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Abstract—We perform THz spectroscopic investigations of the dielectric function of disordered materials in the THz region. Specifically, we consider amorphous materials and perform ultrabroadband THz spectroscopy of chalcogenide glasses where we observe the transition from universal scaling of the absorption at low frequencies to medium-range order and local order at higher frequencies.

I. Introduction

Disordered condensed matter materials are a class of materials which encompass liquids and amorphous solids. While long-range order is absent in this class of materials, a certain amount of local order still characterizes the materials. For instance, liquid water consists of identical water molecules with an average ordering of the local environment in a dynamic and random global environment. This mixture of order and disorder is reflected in the broadband absorption spectrum of liquid water which is dominated by global relaxational dynamics at low frequencies (up to a few THz), vibrational modes of the intermolecular structures at intermediate frequencies (5-15 THz), and molecular vibrations at still higher frequencies (up to approximately 100 THz) [1]. A similar example is amorphous molecular glasses, where the individual molecules (for instance As_2S_3, in the chalcogenide soft glass arsenic trisulfide) are arranged with a certain amount of local and medium-range order, while the global network of the molecules is arranged in a disordered and random fashion. In contrast to liquids, the dielectric properties in amorphous glasses is dominated by a continuous distribution of vibrational modes with universal properties in the low THz range [2] while relaxation dynamics is expected at much lower frequencies due to the rigid network.
Here we discuss the limits of the validity of the universal absorption features in disordered materials [2] by the example of ultrabroadband characterization of the full dielectric function of the soft glass As_2S_3.

II. Experimental technique

Most terahertz time-domain spectroscopy (THz-TDS) studies to date have been focused on the region below 3 THz due to instrumentation limitations (electro-optic generation and detection in ZnTe crystals or photoconductive switches). This limitation of the bandwidth of the spectroscopic system naturally enforces certain limitations to the interpretation of measured spectra, in particular in the case of broad, rather featureless absorption profiles. Larger bandwidth measurements have proven their value in several investigations, as exemplified by references [3-5].
Here we demonstrate a THz-TDS system which employs air plasma generation of ultrashort THz transients [6-7] in combination with air biased coherent detection (ABCD) of the THz transients [8].
The generation and detection processes are both based on four-wave mixing in air. As in any other frequency conversion process, the bandwidth of the process is always limited by the bandwidth of the laser source and the bandwidth of the phase matching. With air as the nonlinear medium the phase matching conditions are perfect over an extremely broad bandwidth range, and therefore the bandwidth of the generation and detection processes is in practice limited only by the laser bandwidth.
We use a transform-limited 35-fs laser pulse from a standard regenerative amplifier system (SpectraPhysics Spitfire), resulting in THz transients with a spectral coverage from 1 to 40 THz and a THz pulse duration of less than 50 fs.

III. Ultrabroadband permittivity of As_2S_3

The absorption coefficient and index of refraction of the soft glass arsenic trisulfide (As_2S_3) is shown in Fig. 1.

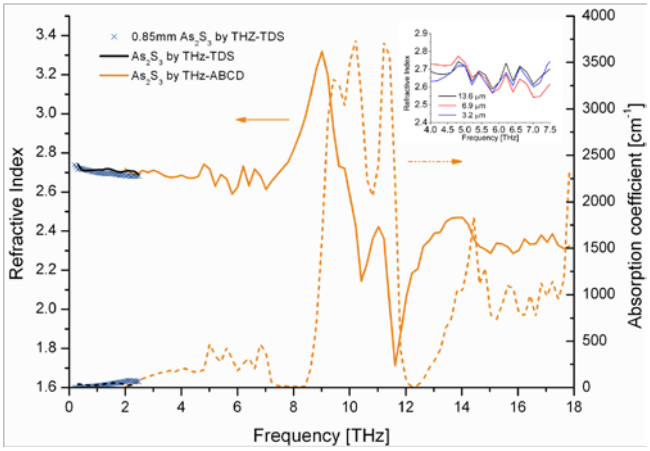

Figure 1: Absorption coefficient (dashed line) and index of refraction (solid line) of As_2S_3 at room temperature. Data represented by black curves and blue symbols are recorded with a low-bandwidth THz-TDS system, data represented by orange curves are recorded with a high-bandwidth THz-TDS system.

The data in Fig. 1 is recorded on three different samples, with two different THz-TDS spectroscopy setups. The low-frequency (0.1-2.5 THz) data are recorded in a conventional THz-TDS system based on photoconductive switches [9]. The high-frequency data are recorded in a THz-TDS system based on air plasma generation of ultrabroadband THz transients [6-7] and air biased coherent detection (ABCD) [8].

At low frequencies we observe the expected broad, monotonously increasing absorption which is a general signature of disordered materials [2]. In the frequency range 4-7 THz we observe highly reproducible (se inset of Fig. 1) absorption features originating from medium-range order in the glass, followed by three strong absorption lines in the 10-12 THz range, originating from intramolecular vibrations in the As_2S_3 molecular unit.

As_2S_3 has very high third-order nonlinearity in the mid-infrared region, and there is a large interest in extending nonlinear optics based on soft glasses into the far infrared. With this future development in mind we find it very interesting that there are two transparency regions in the frequency range that we have investigated, near 8 THz and 12.3 THz.

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