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Piezoelectric effect and ultrasonic attenuation near T_c in single-crystal $YBa_2Cu_3O_{7-x}$ superconductors

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Abstract

The attenuation of ultrasonic waves in single-crystal, high T_c $YBa_2Cu_3O_{7-x}$ has been measured. At T_c a pronounced peak is observed for the longitudinal c_{11} mode. The existence of a piezoelectric effect in the high T_c superconductor has been confirmed in the temperature range from 4 to 280 K in a pulse echo ultrasonic measurement employing a $YBa_2Cu_3O_{7-x}$ crystal as a transducer for the longitudinal c_{33} mode. The peak near T_c for the absorption of the c_{11} mode can be explained by a model taking into account the piezoelectric properties of the material and a treatment of the electron gas according to the Drude theory.

1. Introduction

As reported in a previous publication [1], we have observed an absorption peak for the longitudinal c_{11} mode in single-crystal, high T_c $YBa_2Cu_3O_{7-x}$ (YBaCuO) at T_c. Absorption anomalies have also been reported for other superconductors and have been treated by various models [2–4]. None of these models leading to relatively broad structures around and below T_c is adequate to describe the observed rather narrow absorption anomaly in YBaCuO, which compares in width with the derivative of the a.c. susceptibility [1].

Assuming the presence of a piezoelectric effect in YBaCuO, we have developed a model to describe the experimental results. In materials exhibiting the piezoelectric effect, an acoustic wave is accompanied by an electromagnetic wave [5, 6], which is attenuated in an electrically resistive material. The attenuation has a maximum if the specific internal impedance attributed to the piezoelectric coupling mechanism (ρ_p) is matched with the specific electrical resistivity (ρ_e) of the material. The attenuation is zero for a perfect conductor and small for a material with high resistivity. For $\rho_p < \rho_e$ at a temperature just above T_c this is in accordance with the observed anomaly [1]. Before turning to a quantitative treatment of this effect, we present the results of a search for experimental evidence for the piezoelectric effect in YBaCuO.

2. Experimental results

The possibility of ferro- or antiferroelectric phases in high T_c superconducting materials has already been discussed in the literature. Evidence that YBaCuO exhibits a large static permittivity [7], ferro- or antiferroelectricity [8, 9], piezoelectricity [10–12] and pyroelectricity [11, 13] has been reported. The possible relations between ferroelectricity and high T_c superconductivity have also been discussed [9, 14, 15]. As a consequence of the ferroelectric state for a material which is at least gradually polarized (ferroelectric domains partially aligned), the piezoelectric effect will be present.

We have observed the piezoelectric effect by the detection of longitudinal ultrasonic waves near 10 MHz with YBaCuO single crystals (c cut, T_c of about 90 K) prepared to serve as ultrasonic transducers for the longitudinal c_{33} mode (Fig. 1). For these experiments a YBaCuO transducer is attached to a plane-parallel tantalum single crystal acting as a transmission medium, with a piezoceramic transducer attached to the opposite side. In the temperature regime from about 20 to 250 K we detect with the YBaCuO transducer signals resulting from the transit and echoes of the ultrasonic pulses excited by the piezoceramic transducer. The range of temperatures for which ultrasonic waves can be detected (Fig. 2) is similar to the regime where a hysteresis for the elastic constants has been observed [1]. This hysteresis has already been attributed to ferroelectric phases [9]. A hysteresis is also present in the temperature dependence of the signals at the YBaCuO transducer (Fig. 2).

The signals from the YBaCuO transducer do not disappear below T_c (determined by the a.c. susceptibility). We attribute this to a reduced T_c of the inner part of the YBaCuO transducer. The ultrasonic signal

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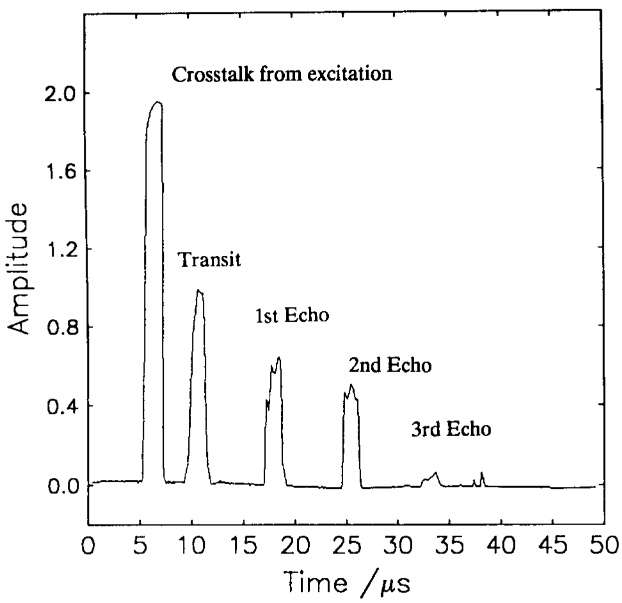

Fig. 1. Time dependence of electrical signals (amplitude normalized to transit; video detection) from pulsed longitudinal ultrasonic waves at 10 MHz with a detecting transducer manufactured from single-crystal YBaCuO (about 2 mm×2 mm×0.2 mm) for the c_{33} mode at a temperature of 70 K.

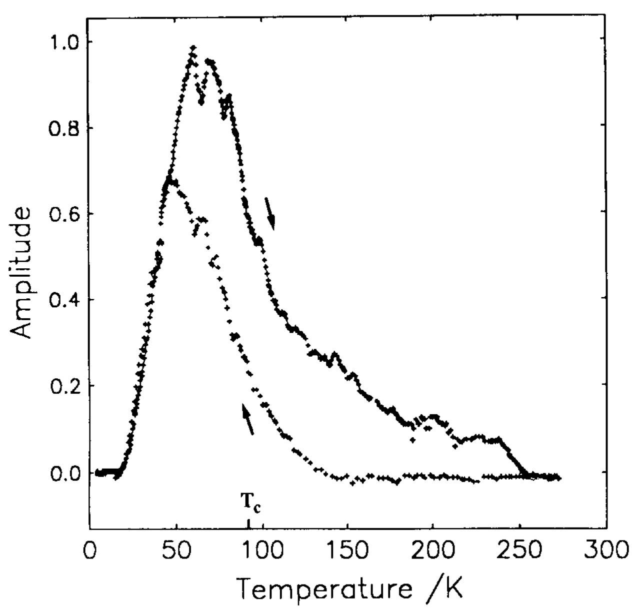

Fig. 2. Temperature dependence of the amplitude of the transit signal (as in Fig. 1) normalized to the maximum value in the range 4–280 K.

decays visibly below about 60 K and finally disappears at 20 K. We expect that in this temperature regime the superconducting layers close to both surfaces, which carry the electrodes serving as a pickup for the observed signals, are growing, which effectively reduces the size of the piezoelectrically active part of the transducer. The signal can also be reduced by low impedance shunts which may develop. Below 20 K the transducer is completely superconducting or a fully superconducting channel is established between the electrodes, in accordance with the observed electrical resistance between the electrodes.

Similar signals (Fig. 1) but different in size have been observed with all three transducers prepared for the experiments. The strongest signals were about 2% (amplitude) of those obtained with a piezoceramic transducer of comparable size replacing the YBaCuO transducer. Owing to the relatively low electrical resistivity of YBaCuO, we were not able to polarize the crystals used for the experiments by applying sufficiently large electric fields while cooling from above the Curie temperature. Since we rely on accidental polarizations, the size of the observed signals sets only a lower limit for the piezoelectric coupling constant of YBaCuO.

3. Discussion and model

We conclude from our experimental results and the crystallographic structure of YBaCuO (related to the perovskites) that the observed piezoelectric effect is caused by the ferroelectric polarization of the material in the temperature range coinciding with the range where ultrasonic signals have been observed with the YBaCuO transducer. Since the voltage induced by the piezoelectric effect will most likely be short circuited below 20 K, the material may still be ferroelectric in that temperature regime. We refrain here from a more detailed discussion, since the treatment of electrically conducting material requires a generalized definition of the piezoelectric effect (not in accordance with the usual definition based on an observable electric field). Owing to the plate-like shape of the rather small crystals (large surfaces normal to the c axis), we could not search for evidence of the piezoelectric effect in other orientations. The presence of the piezoelectric effect for the c_{11} mode in YBaCuO is therefore assumed for the following discussion.

To derive a model for the attenuation of ultrasonic waves in YBaCuO, the material is described as a series of mechanically connected, plane-parallel transducers (short with respect to the wavelength of the longitudinal mode at 10 MHz) which are inhomogeneous with respect to T_c (as described above). This model approximates the structure of the non-perfect "single crystal" used in the experiments for the determination of the ultrasonic absorption. The sample consists of equally oriented, mechanically connected small crystallites separated in part by small amounts of other phases. These impurities serve also as a transport channel for the oxygen in the annealing process used to raise T_c.

The transducers are shunted by a resistor which is determined by the resistivity of the outer part of the

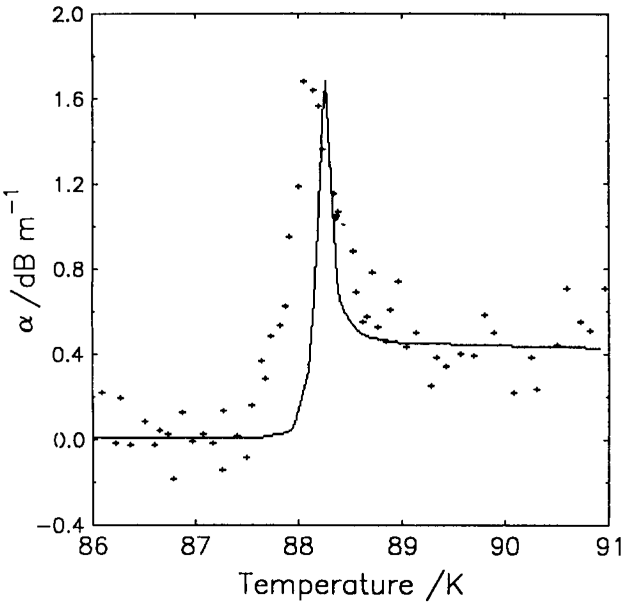

Fig. 3. Experimentally determined temperature dependence (+) of the absorption of ultrasonic waves (longitudinal c_{11} mode at 10 MHz) in YBaCuO corrected for a linear background absorption (according to ref. 1) and results of model calculations (solid curve) for $v=5600$ m s^{-1}, $\rho_d=6300$ kg m^{-3} and $\rho_s=1\times10^{-3}$ Ω m (just above T_c).

crystallites (transducers), with a temperature dependence according to the a.c. susceptibility and a resistivity above T_c typical for YBaCuO. For similar volume fractions of the low and high T_c components in the inhomogeneous (with respect to T_c) single crystal we derive an ultrasonic attenuation

$$\alpha\ (\mathrm{dB\ m^{-1}})=10\,\frac{\omega^2 e_{11}^{\ 2}}{\rho_d v^3}\,\frac{\rho_s}{1+\rho_s^{\ 2}/\rho_p^{\ 2}}\log_{10}\mathrm{e}$$

where ω is the angular frequency of the ultrasonic waves, e_{11} is the relevant piezoelectric constant, ρ_d is the density of the sample, v is the velocity of the longitudinal ultrasonic waves, ρ_s is the specific electrical resistivity, $\rho_p=1/\omega\epsilon_{11}(\omega)$ is the specific internal impedance related to the piezoelectric coupling mechanism and $\epsilon_{11}(\omega)=\epsilon_o\epsilon_d(\omega)$ is the relevant dielectric permittivity.

On the basis of this model a quantitative fit has been obtained for the attenuation near T_c (Fig. 3) with optimized parameters ϵ_d and e_{11}. The dielectric constant of the piezoelectrically active part for 10 MHz ($\epsilon_d=-1.13\times10^7$) is within an acceptable range for this material [9, 16]. The piezoelectric constant ($e_{11}=37$ C m^{-2}) selected for the best fit to the experimental results is large, but within an order of magnitude with respect to the range for other materials of the perovskite family.

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