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Elastic consequences of the dielectric behavior of $YBa_2Cu_3O_{7-\delta}$

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Abstract

Owing to the fact that superconductivity can be described as a dielectric phenomenon, where the screening of the repulsive electron–electron interaction is accounted for by introducing a temperature-dependent dielectric function $\epsilon(\omega, \mathbf{k})$, in this paper emphasis is put on a brief discussion of values of ϵ consistent with high superconducting transition temperatures *in principle* as well as on *experimental techniques* which allow determination of the temperature-dependence of the dielectric function $\epsilon(\omega, \mathbf{k})$.

1. Introduction

The actually known high-T_c superconductors are oxides and therefore belong to a class of materials which stands out for having normally poor electric conductivity and the tendency to transform into an electrically polarized state. The still unexpectedly high superconducting transition temperatures in high-T_c cuprates together with the generally enormous sensitivity of the electric polarizability of apical oxygen to displacements (and distortions of the surrounding lattice) in perovskites, has stimulated the search for possible links between the dielectric behavior of these good conducting oxides and superconductivity. Furthermore, it should be kept in mind that the usually incompatible phenomena (high electric conductivity and static polarization) could coexist in a system where the conducting and polarizable parts are well separated, as is the case for the sandwich-type structure of $YBa_2Cu_3O_{7-\delta}$ (YBCO). Here two layers with metallic conductivity (Cu–O planes) are separated by an intermediate sheet of less-conducting and highly polarizable interlayers.

An attempt to understand high-T_c superconductivity in terms of a dielectric function is straightforward because already conventional superconductivity can be described as a dielectric effect. Lattice distortions (phonons) originating from the motion of conduction-charge carriers are associated with dynamic electric polarization capable of overscreening the repulsive Coulomb interaction. This screening can be described by means of a dielectric function $\epsilon(\omega, \mathbf{k})$. The complexity of electron–electron, electron–lattice, electron–exciton (electron–anything) interactions is now devolved to the dielectric function, determination of the correct form of which is a task not to be approached here.

2. T_c formulae

An extension of the well-known Bardeen–Cooper–Schrieffer (BCS) expression [1] for the superconducting transition temperature in a weak coupling superconductor to the strong coupling case leads to expressions of the McMillan type [2, 3] which can be written in the general form [4]

$$T_c \leqslant \Theta \, e^{-1/\lambda - \mu^*} \tag{1}$$

Here $\Theta = \hbar\omega_0/k_B$ is a measure for the maximum response frequency ω_0 of the coupling subsystem (phonons in BCS superconductors), λ stands for some attractive electron–electron interaction while μ^* is the frequency-dependent Coulomb interaction related to the pure Coulomb interaction μ by [5]

$$\mu^* = \mu \Bigg/ \left[1 + \mu \ln\left(\frac{E_F}{\hbar\omega_0}\right)\right] \tag{2}$$

Here E_F denotes the Fermi energy. The dielectric function can be used to describe the reduced Coulomb repulsion. It can be shown that [4–6]

$$\mu - \lambda = \left\langle \frac{(\epsilon_c(0, \mathbf{k}) - 1)}{\epsilon(0, \mathbf{k})} \operatorname{sign} m^* \right\rangle \tag{3}$$

where the total permittivity

$$\epsilon(0, \mathbf{k}) = \epsilon_b(0, \mathbf{k}) + \epsilon_c(0, \mathbf{k}) - 1 \tag{4}$$

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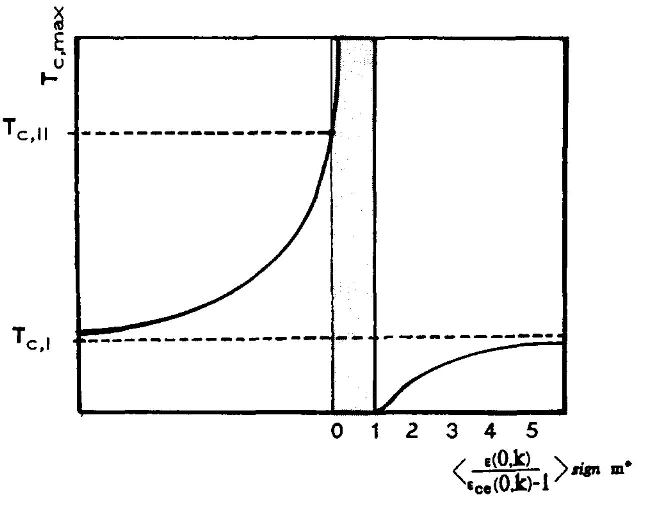

Fig. 1. The dependence of T_c on the dielectric function (see text). The shaded area marks the region of forbidden values for $(\epsilon(0, \mathbf{k})/(\epsilon_c(0, \mathbf{k})-1))$.

includes the permittivity of the bound charges $\epsilon_b(0, \mathbf{k})$ and the permittivity of the conduction charge carriers $\epsilon_c(0, \mathbf{k})$, and sign m^* guarantees the nominator $((\epsilon_c-1)*\text{sign } m^*)$ to be positive for electrons as well as for holes. $\langle \ldots \rangle$ stands for some averaging about E_F. As is obvious from Fig. 1 where the dependence of T_c on the electric permittivity (see eqns. (1)–(3)) is plotted, there are two regions which would be consistent with a high T_c. If the classical restrictions on $\epsilon(0)$ are valid $(\epsilon(0) \geqslant 1)$, T_c would reach a low limiting value at $(\epsilon/(\epsilon-1)) \rightarrow \infty$, *i.e.* $T_{c,I}$. Ginzburg and Kirzhnitz [5] give arguments allowing $\epsilon(0, \mathbf{k})$ to become negative. This allows T_c to approach much higher values, *i.e.* $T_{c,II}$. So the highest T_c should be obtained in materials where $\epsilon(0, \mathbf{k})$ is negative and as small as possible. Equivalently, the electric polarizability $\alpha = (\epsilon-1)/\epsilon$ should be large and because $(\epsilon_c - 1) \leqslant 0$, the charge carriers should be holes. Having in mind the large electric polarizability of the apex oxygen ions $(\alpha \gg 1)$ in perovskites [7] and recalling the fact that in most of the known high-T_c materials the charge carriers are indeed holes, the question arises as to whether or not these high-T_c systems represent examples of the above-mentioned ideas.

3. Experiments

In this context *experimental* verification of a large electric polarizability or at least anomalous dielectric behavior above T_c, is highly desirable. None the less, regarding the high electric conductivity of high-T_c cuprates, experimental determination of the dielectric behavior of the bound charges with conventional techniques (such as, for example, capacitance measurements) is hindered by the highly mobile conduction charge carriers. There are, however, indications for an anomalous dielectric behavior of YBCO [4]. It was shown, for example [4], that in the presence of a strong external magnetic field, the sound-induced r.f. electromagnetic fields can be evaluated and the temperature-dependence of the electric polarizability can be determined. Figure 2 shows the temperature dependence of the electric polarizability (α'') of an oxygen-deficient sinter-sample of $YBa_2Cu_3O_{7-\delta}$ ($T_c \approx 89$ K) in the presence of a 6 T magnetic field, obtained from analyzing the ultrasonic quantities and the quantities of the sound-induced electromagnetic fields as described in ref. 4. The necessity to apply high magnetic fields as well as the very high acoustic quality of the samples required obviously limits the applicability of the described technique. Nevertheless, with the above-mentioned approach, first experimental indications of the anomalous dielectric behavior of YBCO above T_c were obtained.

Because in classical (isolating) dielectrics the dielectric behavior is known to influence the temperature-dependent elastic properties, depending on electric boundary conditions, the experimental work presented here was stimulated by the search for the influence of the dielectric behavior on the elastic properties in this material.

It is known that in conventional (isolating) dielectrics domains with different orientation of electric polarization appear during the transition into the polarized state, in order to minimize the electric energy $(-\mathbf{E}\cdot\mathbf{P})$ between the electric polarization $\mathbf{P}$ and the concomitant electric field $\mathbf{E}$ of the separated charges. The size of

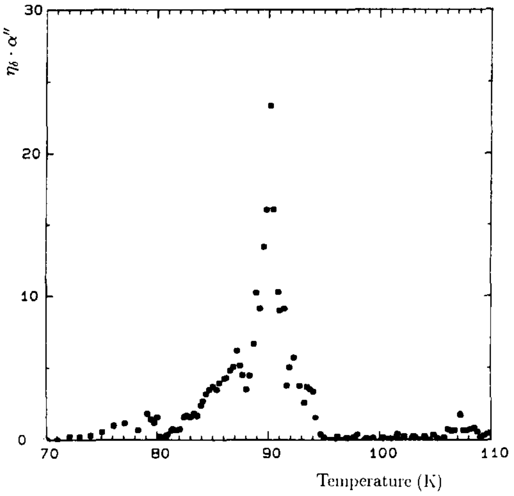

Fig. 2. The temperature-dependence of the imaginary part of the electric polarizability of the bound charges (α'') in oxygen deficient $YBa_2Cu_3O_{7-\delta}$. Since the polarizability could be measured only within the skin depth, the absolute values of α'' are reduced by a prefactor η_δ which gives the fraction of the effective contributing (skin-)volume divided by the sample volume (see text).

the domains and the mobility of the domain walls, the dynamics of generation and decay of the domains, depends on grain sizes as well as on the electric boundary conditions. Elastic behavior, on the other hand, is influenced by the dynamics of the domains and therefore can easily be modified by modifying the grain size (which gives an upper limit for the domain dimensions) and the electric boundary conditions. Besides these effects, the elastic behavior of classical ferroelectrics *with compensated polarization charges* is known to be influenced by the temperature-dependence of the dielectric function.

The aim of the work presented here was therefore to show that in good conducting YBCO, the temperature-dependence of the elastic moduli is influenced by the dielectric behavior and that this influence depends sensitively on intergranular contacts.

3.1. Elastic behavior of a classical ferroelectric

Seignette salt ($C_4H_4KNaO_6 \cdot 4H_2O$) is known to undergo a transition from a high-temperature paraelectric phase into an electrically polarized state at $T^* \sim 294$ K, and to transform into another paraelectric low-temperature phase at $T^{**} \sim 254$ K. Because large single crystals are easily grown, because the transition temperatures lie in an easily accessible temperature interval and because the temperature-dependent behavior of the dielectric function could be determined by conventional techniques, this system was chosen to demonstrate the influence of the dielectric behavior on the elastic properties of the system. As is obvious from Fig. 3, the elastic behavior depends sensitively on the electric state of the system. Figure 3 shows the temperature-dependent behavior of an acoustic resonance frequency for a standing acoustic wave (which is proportional to the acoustic phase velocity v_P) during cooling for an electrically isolated crystal and a crystal covered with gold and therefore electrically short-circuited. The most prominent feature is the pronounced elastic softening in the vicinity of the Curie temperatures in the gold-covered crystal which is absent in the case of an isolated crystal.

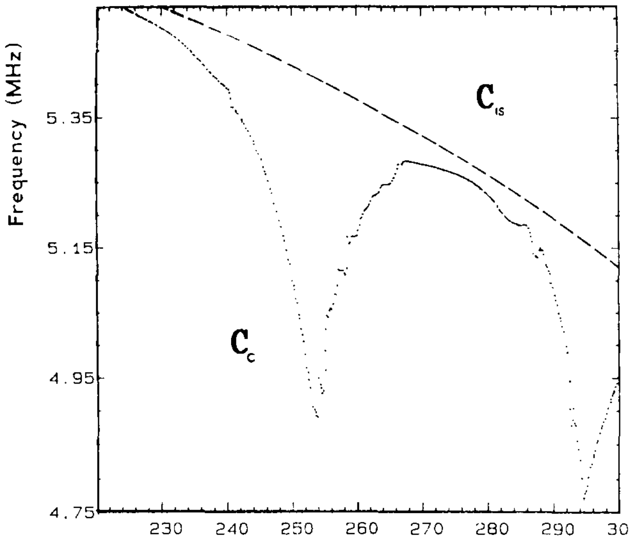

Fig. 3. The temperature-dependence of the resonance frequency for a standing acoustic wave in isolated (C_{is}) and short-circuited (C_c) Seignette salt. The behavior is the same for heating and cooling.

This behavior can be described by means of a Landau-type theory for second-order phase transitions by introducing as two coupled order parameters electric polarization and elastic strain. By this the elastic behavior as consequence of electric polarization can be described.

If only piezoelectric coupling is taken into account, the expression for the elastic modulus C_c of a system with compensated polarization charges reads as

$$C_c = C_{is} - f^2(\epsilon - 1) \tag{5}$$

Here f is the piezoelectric coupling constant and ϵ is the temperature-dependent dielectric function, while C_{is} is the mainly temperature-independent elastic modulus in the case of non-compensated polarization charges. Note that the elastic modulus C is connected with the mass density ρ of the material and the sound velocity v via $C = \rho v^2$.

3.2. Elastic behavior of $YBa_2Cu_3O_{7-\delta}$

Ultrasonic investigations of the elastic behavior of YBCO yield differing results, depending on the samples used, and many seemingly contradictory assertions even from the same group [8, 9] about the temperature-dependence, especially in the vicinity of the superconducting transition temperature T_c, are reported. Consequently, this problem of comparability of experimental results was often *solved* in defining *good* and *bad* samples.

None the less, it can be shown that these seemingly contradictory results are understandable when taking into account the important influence of the electric boundary conditions on the elastic behavior of a dielectric also in the case of YBCO. Having in mind the increasing number of reports about anomalous dielectric behavior of this high-T_c system in the vicinity of the superconducting transition temperature [4, 10–13], and regarding the large anisotropy of the electric conductivity as well as the varying and usually low electrical intergrain contacts, it is not hard to assume that the temperature-dependent elastic behavior should depend not only on grain size (as in conventional dielectrics – and indeed as reported for YBCO [14]) but also on the actual quality of the intergrain contacts. This could be seen nicely in the experiment displayed in Fig. 4. While a freshly prepared pressed (but not sintered) YBCO powder sample shows a clear step-like anomaly in the

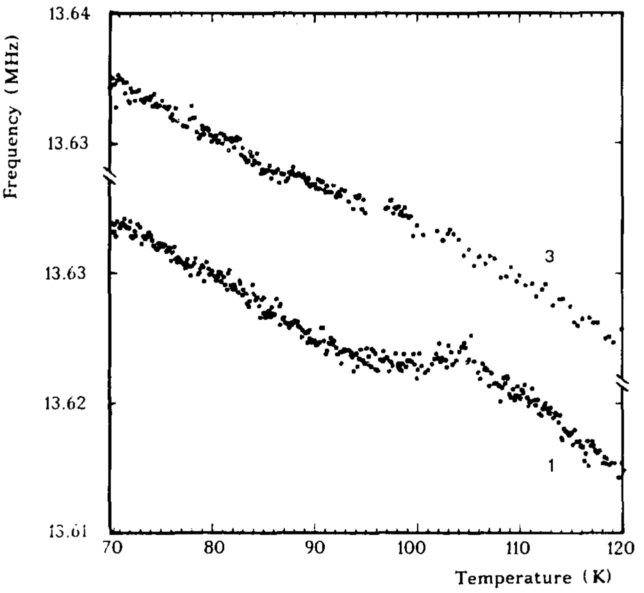

Fig. 4. The temperature-dependence of the resonance frequency of a standing acoustic wave in a freshly prepared YBCO sample during the first (1) and the third (3) heating run. The step-like anomaly in the resonance frequency disappears by thermally cycling the sample.

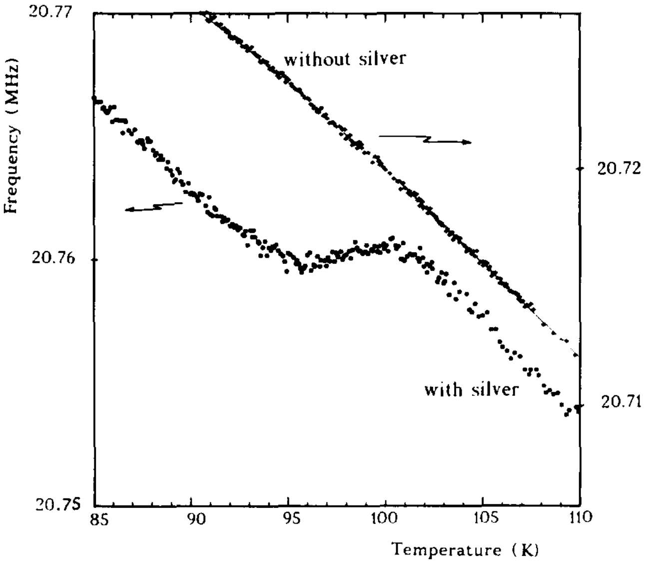

Fig. 5. The temperature-dependence of the resonance frequency of a standing acoustic wave in a pressed sample with low electric intergranular contact (*without silver*) and with artificially enhanced electric intergranular contact (*with silver*).

temperature-dependence of the sound velocity above T_c (curve 1), already two runs later, the step is no longer visible (curve 3). Since this behavior clearly demonstrates the great importance of the intergranular contacts for elastic behavior, and these intergranular contacts decrease in quality during temperature cycles, in a further experiment pressed powder samples with increased and stabilized intergranular contacts were investigated. The samples were prepared from the same batch of high quality YBCO powder. Both sample types did not experience any heat treatment after pressing.

The results of the investigations are given in Fig. 5. The temperature-dependence of the sound velocity of loosely pressed (*without silver*) samples is compared with the one of well-pressed samples with artificially enhanced intergranular contacts (*with silver*). The step-like behavior of the sound velocity of samples with increased intergranular contacts is very reminiscent of the behavior of freshly pressed YBCO powder in the first run (see Fig. 4) but is stable against temperature-cycling.

4. Conclusion

It was shown that, similar to what is known from *conventional* ferroelectrics, in polycrystalline samples of the high-T_c superconductor YBCO the temperature-dependent elastic behavior is influenced by the temperature-dependence of the dielectric function. An elastic anomaly above T_c only in samples with artificially enhanced intergranular contact can be understood to be a consequence of the anomalous dielectric behavior reported for YBCO above the superconducting transition temperature [4, 13]. Besides the fundamental importance of understanding the dielectric properties of these high-T_c systems, reports of contradictory elastic behavior of different samples of YBCO (sintered compared with melt-textured samples as well as single crystals) could now be understood as resulting from the difference in the electrical *intrasample* conductivity, *e.g.* the different intergranular conductivity in sintered specimens (where the highly anisotropic resistive behavior plays an important role), and the different conductivities across domain walls in single crystals. These electric boundary conditions modify the influence of the permittivity on the temperature-dependent elastic behavior.

Due to the limited space, it should only be mentioned that the dependence of the elastic behavior of YBCO on the electric boundary conditions can be qualitatively understood in terms of a Landau-like theory for phase transitions with two coupled order parameters (electric polarization and elastic strain) when piezoelectric as well as electrostrictive coupling is taken into account [15].

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