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# Femtosecond optical Kerr effect study of amorphous chalcogenide films

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**Abstract**

The non-resonant third-order non-linear optical properties of some amorphous chalcogenide films were studied experimentally by the method of the femtosecond optical heterodyne detection of the optical Kerr effect. The real and imaginary parts of the complex third-order optical non-linearity could be effectively separated and their values and signs could be also determined. Amorphous chalcogenide films showed a very fast response in the range of 200 fs under ultrafast excitation. The ultrafast response and large third-order non-linearity are attributed to the ultrafast distortion of the electron orbitals surrounding the average positions of the nucleus of chalcogen atoms. The high third-order susceptibility and a fast response time of amorphous chalcogenide films make them promising materials for application in advanced techniques especially in optical switching.


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*Keywords:* X-ray diffraction; Chalcogenides; Non-linear optics; Ultrafast processes and measurements

## 1. Introduction

Many advanced techniques, including optical switching, modulation, optical information processing and ultrafast optical communications, need the devices made from materials with fast optical non-linear responses. Especially, the rapid development of optical communications requires novel materials with large and ultrafast non-linear optical responses in the femtosecond domain for fabricating ultrafast optical switches and processing devices [1,2]. A number of approaches for measuring the magnitude and dynamics of third-order optical non-linearities have been developed in the past years [3–5]. One of the essential problems is that the third-order susceptibility $\chi^{(3)}$ must be defined as a complex quantity. Optical Kerr effect (OKE) is one powerful technique to observe the third-order non-linearities and relaxation phenomena, but there exists the same problem. On the basis of OKE, a new method called femtosecond optical heterodyned detection of optical Kerr effect (OHD-OKE) was developed. The real and imaginary parts of complex third-order optical non-linearity can be effectively separated and their values and signs can be determined [6,4,7]. Because of high refractivity and structural flexibility, chalcogenide semiconductors are susceptible to photoinduced changes, and their non-linearities should be large. In the present paper, some amorphous chalcogenide films were prepared and the optical non-linear property was studied by the femtosecond optical heterodyned detection of optical Kerr effect (OHD-OKE) [6,4,7] method. Some new results on non-linear optical materials for optical switch are obtained.

## 2. Experimental

Bulk $GeSe_2$, $Ge_{20}As_{25}Se_{55}$ and $Ge_{10}As_{40}S_{30}Se_{20}$ chalcogenide glasses were firstly prepared from 5N purity of Ge, As, S and Se elements (99.999%) in an evacuated sil-

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ica ampoules ($10^{-3}$ Pa) at temperature of ~1000 °C for about 72 h. The films of ~300 nm on $K_9$ substrates were prepared from bulk chalcogenide glasses at a rate of 15 Å/s in a $1 \times 10^{-3}$ Pa vacuum. The thickness and refractive index were measured by NKD-7000w System Spectrophotometer (AQUILA Company, UK) and the refractive index of the film at the 805 nm wavelength is ~2.5.

The third-order susceptibility and the ultrafast response of amorphous chalcogenide films were measured by the method of OHD-OKE. Fig. 1 shows the OHD-OKE experimental set-up. The mode-lock Ti:Sapphire laser pumped by the Millennia Vs laser which provides 4.1 W of green 532 nm, is operated at 82 MHz and has an average output power of 680 mW, a wavelength of 805 nm and a pulse duration of 80 fs. The output beam is divided into a pump and a probe beam ($I_{probe}$:$I_{pump}$ = 1:10) by a beam-splitter. The polarizer P1 is directed at 45° with respect to that of the pump beam. The pump and probe beams, with the average power 20 mW and 2 mW respectively, are focused into the sample, and their confocus length is about 200 μm. A polarization analyzer is placed behind the sample at the crossed polarization direction to the input polarizer. A quarter-wave plate is inserted between the input polarizer and the focusing lens to measure the real part of $\chi^{(3)}$ in the OHD-OKE experiments. The optical axis of the quarter-wave plate is also directed at 45° to the polarization direction of the pump beam. During the process of the OHD-OKE experiments, we can detect the real signal with the quarter-wave plat and the imaginary signal without it. Furthermore, the OHD-OKE effect will be observed only when the angle (heterodyne angle) between the optical axis of the quarter-wave plate and polarization direction of the pump beam is very small (<5°). So we selected 2° heterodyne angle in our experiments of OHD-OKE [6,4,7].

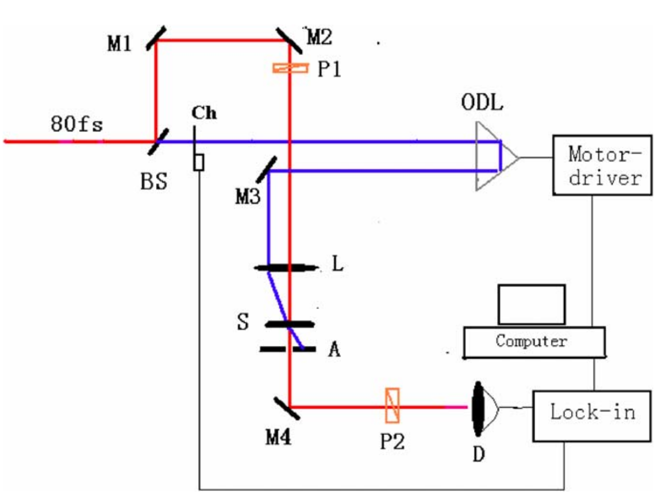


Fig. 1. The OHD-OKE experimental set-up.

## 3. Results and discussion

It is shown from Fig. 2 that $Ge_{10}As_{40}S_{30}Se_{20}$ bulk obtained by water-quenched method and film prepared from bulk $Ge_{10}As_{40}S_{30}Se_{20}$ glass powder are both amorphous. No diffraction peaks occurs except two wide diffraction peaks occur in the X-ray diffraction spectrum of $GeSe_2$ film due to the effect of $K_9$ glass substrate. The same results were obtained in $GeSe_2$ and $Ge_{20}As_{25}Se_{55}$ systems.

For measuring the third-order non-linear susceptibility of amorphous chalcogenide films, $CS_2$ was used as the reference sample and was firstly measured to examine the reliability of our OHD-OKE set-up [6]. The OHD-OKE signal of $CS_2$ at two different heterodyning angles: +2° and −2° is shown in Fig. 3. The value of $(\chi^{(3)})_R$ (real part of $\chi^{(3)}$) for $CS_2$ is $0.67 \times 10^{-13}$ esu in femtosecond time scale (whose $\chi^{(3)}$ is positive) [7,9]. The OHD-OKE signal (real and imaginary parts) of amorphous $Ge_{10}As_{40}S_{30}Se_{20}$ film is shown in Fig. 4, respectively. It is indicated from the symmetry of the signal that amorphous $Ge_{10}As_{40}S_{30}Se_{20}$ film shows a very

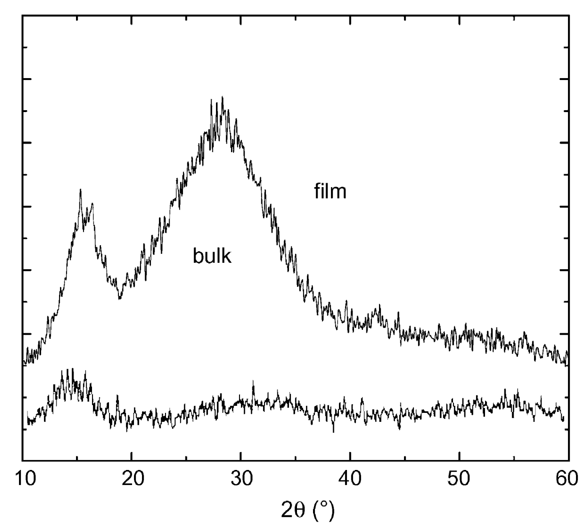


Fig. 2. The XRD spectrum of $Ge_{10}As_{40}S_{30}Se_{20}$ bulk and film.

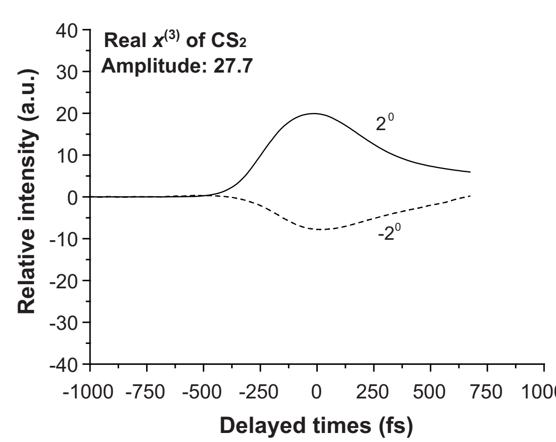


Fig. 3. OHD-OKE signals of $CS_2$ at two different heterodyne angles: −2° and +2°.