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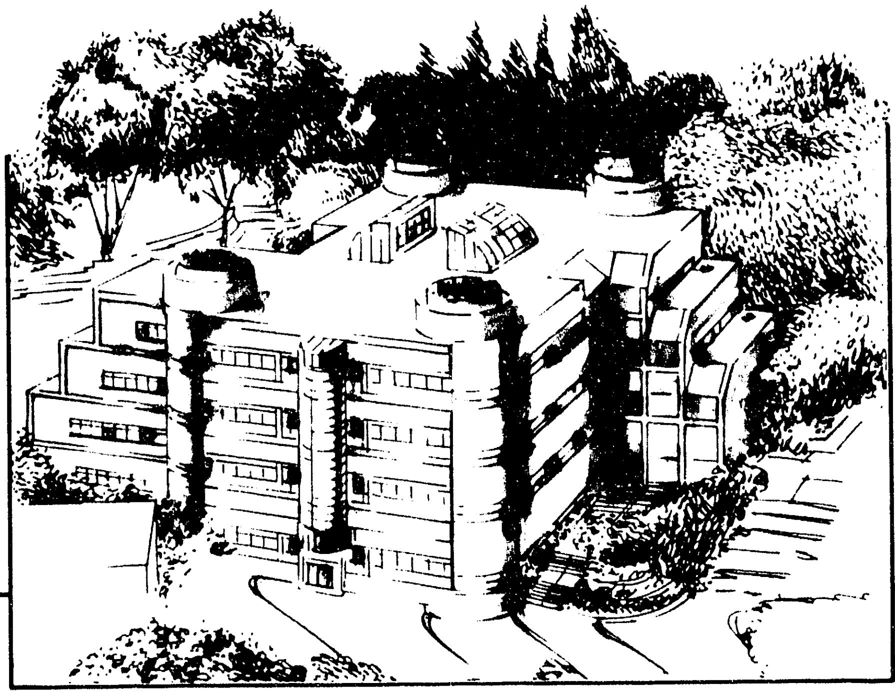

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ABSTRACT - We have used a bolometric technique to obtain accurate low temperature loss data for epitaxial thin films of $Ba_{0.6}K_{0.4}BiO_3$ from 30 cm^{-1} to 700 cm^{-1}. These films were grown on MgO and $SrTiO_3$ substrates by MBE, off-axis sputtering and laser deposition techniques. All films show a strong absorption onset near the BCS tunneling gap of $3.5k_BT_c$. We have analyzed these data using a Kramers-Kronig transformation and have corrected for finite film thickness effects. Our results indicate that the absorption onset is consistent with a superconducting energy gap. Comparison is made with predictions based on strong coupling Eliashberg theory using $\alpha^2F(\omega)$ spectra obtained from the literature. While we are able to fit the overall measured absorptivity, we are unable to fit the structure observed in our data.

Keywords: high temperature superconductivity, thin films, infrared spectroscopy, BKBO

INTRODUCTION

The oxide superconductor $Ba_{1-x}K_xBiO_3$ (BKBO) is an interesting system because of its differences to other high-T_c materials. BKBO has a cubic structure in contrast to the cuprate superconductors, which are highly anisotropic. The parent compound of BKBO shows charge density wave ordering[1] rather than the antiferromagnetic ordering seen for example in the parent compound of $YBa_2Cu_3O_{6+x}$ (YBCO). Tunneling measurements have shown evidence for an energy gap and a BCS density of states in BKBO,[2-4] but not in YBCO. Reflectivity measurements on BKBO thin films show features which have been interpreted as a superconducting gap.[5] In this work we have measured the absorptivities of three BKBO films with x ~ 0.4, listed in Table 1. The films were grown by molecular beam epitaxy,[6] pulsed laser deposition[7] and off-axis magnetron sputtering.[8]

	Institution	Growth	Thickness	Substrate	T_c
A	AT&T	MBE	350 nm	MgO	15 (20) K
B	U. Illinois	Laser Deposition	300 nm	MgO	22 (28) K
C	H.P.	Off-axis Sputtering	400 nm	$SrTiO_3$	19 (21) K

Table 1: List of samples used in this study. T_c values correspond to the low temperature end of the transition as measured by an ac susceptibility technique. The onset of the transition is given in parentheses.

EXPERIMENT

We measure the submillimeter absorptivity from 30 cm^{-1} - 700 cm^{-1} by using the high-T_c film as the absorber in a composite bolometric detector. A thermometer is attached to the back of the substrate to measure absorbed power. The incident power is normalized against an absorber of known optical properties. This technique has previously been used to measure the absorptivities of $YBa_2Cu_3O_7$, [9,10] $Tl_2Ca_2Ba_2Cu_3O_{10}$ and $Tl_2CaBa_2Cu_2O_8$ thin films.[11] In general, this technique is more accurate than reflectivity measurements when the sample reflectivities are close to unity.

The measured absorptivities for the three films are displayed in Fig. 1. The BKBO absorptivities are larger than for a high quality epitaxial YBCO film, with x~ 1, which is also shown in Fig. 1 for comparison. Large sample-to-sample variations are present among the three BKBO films. However, there are similar structures present for all films. There are steps in the absorptivity at 525 cm^{-1} and near 300 cm^{-1}. There is also a strong absorption onset observed for all films near the tunneling gap at $2\Delta = 3.5\ k_BT_c$.[2] However, the frequencies of the observed absorption onsets do not scale with the measured T_c values as listed in Table 1.

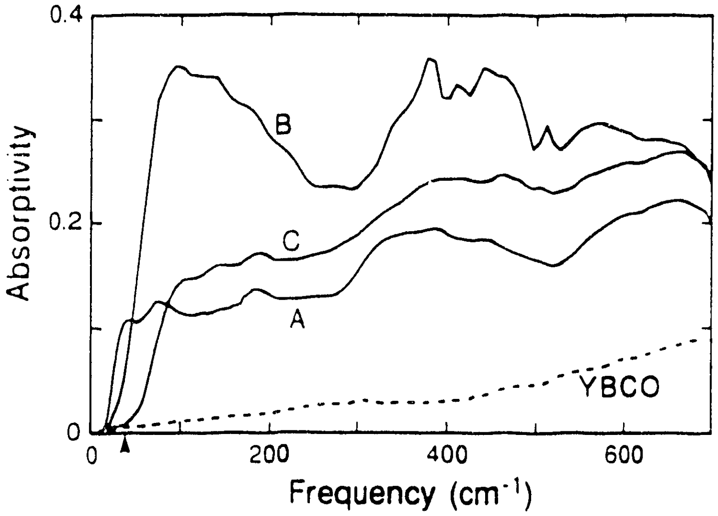

Fig. 1: Measured absorptivities of films A, B and C at 2K. The measured absorptivity of a $YBa_2Cu_3O_7$ film (dotted line) is also shown for comparison. The arrow indicates the approximate tunneling gap value of $2\Delta = 3.5\ k_BT_c$ for film A. The frequencies of the observed absorption onsets do not scale with the measured T_c values as listed in Table 1.

ANALYSIS

Two distinct physical phenomenon, a zero crossing of $\varepsilon_1(\omega)$ and an onset in $\sigma_1(\omega)$ can give rise to an onset of absorption. The former corresponds to a plasma edge; the latter to a superconducting gap. For example, an onset is observed in the absorption of polycrystalline $La_{1.85}Sr_{0.15}CuO_4$ near 50 cm^{-1} which is due to a plasma edge. Historically this feature was confused with a superconducting gap.[12] In order to understand the significance of the low frequency absorption onset it is necessary to obtain information about $\varepsilon_1(\omega)$ and $\sigma_1(\omega)$.

We can estimate the London penetration depth in BKBO as $\lambda_L = c/\omega_p \sim 125$ nm, where c is the speed of light and $\omega_p = 1.59$ eV is the optically determined plasma frequency.[13] Because the films used in this study are only slightly thicker than λ_L we expect that some radiation will be transmitted through the films. Our data contain fringes resulting from interference of waves reflected from the front and back surface of the substrate. We average these fringes which are typically only a few percent of the absorptivity. This is equivalent to neglecting the effects of reflections from the back surface. These data are then compared to a theoretical model which assumes that the substrate thickness is infinite.

We compute the Kramers-Kronig (KK) transform of our data. In order to extend our data for film A to high frequency we use data measured up to 5 eV on a nominally identical sample.[14] We use the results of the KK transform above ~30 cm^{-1}, which are independent of the low frequency extrapolation used. The pseudo-dielectric function $<\varepsilon(\omega)>$ is obtained from the Kramers-Kronig transform of the measured absorptivity, and includes effects of the film/substrate interface. We use the well known expression for the optical properties of a thin film on an infinite dielectric[15] to relate $<\varepsilon(\omega)>$ to the dielectric function $\varepsilon(\omega)$ of the BKBO, the known BKBO film thickness and the known optical properties of the substrate. The unknown quantity $\varepsilon(\omega)$ is solved for numerically. In what follows we describe our results for film A. Similar results are obtained for film B, also grown on an MgO substrate for which the FIR properties are described in the literature.[16] Film C, deposited on a $SrTiO_3$ substrate , poses a special problem, as the low temperature (~4K) FIR properties of this ferroelectric material are not adequately described in the literature.

RESULTS OF ANALYSIS

The pseudo- functions $<\sigma_1(\omega)>$ and $<\varepsilon_1(\omega)>$ and the inferred functions $\sigma_1(\omega)$ and $\varepsilon_1(\omega)$ for BKBO (with and without finite film thickness correction) are shown in Fig. 2 as dotted and solid lines, respectively. The pseudo-dielectric function $<\varepsilon_1(\omega)>$ in Fig. 2(a) crosses zero near the frequency of the low frequency absorption onset. However $\varepsilon_1(\omega)$, which is corrected for the finite film thickness, does not cross zero near the absorption onset, indicating that the absorption onset is not a plasma edge, and may be a superconducting gap. The presence of a zero crossing in $\varepsilon_1(\omega)$ at 400 cm^{-1} , which is near a strong absorption feature in the MgO substrate, may come from an error in our technique for correcting finite film thickness effects. The low frequency onset in the pseudo-conductivity $<\sigma_1(\omega)>$ also becomes more steep in $\sigma_1(\omega)$, as shown in Fig. 2(b).

The absorptivity of BKBO (corrected for the finite film thickness) is shown in Fig. 3(a) as a dotted line. The measured absorptivity for the same sample, not corrected for finite film thickness effects, is shown again for comparison as a dashed line. The feature near 400 cm^{-1} may be due to the strong absorption in the MgO substrate. The remaining structure appears to be a real property of BKBO. It is useful to investigate whether the observed structure is due to phonons via the Holstein effect,[17] expected in a strong coupling superconductor. In order to explore this question we compare our data to an Eliashberg strong coupling calculation.[18] We use the

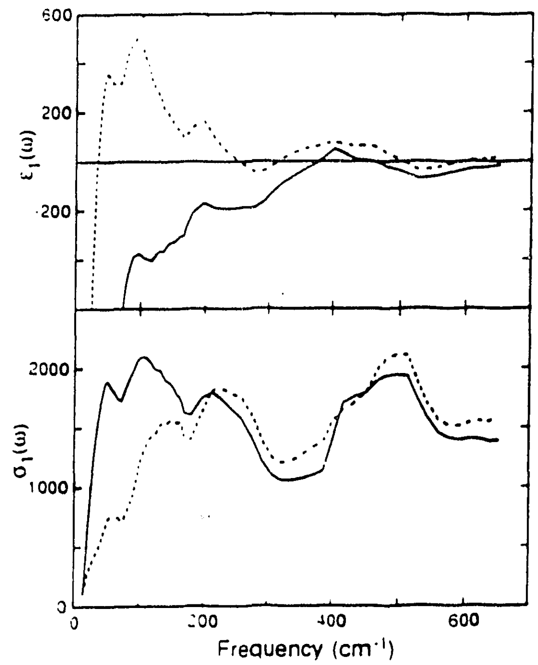

Fig. 2: (a) Pseudo-dielectric function $<\varepsilon_1(\omega)>$ (dotted line) and inferred dielectric function $\varepsilon_1(\omega)$ (solid line) for film A. The pseudo-dielectric function is derived from the Kramers-Kronig transform of the measured absorptivity and is therefore not corrected for finite film thickness effects. The inferred dielectric function $\varepsilon_1(\omega)$ is corrected for the finite film thickness. The feature near 400 cm^{-1} may be due to a strong absorption band in the MgO substrate, which we are unable to remove from our data. (b) Pseudo-conductivity $<\sigma_1(\omega)>$ (dotted line) and inferred conductivity $\sigma_1(\omega)$ (solid line) for film A.

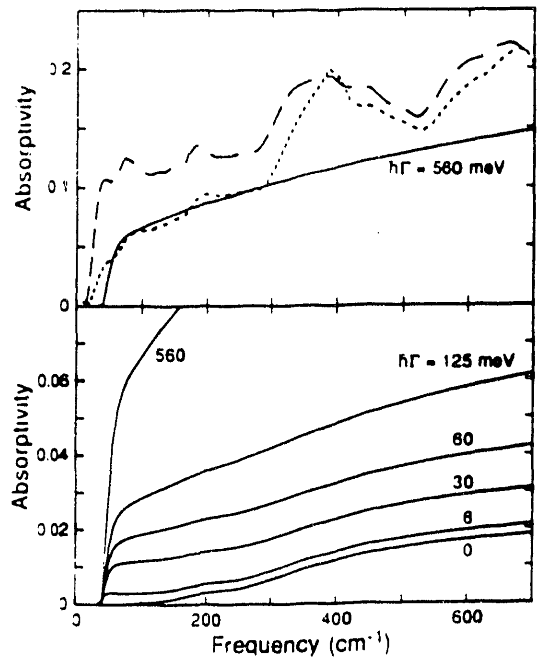

Fig. 3: (a) The dotted line is the absorptivity of film A corrected for finite film thickness effects. The feature near 400 cm^{-1} may be due to a strong absorption band in the MgO substrate, which we are unable to remove from our data. The uncorrected absorptivity for sample A is also shown for comparison as the dashed line. The solid line is a fit to the data using the strong coupling Eliashberg theory with $\omega_p = 1.59$ eV, $\varepsilon_\infty = 3.8$ and $\hbar\Gamma = 560$ meV. (b) Results of the strong coupling Eliashberg theory with $\omega_p = 1.59$ eV, $\varepsilon_\infty = 3.8$ and $\hbar\Gamma = 0, 6, 30, 60, 125$ and 560 meV. Note that as $\hbar\Gamma$ increases, the Holstein structure due to phonon modes is washed out.

experimental plasma frequency ω_p = 1.59 eV[13], $\alpha^2F(\omega)$ from tunneling measurements[3] and various values of the impurity scattering rate $\hbar\Gamma$, as shown in Fig. 3(b). In the clean limit ($\hbar\Gamma$ = 0) no absorption onset is observed. As the impurity scattering rate increases, an absorption onset appears. Structure present in the calculated absorptivity due to phonon modes gets washed out as the scattering rate increases. In order to fit the absolute value of the absorptivity, the inelastic scattering rate used in the Eliashberg calculation is so large ($\hbar\Gamma$ = 560 meV) that all features due to electron-phonon coupling are washed out. Inelastic scattering rates determined from reflectivity measurements range from 250 meV[5] to 900 meV.[13] The structure we observe in the measured optical properties which the model is not able to fit may be due to the presence of insulating regions in the film.[19]

CONCLUSIONS

We have directly measured the FIR absorptivity for three $Ba_{0.6}K_{0.4}BiO_3$ films at 2K . The measured absorptivities are significantly larger than for $YBa_2Cu_3O_7$ at the same temperature. In addition, there is considerable sample-to-sample variation among the films. We observe absorption onsets near 3.5 k_BT_c (~40 cm^{-1}) for all films which do not scale with T_c, as well as pronounced structure at higher frequencies. After performing a Kramers-Kronig analysis and correcting for finite film thickness, we find that the absorption onset is consistent with a superconducting gap. Further measurements using thick films or single crystals are necessary to eliminate uncertainties associated with the finite film thickness correction. A strong coupling Eliashberg calculation does not account for the structure observed in the optical properties.

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