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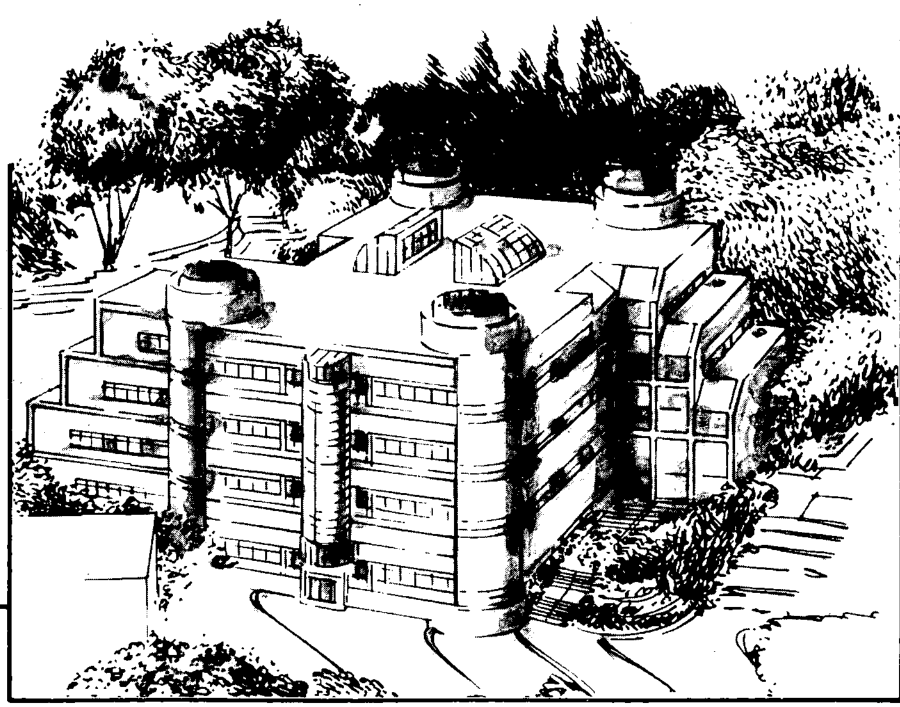

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Abstract

We have used both bolometric and cavity techniques to obtain accurate submillimeter and microwave loss data for epitaxial thin films of $Tl_2Ca_2Ba_2Cu_3O_{10}$ at low temperatures. These films have T_c =121.5 K, are c-axis oriented, contain some volume fraction of the 2:1:2:2 phase, and are characterized by excellent in-plane epitaxy. The absorptivity of these films at 100 cm^{-1} is a factor of five lower than that obtained by others from a reflectivity measurement on a ceramic sample. We observe strong phonon structure for frequencies between 70 and 600 cm^{-1}, which are in agreement with a lattice dynamical calculation. Our results show remarkably similar phonon structure to that observed in the ceramic sample. This is in strong contrast to the case for other high T_c superconductors such as $YBa_2Cu_3O_7$, where phonon structure observed in ceramic samples is absent in epitaxial oriented films and crystals because of the electronic screening due to the high conductivity of the a-b planes. At microwave frequencies the absorptivity follows a frequency squared dependence, and is consistent with the submillimeter results.

We have studied ~ 5000Å thick epitaxial films of nominal composition $Tl_2Ca_2Ba_2Cu_3O_{10}$ which are grown on $LaAlO_3$ substrates. We have used a novel technique for directly measuring the submillimeter losses of the films between 30 and 700 cm^{-1} at 2 K.[1] In this technique, the high T_c film is used as the absorber in a composite bolometric detector for the signal from a Fourier transform spectrometer with a Hg arc source. The data are normalized by comparison with reference detectors with normal metal absorbers whose absorptivities can be calculated. This technique has previously been used to study the residual losses in high quality epitaxial films of YBCO.[1] The losses in the films used for this study were also measured near 10, 30 and 90 GHz (0.3, 1 and 3 cm^{-1}) down to 10 K. Microwave losses at 10 GHz were measured with a parallel plate resonator technique.[2] Microwave losses at 30 and 90 GHz were measured with a confocal resonator technique.[3]

The films used in this study were sputter-deposited onto (1 0 0) $LaAlO_3$ substrates from sintered targets prepared from oxide powder mixtures with an initial cation ratio of 2Tl:2Ca:2Ba:3Cu. These amorphous precursor films were subsequently wrapped with 2:2:2:3 pellets and post-annealed at ~ 850 °C in order to minimize Thallium loss. This technique has been described previously.[4] The zero resistance superconducting transition temperature for these films was measured to be T_c = 121.5 K. Film microstructure for one of the samples (L2) was investigated using a four circle x-ray diffractometer. Within experimental uncertainty, the x-ray $\theta/2\theta$ scan shown in Fig. 1indicates that for this film as much as 70% but as little as 30% of the oriented material is in the 2:2:2:3 phase, with the remainder of the oriented material in the 2:1:2:2 phase, where the c-axis is perpendicular to the substrate surface. X-ray $\theta/2\theta$ scans on similar films indicates that as much as 80% of the oriented material may be in the 2:2:2:3 phase. However, the relatively high background suggests the presence of some volume fraction of amorphous or randomly oriented polycrystalline grains. The x-ray φ scan shown in Fig. 2 is obtained by aligning the x-ray source and detector to observe the (1 0 15) peak of the 2:2:2:3 phase and rotating the film about the axis normal to the substrate. This scan indicates that the c-axis oriented 2:2:2:3 phase material in these films is in excellent registry with the $LaAlO_3$ substrate. An x-ray φ scan using the (1 0 5) peak of the 2:1:2:2 phase also indicates that the c-axis oriented 2:1:2:2 phase material is in good registry with the $LaAlO_3$ substrate.

The submillimeter absorptivity spectra for samples L1 and L2 are shown in Fig. 3 and replotted again in Fig. 4 (solid lines) along with the microwave loss data (filled circles) measured for the same films near 10 K. The curves in Fig. 4 are displaced by factors of 10 to avoid overlap. Losses at low frequencies increase as frequency squared between 10 and 90 GHz and are in good agreement with submillimeter data for both films. The dotted lines in Fig. 4 are frequency squared best fits to the microwave data. The submillimeter spectra contain many phonon modes between 50 and 700 cm^{-1}. Such phonon structure has previously been observed experimentally in the sintered ceramic sample of Zetterer *et al.*, which is plotted in Fig. 3.[5] Our films exhibit remarkably similar structure to that observed in the ceramic sample between 50 and 700 cm^{-1}. Therefore the agreement with lattice vibration calculations found for the phonon structure from the sintered ceramic[5] should also be valid for the structure we measured on the films. However, the overall absorptivity of the epitaxial films is lower than for the ceramic samples.

We expect the high conductivity of the a-b plane to screen out phonon excitations with vibrational motion in the a-b plane. We therefore believe that the phonon modes observed in epitaxial films L1 and L2 correspond to infrared active excitations perpendicular to the a-b plane. The observed phonon structure is in agreement with a lattice dynamical study which show the existence of 8 phonon modes for 2:2:2:3 phase material with $E \parallel c$.[6] Since the incident fields probing the sample in our measurement are predominantly parallel to the surface of the film, the plausible identification of the observed structure to $E \parallel c$ phonon modes suggest the presence of a substantial fraction of the film containing grains whose c-axis is not perpendicular to the film surface. This is consistent with the observation of a large background in the x-ray $\theta/2\theta$ scan, and demonstrates that absorptivity on these highly anisotropic materials are sensitive to the quality of film epitaxy. The x-ray data and the similarity of the submillimeter spectra for the ceramic and film samples suggest that the films used in this study contain well oriented epitaxial grains of both the 2:2:2:3 and the 2:1:2:2 phases in addition to amorphous or randomly oriented polycrystalline grains.

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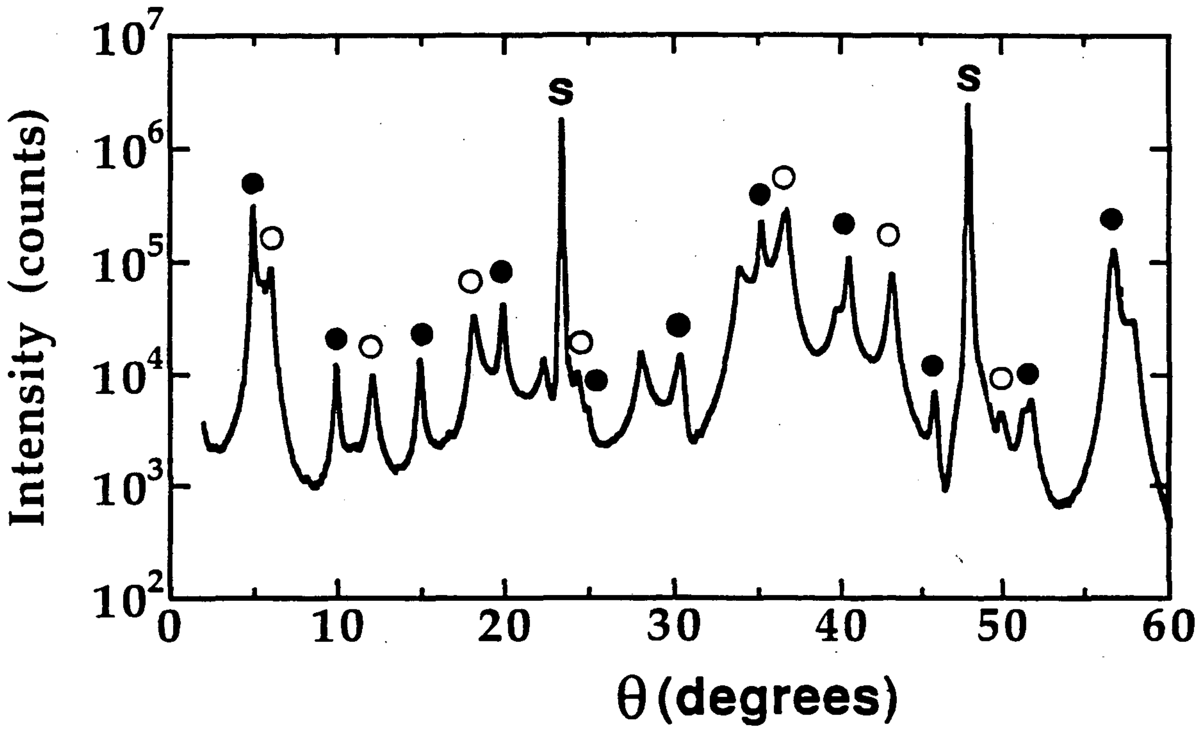

Fig. 1. X-ray θ/2θ scan of the epitaxial nominal $Tl_2Ca_2Ba_2Cu_3O_{10}$ film L2, indicating the presence of both the 2:2:2:3 (filled circles) and the 2:1:2:2 (open circles) phases. Substrate peaks are indicated by "s". The high background counts suggest the presence of some volume fraction of amorphous or randomly oriented polycrystalline grains.

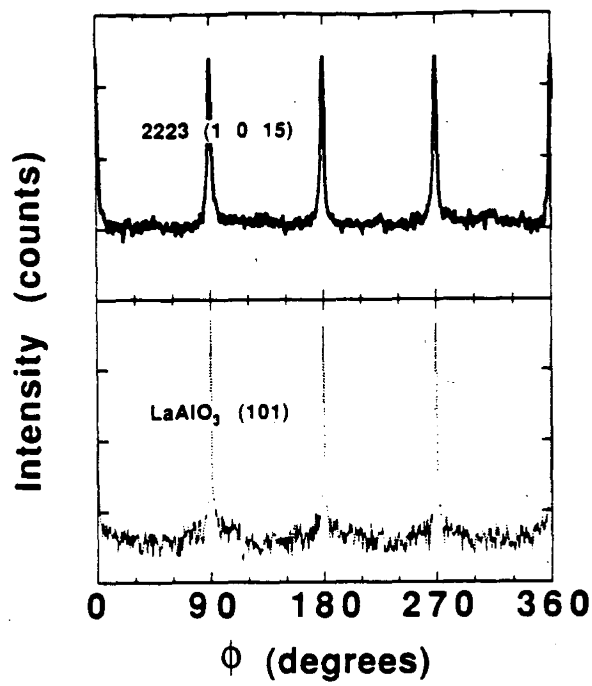

Fig. 2. X-ray φ scan using the (1 0 15) peak of $Tl_2Ca_2Ba_2Cu_3O_{10}$ for the epitaxial film L2, indicating excellent in plane registry of the oriented 2:2:2:3 phase (upper panel). For comparison, a φ scan using the (1 0 1) peak of the bare $LaAlO_3$ substrate is shown (bottom panel).

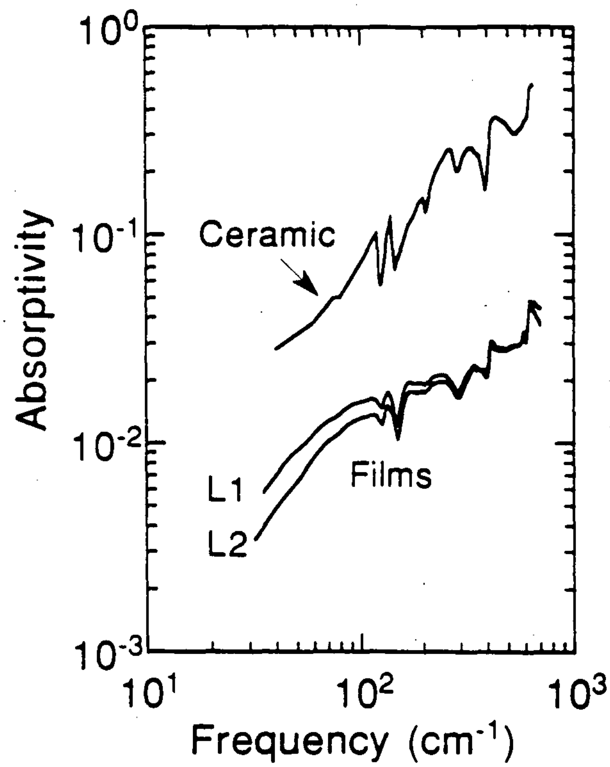

Fig. 3. Submillimeter absorptivity spectra for the epitaxial $Tl_2Ca_2Ba_2Cu_3O_{10}$ samples L1 and L2 plotted logarithmically. Also shown is a reflectivity measurement of a $Tl_2Ca_2Ba_2Cu_3O_{10}$ sintered ceramic sample plotted as an absorptivity. All three data sets show 8 phonon modes, in agreement with calculations for 2:2:2:3 phase material with $E \parallel c$.

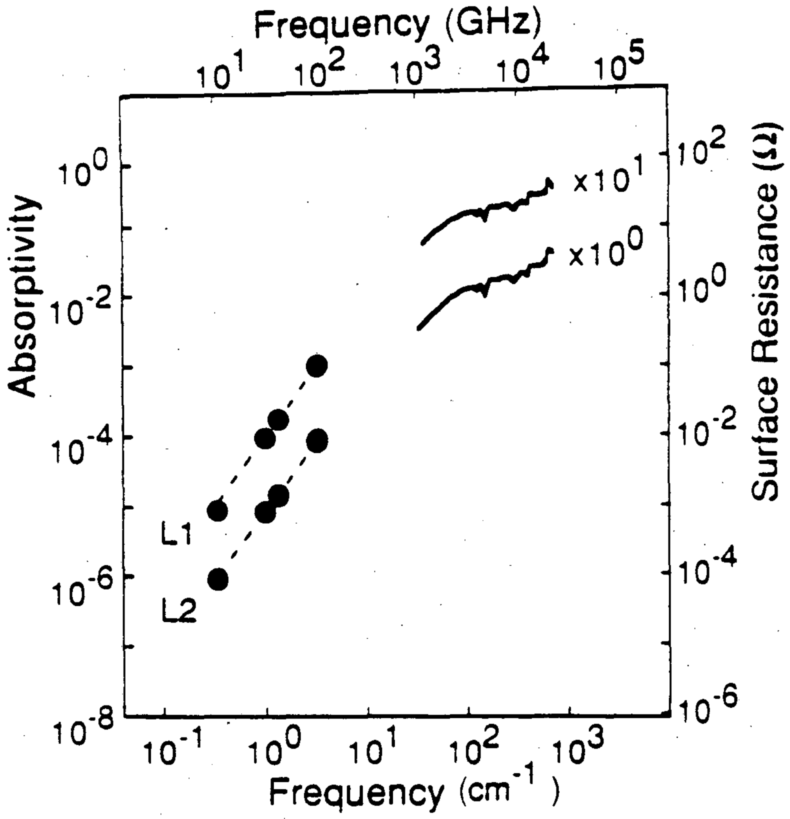

Fig. 4. Measured submillimeter absorptivities at 2K for the epitaxial $Tl_2Ca_2Ba_2Cu_3O_{10}$ films L1 and L2 multiplied by the indicated factors to separate the curves. Values of the microwave surface resistance measured for each sample near 10K between 10 and 90 GHz are shown as filled circles. The dotted lines are frequency squared best fits to the microwave data.

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