THz induced nonlinear absorption in ZnTe

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***Abstract*—Absorption spectra of ZnTe during strong-field THz interaction are investigated. Bleaching of the difference phonon modes below the fundamental TO mode is observed when field strengths higher than 4 MV/cm are applied.**

I. INTRODUCTION

TERAHERTZ (THz) systems are widely used for studies of lowest vibrational modes in crystalline materials. At the same time, the possibility of generating THz field strengths on the order of MV/cm with table-top THz systems [1,2] has enabled nonlinear studies of intermolecular vibrational modes [3] as well as carrier dynamics [4]. ZnTe is an example of a semiconductor, which is often used both for coherent generation and detection of THz pulses due to its high second-order nonlinearities. ZnTe has a number of low-frequency phonon modes below the fundamental TO phonon mode at 5.4 THz, particularly the two bands around 1.6 and 3.7 THz originating from difference frequency modes from longitudinal and transverse optical and acoustic phonons (LO, TO, LA and TA) [5]. Here, the absorption spectrum of these phonon modes is investigated when the incident field strength is increased to several MV/cm using a DSTMS THz source and air-biased coherent detection in order to cover the desired bandwidth from 1 to 4 THz.

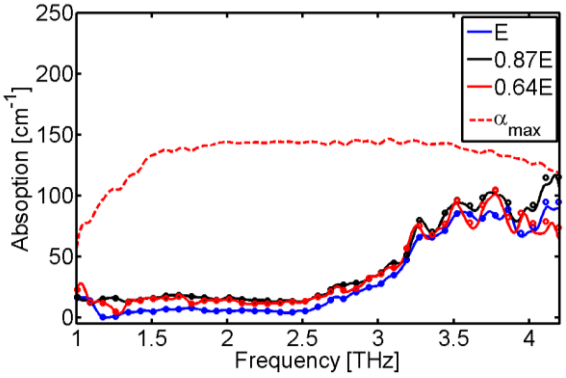

Fig. 1. Absorption spectra measured at three field strengths.

II. RESULTS

THz-TDS spectroscopy is performed on a 0.5 mm thick ZnTe using a DSTMS THz source pumped with 1 mJ pulses centered at 1300 nm and with a pulse duration of 70 fs. This results in field strengths of the focused beam at the sample position of up to 4.9 MV/cm. The field strength incident on the sample is controlled with a pair of wiregrid polarizers.

Figure 1 shows the measured absorption coefficient at three different field strengths together with the maximum absorption, which can be detected due to the dynamic range of the system, α_{max}, here covering the range 1-4.2 THz. Considering the absorption features around 3.7 THz, it is seen that three distinct absorption peaks are observed for the lowest field strength (red curve). When the field strength is increased the peaks are first broadened (black curve), and for the highest field strengths the absorption decreases (blue curve). This suppression of absorption peaks is interpreted as phonon bleaching, which is similar to what was observed in [3] when the population in an anharmonic vibrational potential is redistributed by ladder climbing. The same behavior is seen at lower frequencies, which is expected to be due to a similar mechanism for the broad absorption feature at 1.6 THz.

The average transmission and the integrated absorption as function of field strength are shown in Fig. 2. The averaged transmission (red curve) is calculated from 1.5 to 2.5 THz to represent the lowest difference phonon mode in the optimum α_{max} region, while absorption is calculated between 1.5 and 4 THz (black curve). It is seen that absorption increases slightly after until the field is increased to approximately 4.2 MV/cm after which it decreases abruptly. In agreement, the inverse trend is observed for the transmission. Similar studies have been performed for other samples of lactose and sucrose, from which the nonlinear absorption features are compared.

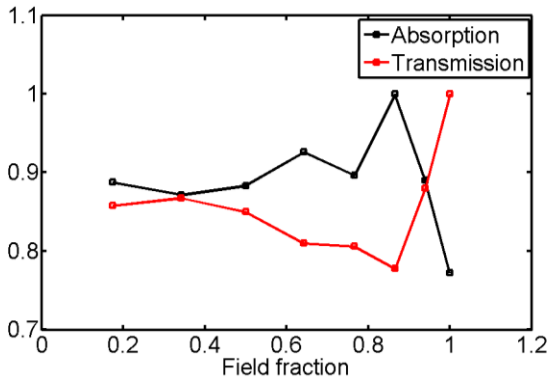

Fig. 2. Field-dependent transmission (red) and absorption (black) in ZnTe.

III. SUMMARY

In summary, nonlinear absorption has been observed for phonon modes in ZnTe. This is expected to be caused by phonon bleaching due to ladder climbing in an anharmonic vibrational potential when high fields are applied.

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