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Transient Conductivity in Single-Crystal Al_2O_3 Probed by THz Time-Domain Spectroscopy

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Abstract: THz time-domain spectroscopy is applied to probe photo-induced charge transport in single-crystal Al_2O_3. The conductivity of the material and its temporal evolution yield the scattering rate, plasma frequency, and dynamics. The temperature dependence indicates that LO phonons play an important role in the scattering process.

1. Introduction

Terahertz time-domain spectroscopy (THz TDS) is a powerful probe of charge transport and ultrafast carrier dynamics in condensed matter. The method is particularly useful when used in conjunction with an optical pulse that generates charge carriers [1,2]. In this work, we report initial results of the application of this technique to investigate transient conductivity over a range of temperatures (77K – 300K) in single-crystal Al_2O_3 (sapphire). Electronic transport in sapphire and other insulators, unlike the case for semiconductors and metals, is very difficult to observe under equilibrium conditions [3]. It can, however, be readily studied by ultrafast pump-probe measurements. Aside from its intrinsic interest, characterization of charge transport in such dielectrics is essential for understanding electrical [4] and laser-induced [5] breakdown phenomena. While optical pump-probe techniques have been applied to study photo-induced carriers in sapphire [6], measurements of the THz response are particularly valuable because of their direct correlation with the transport properties of the mobile charge carriers.

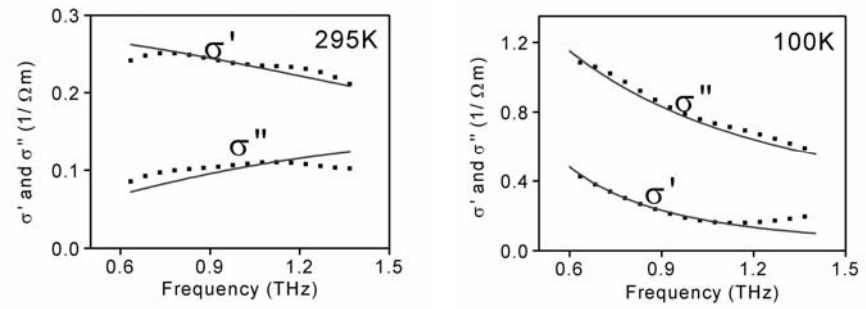
2. Experimental Methods

In these measurements, the laser source was an amplified, mode-locked Ti:sapphire system, which provided pulses of approximately 100-fs duration at a wavelength of 810 nm. Charge carriers in sapphire (band gap of 8.7 eV) were created through a two-photon absorption process by femtosecond ultraviolet pulses at the third harmonic frequency. The pump-induced changes of the material were probed by THz electromagnetic pulses introduced at a defined delay time. The THz pulses were generated and detected, respectively, by optical rectification and electro-optic sampling of the femtosecond laser pulses in ZnTe crystals.

At each sample temperature, the electric-field waveform of THz pulses transmitted through the unexcited sapphire crystal and the pump-induced change in the THz waveform were recorded. The real and imaginary parts of the transient conductivity were then extracted [2].

3. Results and Discussion

Results at two representative temperatures, 295 K and 100 K, are illustrated in Figs. 1 and 2 (dots). We found that the photo-induced conductivity could be described approximately by a Drude model. From such a fit (lines), we obtained the plasma frequency and carrier scattering rate. At 295 K and 100 K, we found widely differing scattering rate of ~ 13.8 and 1.6 THz, respectively, corresponding to scattering times of 70 and 640 fs. The present approach allows us to separate the behavior of the scattering rate from other temperature-dependent effects (such as carrier capture in shallow traps) that complicate the interpretation of conductivity data [7]. The strong increase of the carrier scattering rate with temperature could be described roughly (above the lowest temperatures) by an activation energy of ~ 0.1 eV. This behavior suggests that LO phonons play a major role in the scattering process, as has been predicted theoretically [8].



Figs. 1, 2. Frequency dependence of the change in real ($\Delta\sigma'$) and imaginary part ($\Delta\sigma''$) of the conductivity (dots) at 295 and 100 K. The UV pump fluence was 0.3 mJ/cm^2 at a pump-probe delay of 5 ps. The solid lines are from a fit to the Drude model.

By varying the delay time between the UV pump pulse and the THz probe pulse, we recorded the temporal evolution of the density of the photo-generated charge carriers (Fig. 3). We observed a decay time in the range of 10's of picosecond, which varied appreciably from sample to sample. The measured lifetimes showed no significant dependence on the pump fluence and relatively weak dependence on temperature.

The time scale for the decay of the induced THz response in our studies was found to be similar to that of an earlier measurement of optically induced THz response in sapphire reported by Haran *et al.* [9]. In this study somewhat different excitation conditions were used and the decay was attributed to phonon relaxation, rather than to the dynamics of charge carriers. To confirm our interpretation of the electronic nature of the response for our excitation conditions, we performed a

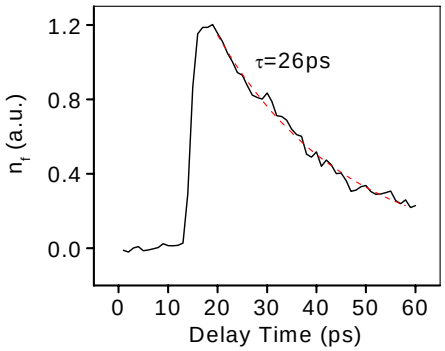


Fig. 3. Time-resolved measurements of the density of mobile carriers in sapphire for photo-excitation at a fluence of 25 J/m^2 by THz pump-probe spectroscopy. The fit is an exponential decay with a time constant of 26 ps.

complementary time-resolved measurements with near-infrared probe pulses. A similar decay was observed. This situation is expected for an electronic effect, but is difficult to explain as the consequence of an induced response of phonons. With respect to the measured decay rate of THz conductivity, the time scale of 10's of picosecond is too short to be attributed to the loss of charge carriers by radiative electron-hole recombination. Further, as noted above, the decay rate was found to vary from sample to sample. This behavior suggests the decrease in the density of mobile carriers results from localization at trap sites, such as impurities and defects.

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