor’kov theory developed further by Maki, de Gennes and others [16]). In the limit of moderate fields ($H \ll H_{c2}$), Caroli, de Gennes and Matricon [17] have shown that the main contribution to the density of states at the Fermi energy comes from the low lying states localized in a vortex core. Each isolated vortex gives a contribution equivalent to a normal region of radius $\xi$ yielding for the density of states at the Fermi level $\rho(0)$

$$\rho(0) \propto \rho_n(0) \xi^2,$$

where $\rho_n(0)$ is the normal state DOS at the Fermi level. Thus, the total averaged density of states at the Fermi level is proportional to $\rho_n(0) \xi^2$ per area ($H_{c2}/H$) $\xi^2$ occupied by one vortex giving $\rho(0) \approx \rho_n(0) H / H_{c2}$. However, of more relevance for critical-field data, also close to $H_{c2}$ a linear field dependence of $\rho(0)$ has been found by solving the linearized Ginzburg-Landau equation in the mixed state [18]. A very sensitive method to determine the upper critical field from tunneling experiments is to display the normalized zero-bias tunneling conductance as a function of the field strength [19]. The observation of a sharp transition is then taken as a proof of a good homogeneity of the sample.

In fig. 4 we present the zero-bias tunneling conductance as a function of the applied magnetic field for different temperatures. A linear dependence of $dI/dV(0)$ as a function of the applied field can be found in a limited field range. At the highest fields a “tailing” of the zero-bias conductance towards the normal-state value is observed, and a finite value of the zero-bias conductance is found already at zero magnetic field. The latter effect is obviously due to the $\Gamma$ broadening. Also the tailing effect at the highest fields could be related to the same cause as the $\Gamma$ broadening, i.e. a certain inhomogeneity in the sample. The observed tailing effect resembles the behavior in the resistive transition close to the transition into the normal state (see fig. 1). A linear extrapolation of the zero-bias conductance to the field where the normalized conductance equals unity, as shown by the full lines in fig. 4, has been used to
determine the upper critical field. Figure 5 shows the obtained temperature dependence of the upper critical field $H_{c2}(T)$. Also the points $H^*(T)$ obtained from 90% of the magnetoresistance transition ($R/R_0 = 0.9$) are indicated. The slope of $H_{c2}(T)$ near $T_c$ is different from that of the transition field $H^*(T)$ determined from resistance data. We note that the tunneling critical fields $H_{c2}(T)$ are at fields where the bulk resistivity is very close to the onset of superconductivity (as indicated by the arrows for $H_{c2}$ in fig. 1).

As a very significant result, $H_{c2}(T)$ shows a clear saturation at the lowest temperatures as expected for the WHH theory. From all dissipative measurements on different samples of Ba$_{1-x}$K$_x$BiO$_3$ so far done [6], [12], [15], only a linear increase of $H^*(T)$ for decreasing temperatures has been obtained. Also recent susceptibility measurements reveal this effect in the temperature dependence of the irreversibility field [20], [21] down to the lowest temperatures (0.4 K in [20]).

Besides the above-mentioned tailing effect in the zero-bias tunneling-conductance and the bulk resistive transition near the superconducting transition, the same tailing can also be observed near $T_c$ in $\Delta(T)$ (see fig. 2) and in $H_{c2}(T)$ (see below in fig. 5). We suppose that, despite the quality of our sample, stoichiometric inhomogeneity could play a role in this phenomenon. However, as we will discuss below, a more intrinsic cause related to superconducting fluctuations could also explain this broadening in the superconducting transition.

Klein et al. [12] suggested that in Ba$_{1-x}$K$_x$BiO$_3$ a vortex-glass melting transition driven by fluctuations can obscure the magnetotransport determination of $H_{c2}$. Their measurements of the electric field vs. current density $E-J$ show that a second-order phase transition from a vortex glass to a vortex liquid state does exist in this system. The presence of the liquid phase can induce strong fluctuations below $H_{c2}$ related to the motion of the flux lines, but these fluctuations can be quite small above $H_{c2}$. This can be the reason why $H_{c2}$ (see arrows in fig. 1) is quite close to the onset of the resistive transitions. In this approach the foot of the resistive transition is determined by the melting of the vortex lattice giving a positive curvature in the temperature dependence of the line $H_g(T)$ for the liquid-solid transition. The feet of the curves in fig. 1 could be indeed well fitted as $R \sim [H/H_g(T) - 1]^\beta$ corresponding to the vortex glass melting theory as introduced by Fisher et al. [22], where $H_g(T)$ is the magnetic field of the melting transition. The resulting fitting parameter $\beta = 4.1 \pm 0.5$ is in perfect agreement with the value obtained on a different Ba$_{1-x}$K$_x$BiO$_3$ crystal with $T_c \approx 31$ K [12]. The melting line $H_g(T)$ reveals a positive curvature and can be described by the power law temperature dependence $H_g = H_0(1 - T/T_c)^{3/2}$ as shown in fig. 5. This is also in good agreement with the recent measurement of the irreversibility field on a similar sample [20] indicating that the melting and irreversibility lines coincide in Ba$_{1-x}$K$_x$BiO$_3$.

In the $H-T$ phase-diagram of fig. 5 the initial slope of the upper critical field $(-dH_{c2}/dT)_{T_c}$ is about 1.7 $\pm$ 1.8 T/K. To emphasize more the fact of the saturation of the upper critical field at the lowest temperatures, we note the closeness of the zero-bias conductance data for 1.5, 3 and 4.2 K in comparison with the data taken at other temperatures in fig. 4. In fig. 5 we also present the WHH upper critical-field line with an uncertainty comparable to the experimental error bars. Taking into account that in a system with important fluctuations the $H_{c2}$ boundary should not be very sharp, a satisfactory agreement is found.

We have presented here a direct non-dissipative determination of the upper critical field in Ba$_{1-x}$K$_x$BiO$_3$ using the tunneling effect. $H_{c2}(T)$ can be satisfactorily described by the Werthamer-Helfand-Hohenberg theory. In the Cu oxides, the existence of a resistive state within a large part of the $H-T$ diagram complicates an unambiguous determination of the critical field from transport data. Therefore, it would be very interesting (and decisive for certain proposed superconducting mechanisms) to study the upper critical field in the cuprates with the non-dissipative tunneling method.
We acknowledge fruitful discussions with S. I. Vedeneev and support of the EU grant No. CIPA-CT93-0183 and the Slovak VEGA contract No. 2/1357/94.

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