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Title: TERAHERTZ EMISSION FROM YTTRIUM BARIUM COPPER
OXIDE THIN FILMS

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Terahertz Emission from $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin films.

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Abstract

We report the first observation of terahertz emission from unbiased $\text{YBa}_2\text{Cu}_3\text{O}_7$ thin films at room temperature excited by 1.5 eV femtosecond laser pulses. The strength of the radiated terahertz field depends on the incident angle (θ) and depends linearly on incident power. The terahertz emission also depends strongly on the rotation of the film about its surface normal (azimuthal angle, ϕ). The strong anisotropy, with 4ϕ symmetry is demonstrated for different polarizations (incident and detected) and found to be consistent with optical rectification. Because YBCO is centrosymmetric, the second order nonlinear susceptibility vanishes. The next order term in the nonlinear polarizations is the electric quadrupole source term which describes the observed data quite well. The terahertz radiation was found to decrease with decreasing oxygen content, indicating that the carriers in the doped sample enhance the nonlinearity responsible for the terahertz emission.

Keywords: Terahertz generation, YBCO, ultrafast nonlinear optics.

Introduction

The generation of subpicosecond, terahertz bandwidth radiation via the illumination of nonlinear materials with visible femtosecond pulses is of interest both as a spectroscopic source and as a probe of the electronic and vibrational properties of nonlinear materials. Such terahertz emission experiments are based on the optical excitation of coherent phenomena in a material and the temporal evolution of the far-infrared dipole moment associated with the excitation. Previous studies have included terahertz emission from semiconductors

such as GaAs and InP where such emission is a result of optical rectification [1] in the material and/or the evolution of a transient photogenerated current.[2, 3] Using this technique the effects of magnetic and electric fields on the emission were also studied.[4, 5] The dynamics of optically excited wavepackets in quantum well structures have also been studied using terahertz emission spectroscopy, leading to a demonstration of the existence of Bloch oscillations in superlattices.[6] Finally, terahertz emission spectroscopy has led to the detection of coherent infrared phonons in tellurium,[7] as well as the detection of coherent atomic vibrations in molecular crystals.[8] Terahertz radiation has recently been detected from biased $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin-film antennas at temperatures below T_c , [9] and from unbiased YBCO samples ($T < T_c$) that have been exposed to a magnetic field.[10] In this paper, we describe not only the first observation of terahertz emission from unbiased $\text{YBa}_2\text{Cu}_3\text{O}_7$ thin films at room temperature, but also the first clear observation of terahertz generated by optical rectification due solely to the electric quadrupole source term. Second harmonic generation from an electric quadrupole source in cubic crystals has been studied both experimentally,[11, 12] and theoretically.[13]

Experimental Setup

The experimental arrangement is a standard setup used for terahertz spectroscopy. The incident optical beam is provided by a Ti:sapphire chirped pulse amplifier laser system producing 1 mJ, 800 nm, 150 fs pulses at a repetition rate of 1 kHz. This beam is split into two pulses: one pulse is used to excite the sample, while the other is used to gate a photoconducting detector. A chopper rotating at 200 Hz is located in the excitation beam path so that synchronous detection can be used. The resultant terahertz emission is collected in transmission

with a pair of gold coated parabolic mirrors and focused onto a photoconducting detector consisting of 2.5 mm gap electrodes deposited on radiation-damaged silicon-on-sapphire with a temporal response of about 1 ps. An optical delay line in the gating pulse pathway is used to measure the temporal profile of the radiated terahertz pulse. Approximately 75% of the incident optical beam is absorbed in the film, while terahertz transmission experiments indicate that about 85% of the incident terahertz radiation is absorbed by the film. A wire grid polarizer is used to ensure detection of either P or S-polarized terahertz.

The sample consists of a ~ 300 nm thick, c-axis oriented, twinned $\text{YBa}_2\text{Cu}_3\text{O}_7$ (YBCO) thin film epitaxially deposited on a $10 \times 10 \times 1$ mm MgO (100) substrate using DC off-axis sputtering.[14] The critical temperature is measured to be 88 K, but all terahertz emission measurements were performed at room temperature where the film is metallic, rather than superconducting. The films were analyzed by X-ray diffraction and were found to be very well aligned with the c-axis normal to the surface and the a,b axes aligned with the MgO cube axes. For comparison, we made many of the same measurements on a 0.5 mm thick InP (100), semi-insulating, Fe doped sample using the same geometry and experimental conditions as those used for the YBCO film.

Results

We measured terahertz radiation emitted from a YBCO film for four different polarization geometries; incident radiation (800 nm) polarized in the plane of incidence (P-polarized) and perpendicular to the plane of incidence (S-polarized), and detecting P-polarized and S-polarized terahertz radiation. Terahertz waveforms emitted from $\text{YBa}_2\text{Cu}_3\text{O}_7$ are shown in Fig. 1 for the four different polarization combinations. The greatest THz signal is detected with the incident pulses polarized in the plane of incidence (P-polarized) and detecting P-polarized THz. When S-polarized radiation is incident on the film, no THz (P or S-polarized) is detected at $\phi = 0^\circ$, (Fig. 1) but as the film is rotated about its normal to $\phi = 22^\circ$, (Fig. 2) both S and P-polarized THz radiation is detected. The orientation dependence of the THz radiation is discussed in more detail below.

The THz field radiated from the YBCO film depends linearly on the incident 800 nm power (Fig. 3), in other words, the THz intensity depends quadratically on the incident power. The incident laser pulses were focused to a spot size on the film of ~ 2 mm across, giving a fluence of ~ 6 mJ/cm² and a photo-carrier density of $\sim 6 \times 10^{21}$ cm⁻³ at 100 mW. Terahertz radi-

Figure 1: Terahertz waveforms from $\text{YBa}_2\text{Cu}_3\text{O}_7$ at $\theta = 50^\circ$, $\phi = 0^\circ$ for a) S-polarized incident and S-polarized detected radiation, b) S-polarized incident, P-pol. detected, c) P-pol. incident, S-pol. detected, and d) P-pol. incident and P-pol. detected.

Figure 2: Terahertz waveforms from $\text{YBa}_2\text{Cu}_3\text{O}_7$ at $\theta = 50^\circ$, $\phi = 22^\circ$ for a) S-polarized incident and S-polarized detected radiation, b) S-polarized incident, P-pol. detected, c) P-pol. incident, S-pol. detected, and d) P-pol. incident and P-pol. detected.

Figure 3: The radiated THz field depends linearly on the incident 800 nm power. The spot size on the film had a diameter of ~ 2 mm.

ation emitted from an InP (100) semi-insulating wafer was approximately 5 times stronger than the THz generated by the YBCO film with the same experimental conditions ($\theta = 50^\circ$, incident power of 90 mW).

Fig. 4 shows that the THz radiation depends strongly on incident angle and disappears at normal incidence ($\theta = 0^\circ$). The bottom graph in Fig. 4 shows the same data but its plotted as a function of the internal angle, θ_f . The internal angle was calculated using an index of refraction of 2.0 for YBCO at room temperature for 800 nm light.[15] The data was taken with the incident light P-polarized and detecting P-polarized THz (PP) The solid curves in Fig. 4 are fits of the data to

$$E_{PP}(\theta) = A \sin(2\theta) + B \sin(4\theta), \quad (1)$$

which is predicted by optical rectification from the bulk electric quadrupole term for a twinned orthorhombic crystal. The dependence of the emitted terahertz radiation on sample orientation (rotation of the sample about its normal, ϕ) for the four polarization geometries is shown in Fig. 5 where $\theta = 50^\circ$ and the incident power is 90 mW. The solid curves are fits to functions obtained from the bulk electric quadrupole contribution to optical rectification:[11, 13]

$$E_{PP}(\phi) = A_1 \cos(4\phi) + C_1, \quad (2)$$

$$E_{SP}(\phi) = -A_2 \cos(4\phi) + C_2, \quad (3)$$

$$E_{PS}(\phi) = A_3 \sin(4\phi), \quad (4)$$

Figure 4: Radiated THz field is plotted as a function of incident angle ($-\theta_i$) in the top figure and as a function of the internal angle, (θ_f), in the bottom figure. The solid circles represent data taken with P-polarized 800 nm incident and detecting P-polarized THz. The solid curves are fits, see text for more information.

Figure 5: The peak-to-peak THz field amplitude is plotted as a function of azimuthal angle (crystal orientation, ϕ). The data were taken with 90 mW of P- (filled symbols) or S-polarized (open symbols) 800 nm pulses incident on the film at $\theta = 50^\circ$ and detecting P- (circles) or S-polarized (squares) THz.

$$E_{SS}(\phi) = -A_4 \sin(4\phi), \quad (5)$$

where, A and C are constants that depend on the crystal structure and Fresnel coefficients. The formulas above were calculated for an orthorhombic crystal where the x and y axes are interchangeable to take into account the twin domains.

Varying the oxygen content in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ changes the density of carriers in the film, as well as changing T_c . We used three different films to look at the THz dependence on carrier concentration (Fig. 6) at room temperature. A very small THz signal was detected from an insulating film with $\delta = 0.8$ (not superconducting), slightly stronger emission was detected with $\delta = 0.5$ ($T_c = 55$ K), and the strongest THz signal was generated with $\delta = 0.0$ ($T_c = 88$ K). These results indicate that increasing the concentration of carriers enhances the mechanism responsible for the terahertz emission.

Figure 6: THz radiation from $\text{YBa}_2\text{Cu}_3\text{O}_{6.2}$, $\text{YBa}_2\text{Cu}_3\text{O}_{6.5}$ and $\text{YBa}_2\text{Cu}_3\text{O}_7$. $\theta = 50^\circ$, $\phi = 0^\circ$, and $T = 300$ K for all three measurements.

Conclusions

In semiconductors several mechanisms have produced terahertz emission via femtosecond optical excitation, including phonon generation,[7] difference frequency mixing from the bulk second order susceptibility (bulk optical rectification),[16] dipole radiation from a transient current of photoinjected carriers accelerated in a surface field (current surge),[2] and difference frequency mixing from an effective second order susceptibility proportional to the surface field (surface optical rectification).[1] In our experiment several of these mechanisms are not relevant. Our bandwidth is insufficient to detect coherent phonons, since the lowest infrared active phonon frequency is greater than 3 THz in YBCO. YBCO has a center of inversion symmetry, therefore its second order susceptibility vanishes and bulk optical rectification is not possible. The current surge mechanism is a possible candidate for the observed emission, with the driving field produced by the carrier gradient resulting from the photogeneration of carriers. However, it is unlikely that such a mechanism alone could produce the strong orientational dependence shown in Fig. 5. Electric quadrupole optical rectification in YBCO is consistent with the strong orientational dependence seen in the data. We calcu-

lated the dependence of the emitted THz radiation on incident angle and the complex dependence on crystal orientation for the different incident and detected polarizations, for a twinned orthorhombic crystal. We have demonstrated not only the first observation of terahertz emission from unbiased $\text{YBa}_2\text{Cu}_3\text{O}_7$ thin films at room temperature, but also the first clear observation of terahertz generated by optical rectification due solely to the electric quadrupole source term.

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