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Magnetoresistivity of ultra-thin YBa$_2$Cu$_3$O$_{7-\delta}$ films on sapphire substrate

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Abstract

Magnetoresistivity of YBa$_2$Cu$_3$O$_{7-\delta}$ ultra-thin films with thicknesses between 7 and 100 nm deposited on sapphire substrate using CeO$_2$ and PrBa$_2$Cu$_3$O$_{7-\delta}$ buffer layers has been measured to analyze temperature dependence of the second critical magnetic field $B_{c2}$. $B_{c2}$ was determined using $T_c$ values obtained by fitting of resistivity vs. temperature data with the fluctuation conductivity theory of Aslamazov-Larkin. At $T \rightarrow T_c$ the $B_{c2}(T)$ dependence shows a crossover from downturn to upturn curvature with the increase of the YBCO film thickness.

Keywords: high-temperature superconductor YBCO; thin film; pulsed-laser deposition; terahertz detector

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1. Introduction

The high-temperature superconductor YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) is a prominent candidate for ultra-fast detector applications due to its strong electron-phonon coupling. Energy relaxation rates in the range of only a few picoseconds have been measured from optical to infrared wavelengths [1]-[3]. These ultra-fast detectors are required for the detection and analysis of terahertz (THz) pulses with pulse widths down to 1 ps generated in electron storage rings [4]. For ultimate time resolution and highly sensitive detectors, the film thickness of the detecting element has to be minimized. Recently we demonstrated the successful detection of synchrotron THz pulses with our thin-film YBCO detectors [5]. These measurements revealed a non-bolometric detection mechanism. Earlier experiments showed a strong influence of the dynamics of vortices on the response of these superconducting detectors to continuous wave THz radiation [6]. Another application for YBCO thin-films are superconducting single-photon detectors for the soft x-ray spectrum range. To determine the cut-off wavelength of these devices, the electron diffusivity is required [7] which is extracted from the temperature dependence of the second critical magnetic field.
We present a detailed and systematic study of the temperature dependence of the second critical magnetic field \( B_{c2}(T) \) of films with thicknesses \( d \) from 7 to 100 nm. The deviation from the linear dependence of \( B_{c2}(T) \) close to the zero-field critical temperature predicted by the Ginzburg-Landau theory is discussed in detail.

2. Sample fabrication

The YBCO thin films were fabricated by pulsed-laser deposition (PLD) using a KrF excimer laser (wavelength 248 nm). For the deposition of the YBCO thin films the laser was focused on a rotating stoichiometric YBCO target with an energy density of 1 J/cm². The sapphire substrate was glued to the heater with silver paste to ensure good thermal contact during deposition. The distance between substrate and target was 50 mm in on-axis position.

The YBCO PLD process was optimized with focus on ultra-thin YBCO films with transition temperatures well above 77 K. To fabricate detectors for the THz frequency range with low dielectric losses, the YBCO films were grown on sapphire substrates. To reduce the crystalline mismatch between sapphire and YBCO and to prevent the diffusion of aluminum into the YBCO film at high deposition temperatures, buffer layers were used in the deposition process. A standard buffer material for YBCO on sapphire is cerium oxide (CeO₂). A CeO₂ buffer layer with a film thickness of 8 nm was deposited at a substrate temperature of 800°C and a partial oxygen pressure of \( p_{O2} = 0.9 \) mbar. YBCO films with thicknesses between 7 and 100 nm were then deposited in-situ on top of the CeO₂ layer at the same temperature with a partial oxygen pressure of \( p_{O2} = 0.7 \) mbar. After the YBCO deposition the oxygen pressure was increased to 900 mbar and the substrate temperature was ramped down to 530°C with a rate of 10°C/min. The temperature was kept constant at 550°C for ten minutes for annealing of the YBCO film. Afterwards the heater was ramped down to 400°C before switching off and cooling down exponentially to room temperature.

The as-deposited films were characterized by measuring the temperature dependence of the resistance. The zero resistance transition temperature \( T_{c0} \) was defined as the temperature at which the resistance decreases below 1% of the normal state resistance above the transition. The dependence of \( T_{c0} \) on the YBCO film thickness for the 8 nm thick CeO₂ buffer layer is shown in Fig. 1 (triangles).

For YBCO film thicknesses above 30 nm \( T_{c0} \) is nearly constant and well above 77 K. For films thinner than 30 nm \( T_{c0} \) decreases significantly. This can be explained by the above-mentioned lattice mismatch leading to oxygen deficiency in the YBCO film, which results in a reduction of \( T_{c0} \) down to 5 K for 9 nm thick films. The crystalline mismatch was reduced by the introduction of an additional buffer layer made of \( \text{PrBa}_2\text{Cu}_3\text{O}_7 \) (PBCO). The lattice mismatch of approximately 1% between YBCO and PBCO allows for growing of very thin superconducting YBCO films with high crystalline quality. To protect the YBCO thin film during the patterning process, a PBCO protection layer was deposited on top of the YBCO film. The introduction of the 25 nm PBCO buffer layer combined with the PBCO protection layer of the same thickness resulted in \( T_{c0} = 79 \) K of YBCO films with thicknesses of only 10 nm (see circles in Fig. 1).

Using standard photo-lithography and ion-milling technique the multi-layers were patterned into micrometer-sized bridges (width \( w = 20 \) µm, length \( l = 200 \) µm – 1000µm) for four-point magnetoresistivity measurements.

3. Magnetoresistivity of YBCO thin films

Fig. 2 shows magnetoresistivity data near the superconducting transition for the 50 nm thick sample measured in magnetic fields up to 9 T parallel to the c-axis of the sample. At temperatures in the vicinity of \( T_{c0} \) the temperature dependence of the second critical magnetic field \( B_{c2}(T) \) is described as

\[
B_{c2}(T) \propto (1 - T / T_{c0})^n
\]

with \( n = 1 \) for conventional low-\( T_{c0} \) superconductors according to Ginzburg-Landau (GL) [8].
The definition of the critical temperature \( T_c \) has a major influence on \( B_{c2}(T) \) for the high-temperature superconductor YBCO as it was already mentioned by Oh et al. [9]. Therefore, to find the correct transition temperature the fluctuation conductivity theory of Aslamazov and Larkin (AL) [10] was used. Expressed in terms of the measured resistance \( R \) and arbitrary dimensionality \( m = 1, 2, 3 \) it renders

\[
R(T) = \frac{R_n}{1 + CR_n \frac{\xi}{A_n} \left( \frac{T}{T - T_c} \right)^{2-0.5m}}
\]

with \( A_n \) a constant factor depending on the dimensionality of the studied superconducting system [11]. \( R_n \) is the normal state resistance, \( A = wd \) is the cross-section of the bridges and \( C \) a fitting parameter.

The \( R(T) \)-dependencies were fitted by (2) in a narrow temperature range in the vicinity of the onset of the superconducting transition using all three values for \( m \). The model for two-dimensional (2D) superconducting systems \((m = 2)\) with \( A_2 = e^2/(16hd) \) fit our data best with the smallest values of \( C \). Here \( h \) is the reduced Planck constant and \( e \) is the elementary charge. The obtained results are in agreement with earlier reported data [9] and our expectations since we consider temperatures very close to \( T_c \) where \( \xi \) becomes large and therefore the two-dimensional approximation \( d \ll \xi \) [11] is justified.

A typical result is shown in Fig. 3 where the temperature dependence of resistance (circles) of the 8 nm thick YBCO film is fitted by the AL fluctuation theory according to equation 2 (solid line) with \( T_c = 75.3 \text{ K} \) indicated by the arrow.

For all film thicknesses the extracted \( T_c \) values are close to the midpoint of transition. Therefore, we chose the 0.5\( R_n \)-level of transition commonly used to evaluate the temperature dependence of the second critical magnetic field. Fig. 4 shows the series of the \( B_{c2} \) temperature dependence for film thicknesses from 7 to 100 nm. The solid lines represent fits according to equation 1 using \( n \) as a fitting parameter.

At temperatures \( T < 0.95T_c \) (magnetic fields above 3 T) all films show a linear dependence of \( B_{c2} \) on temperature according to the GL theory for conventional superconductors [8] (equation 1 with \( n = 1 \)). In Fig. 4 this is emphasized by linear fits (dashed lines) to the data points at high magnetic inductions which show the same slope for all film thicknesses.

At \( T > 0.95T_c \) the \( B_{c2}(T) \) dependencies vary strongly with film thickness. We observe a downturn curvature of the \( B_{c2}(T) \) dependence close to \( T_c \) for ultra-thin films with thicknesses between 7 and 20 nm with a minimum exponent of \( n = 0.8 \) for the 7 nm thick sample. The exponent \( n = 0.5 \) has been observed by Fang et al. [12] for twinned YBCO 150 \text{ nm} sized crystals.

The exponent \( n \) increases with increasing film thickness converging to the linear GL dependence \((n = 1)\) for \( d = 30 \text{ nm} \). The films thicker than 30 nm reveal an upturn curvature of \( B_{c2}(T) \). Similar \( B_{c2}(T) \) behavior has been demonstrated by Yeshurun [13] and Tinkham [14] with an exponent of \( n = 1.5 \) for 100 \text{ nm} YBCO crystals. Also Oh et al. [9] reported an upturn curvature with \( n = 1.3 \) for 1 \text{ µm} thick epitaxial YBCO films using both a 0.9\( R_n \) criterion and the critical fluctuation theory. The thickest studied YBCO film in this report with \( d = 100 \text{ nm} \) shows a \( B_{c2}(T) \) dependence with the exponent \( n = 1.14 \).

Since the activation energy \( U \) for flux motion is inversely proportional to the applied magnetic field a possible explanation for the observed behavior may be a change in the strength of vortex pinning in films with different thicknesses. However, since the excitation current during all magnetoellistivity measurements was 100 nA any influence by depinning of vortices may be neglected.
4. Conclusion

We have developed a pulsed-laser deposition process for ultra-thin YBCO films used for fast THz detectors and single-photon detecting applications. A detailed study of films with $7 \text{ nm} < d < 100 \text{ nm}$ in magnetic fields up to 9 T revealed a deviation of the linear upper critical field line predicted by GL. Employing the fluctuation conductivity theory by AL showed that this deviation cannot be explained by the correct choice of the transition temperature. Therefore, we conclude that the downturn curvature of the upper critical field line for ultra-thin films below 30 nm reveals a real suppression of the transition temperatures at low magnetic fields, which might have influences on the detection mechanism of our detectors. Further experiments to study the origin of this suppression of the superconducting properties of the ultra-thin YBCO films close to zero-field transition will be carried out.

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