Analele Universităţii de Vest din Timişoara
Vol. L, 2007
Seria Fizică

ULTRA SHORT LASER PULSE INDUCED ELECTRON-PHONON DYNAMICS IN CRYSTALS

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Abstract
The subpicosecond time resolved spectroscopy is a very versatile tool for identification of carrier (and phonon) dynamics in subsurface layers of solids. We investigate the reflectivity behaviour of TeO_2 optical crystals due to irradiation by femtosecond IR laser pulses (pump and probe method) with resolution better than 1 ps. From Fourier spectra of reflected signal obtained we conclude that the appearance of most intense signals depends in different time frames on the angle between pumping beam polarisation and the main crystal axis. This might be convicted with appearance of different phonons and quick rearrangement of short electron-phonon coupling due to dynamic shock waves induced by absorbed laser energy.
Keywords: ultrafast process, dynamic reflectivity, tellurium-dioxide

1. Introduction

The advents of femtosecond lasers have spawn a whole new field of scientific investigations. These lasers have a vide range of applications weather its high intensity or its short pulses. A short pulse can be used to measure ultrafast processes. Phonon vibration, electron-phonon scattering and chemical bond changing can all be directly time resolved by femtosecond pulses. Paratellurite (TeO_2) is commonly used in acousto-optical devices due to their high acousto-optic figure of merit. As will be shown in this paper tellurium dioxide exhibits ultrafast dynamics. The typical experimental realization of such a time resolved measurement is based on a pump and probe system, where the pump pulse is used to modify the sample, and the probe is used to monitor this modification [1-2]. The excited states created by the pump pulse can be directly inspected, e.g., via a spectral analysis. In our measurements the probe pulse is reflected from the excited sample after a controllable time delay. In addition, by varying the delay time of the probe pulse, a time-resolved spectrum of the excited sample is obtained, which in turn tracks the ultrafast process that characterizes the relaxation of the system. Changes in the optical properties of a material provide a window on the electron and lattice dynamics because optical properties depend on the structure (e.g.

crystalline or amorphous) and electronic configuration (e.g. the number of valence electrons per ion and the number of excited electrons)[3-5]. Structure determines the allowed electron energy states, and the electronic configuration describes which levels are occupied. Thus we can extract information about the vibrational dynamics of the sample and the changes in the electron configuration can be concluded. The dynamic reflectivity yields not only the vibrational frequencies, but also characterizes how the amplitudes of the vibrations evoluting with time. We report on dynamic reflectivity measurements of tellurium dioxide crystals.

2. Measurement system

We used a pump and probe system for time resolved measurements *fig.1*. A self-mode-locked titanium-sapphire laser (KMlabs model TS) pumped with a solid-state green laser (Coherent 5W Verdi) generated the femtosecond pulses. The centre wavelength of the pulses was at 810 nm with 40 nm bandwidth (FWHM). A pair of prisms was used to pre-compensate the group velocity dispersion caused by the air and the larger number of optical elements used in the path of the pump pulse. The durations of the pump and probe pulses at the sample location were measured to be 25 fs by frequency-resolved-optical-gating (FROG) technique [6]. The noise reduction was a crucial problem of the measurements, because the system had to be able to detect changes in the reflectivity at the level of about one part in a million. To reduce noise, an acousto-optic modulator (AOM) was used in the pump beam path to act as a high-speed optical chopper. The chopping frequency of the AOM was set to 25 kHz.

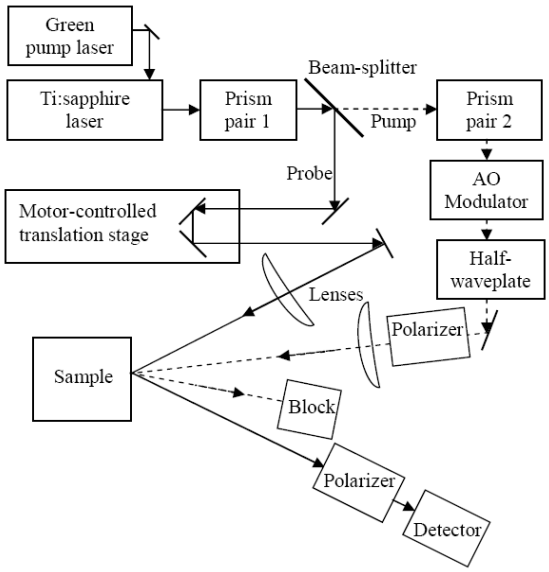

Fig. 1. Schematic diagram of dynamic reflectivity measuring system

A lock-in amplifier was used to detect only the reflected probe light oscillating at the frequency of the chopped pump beam. The polarization of the probe beam was set to be perpendicular to the polarization of the pump beam by using a half-wave plate.

3. Results

The dynamic reflectivity of TeO_2 is shown on *fig.2*. The main change in the reflectivity is induced directly by the arriving pump pulse and is attributed to excitations of the electrons in the sample. The quick rise and fall — *which is about* 50 *fs (FWHM) for* TeO_2 — of the initial peak is followed by smaller amplitude reflectivity oscillations persisting for approximately 10 to 50 ps depending on the sample orientation. The orientation angle of the sample is defined between the polarization angle of the pump pulses and the *c* crystal axis. We have measured the transmission spectrum of the tellurium dioxide TeO_2, and found that the transmission is depending weakly on the polarization angle.

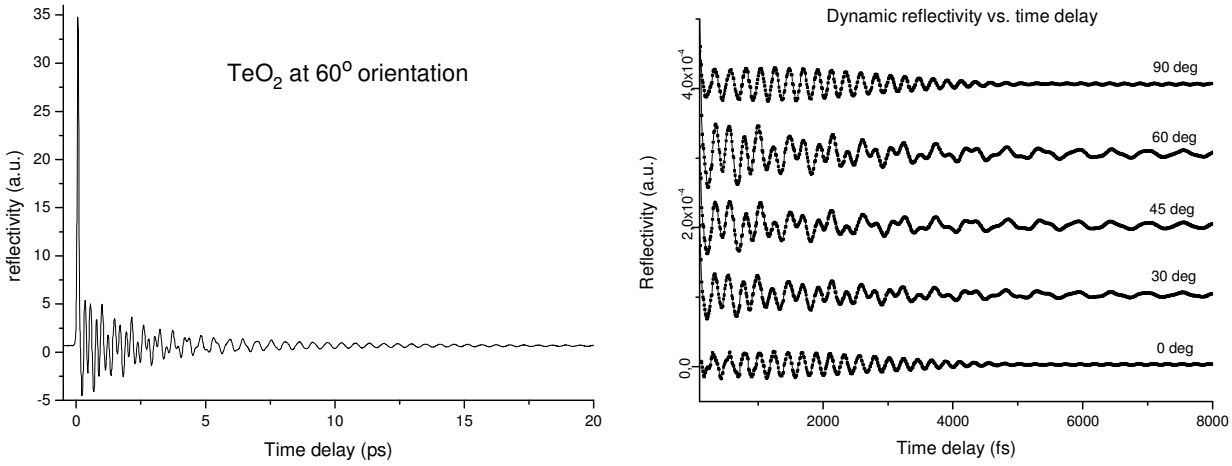

Fig. 2. Dynamic reflectivity of TeO_2 sample.

The initial peak of reflectivity curve increases with the orientation angle, corresponding to the polarization-dependent absorption of the TeO_2. The rapid rise and fall of the main change in the reflectivity is so that it is intrinsically limited by the duration of the laser pulses. The following oscillations of the reflectivity have different relaxation times depending on the sample orientation. At the orientation of 0° and 90° the relaxation times are 8 ps, at 30° and 60° orientation the oscillations persist no longer than 28 ps. The reflectivity change has the longest relaxation time (~50 ps) at 45° orientation.

Fig.3a shows the Fourier transform of the dynamic reflectivity of TeO_2 at 60° orientation. Three different time windows are defined to study the temporal evolution of the oscillations.

The first time window runs from 500 fs and ends at 4 ps. Three different oscillations is induced directly by the laser pulses with a wavenumber of 150 ± 3 cm^{-1}, 133 ± 3 cm^{-1}, 62 ± 2 cm^{-1}. Immediately after the fall of the initial peak the largest wavenumber oscillation dominates, and the oscillation corresponds to 133 cm^{-1} wavenumber has the smallest amplitude. The amplitude of the 149 cm^{-1} wavenumber oscillation decreases, while the relative amplitude of the 62 cm^{-1} wavenumber oscillation increases with time. As *fig.3a* shows, the energy flows from the larger wavenumber oscillations to the smaller wavenumber ones with time. The structures of the oscillations at different sample orientation are very similar to each other. All of the periodic reflectivity change consists of the same oscillations; the only difference is the amplitude of each oscillation. These oscillations also behaving the same way at different sample orientation, the amplitude ratio of the smaller and larger wavenumber oscillations initially shows the dominance of the large wavenumber ones, but the dominance decreases with time beside the relative increase of the amplitude of the small wavenumber oscillation, which vanishes the larger wavenumber oscillation.

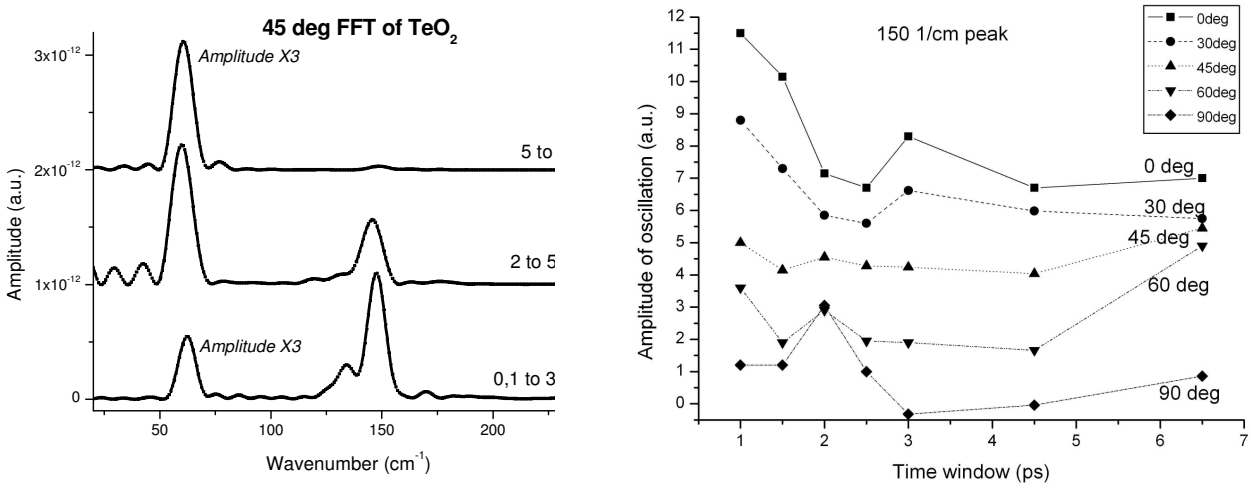

Fig 3. a. Fourier spectra of oscillations at 45-degree; b. Amplitude variation of the 150 1/cm peak.

Fig.3b shows how the amplitude of the 150 cm^{-1} peak changes in different time windows at different orientations. On the horizontal-axis each dot is corresponds with the middle of the time window where the Fourier transform had been taken. On the vertical-axis the amplitudes of the oscillations are noted. We can see in fig.3b, that the behaving of this oscillation similar at 45°, 60°, 90° orientation. Both starts with a slight decrease, which turns into increase at 1.5 ps, reaches the local maximum at 2 ps, than decrease again until 3 ps and in the latest

regime a small increase can be noted. At 0°, and 30° a continuous decreasing is observed except the interval between 2.5 ps to 3 ps where a small increase can bee seen.

These behaves cannot be explained by the classical picture of the structure of TeO_2. This phenomenon more likely pointed out that the α-TeO_2 cluster structure with its relative oscillator strength responsible for these changes.

4. Conclusions

Femtosecond laser pulses can induce ultrafast changes in optical properties of TeO2. The laser pulses excite acoustic vibrations at different wavenumbers. The amplitudes and the relaxation time of the oscillations strongly depend on the angle between the crystal axes and the polarization of the pulses. Oscillations with different wavenumbers dominate in different time scales after. The special behave of the oscillations corresponds to the structure of TeO_2.

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