Probing the Second-Harmonic in ZnCdSe Thin Films

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Abstract

The second-harmonic behavior of thin-film materials is not well understood. These materials have promise to aid in both laboratory and industrial applications, such as optical computing. One of the keys to understanding these effects is to characterize family of similar semiconductors rather than a single binary compound. By systematically exploring a library of materials we will eventually be able to tune for their desired properties depending on the application. We are exploring the $Zn_{1-x}Cd_xSe$ family of ternary thin film alloys for their second-harmonic response. The responses should lie between that of ZnSe and CdSe. In addition, we have access to the $Zn_{1-x}Mg_xSe$ family.

One of the difficulties in examining non-linear responses of a material is differentiating those responses from the linear effects. The linear reflectivity of thin films can be characterized using a waveguide prism coupling technique (PCT). We have built a unique prism coupler technique (PCT) set-up with rotating stages to examine the changing incidence, and with a waveguide mode to accurately measure dispersion curves (index of refraction (n) versus wavelength) in both vertical and horizontal polarizations. In addition, we can accurately determine the amount of light entering the film and can begin to tease apart the linear and non-linear response using the waveguide modes. The second harmonic will be probed with a high-power pulsed 1064 nm laser. While we have not begun the PCT in the second harmonic, we have preliminary results and are hopeful for a positive result.
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Introduction

The nonlinear versus the linear response of materials is analogous to more everyday effects. For example, adjusting the volume normally affects the sound frequencies of speakers in a linear fashion. However, when the volume is turned up to high, the speakers follow a non-linear response to the voltage it is feed. We hear distortions of the original sound including changing amplitudes of frequencies and entirely new frequencies formed from the sum and differences of existing frequencies. This non-linear response can be seen in the spectroscopy of organic and inorganic crystals. For example, using readily available lasers in the visible spectra, crystals can be probed in the UV or IR spectra using sum and differences frequencies. Using nonlinear techniques in the investigation of these materials can give more true interaction with the bulk material.[1] The Maker fringe technique is a proven way to characterize non-linear effects.

Maker fringes

When a laser beam passed through a thin film, the free and bound second harmonic waves can be observed moving in and out of constructive and destructive interference. The bound waves occur as a result of the induced electric field inside the crystal, whereas the free waves are created at the surface of the crystal. Changing the angle of incidence changes the path length of the light within the crystal.[2]

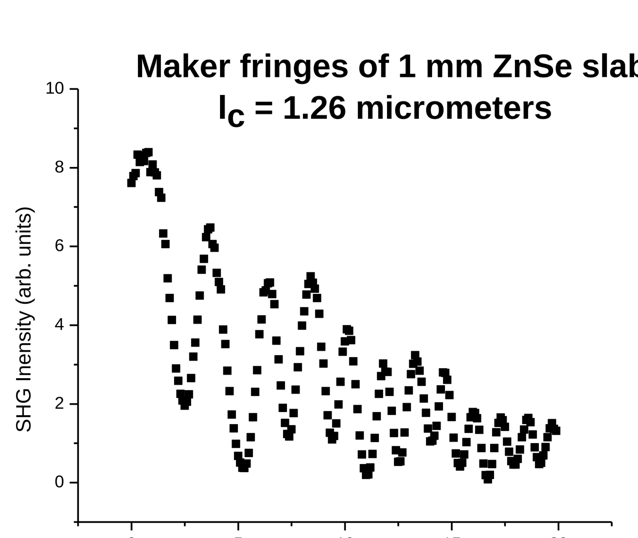


Fig. 1 The interference patterns from the Maker fringes can easily be seen with a bulk slab

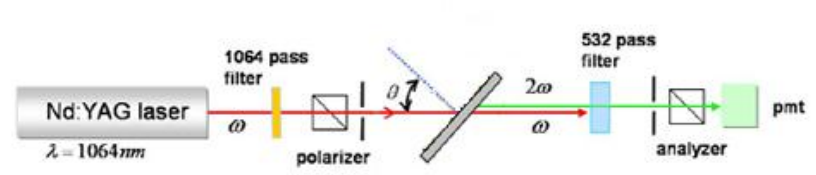


Fig. 2 The general scheme for Maker fringes

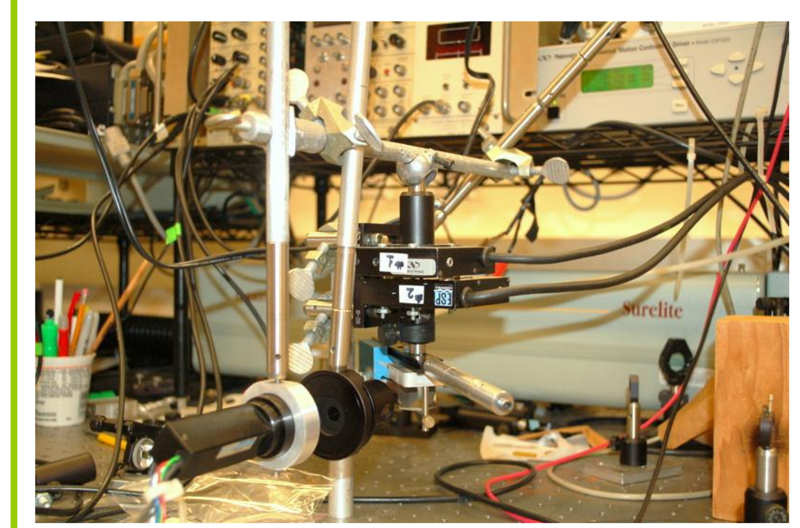
$$l_c = \frac{2\pi}{\Delta k}$$

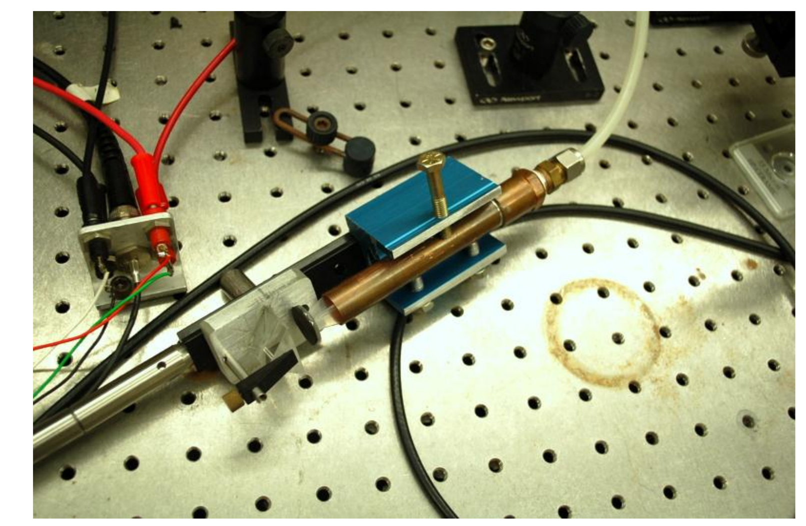
Eq. 1 The change in pathlength must equal an integer multiple of the coherence length, l_c

Experiment

Because the change in pathlength is so small in thin films, Maker fringes are not a feasible method to characterize their second harmonic response. We looked to a waveguide prism coupler technique (PCT) as an alternative.

Prism Coupler Technique

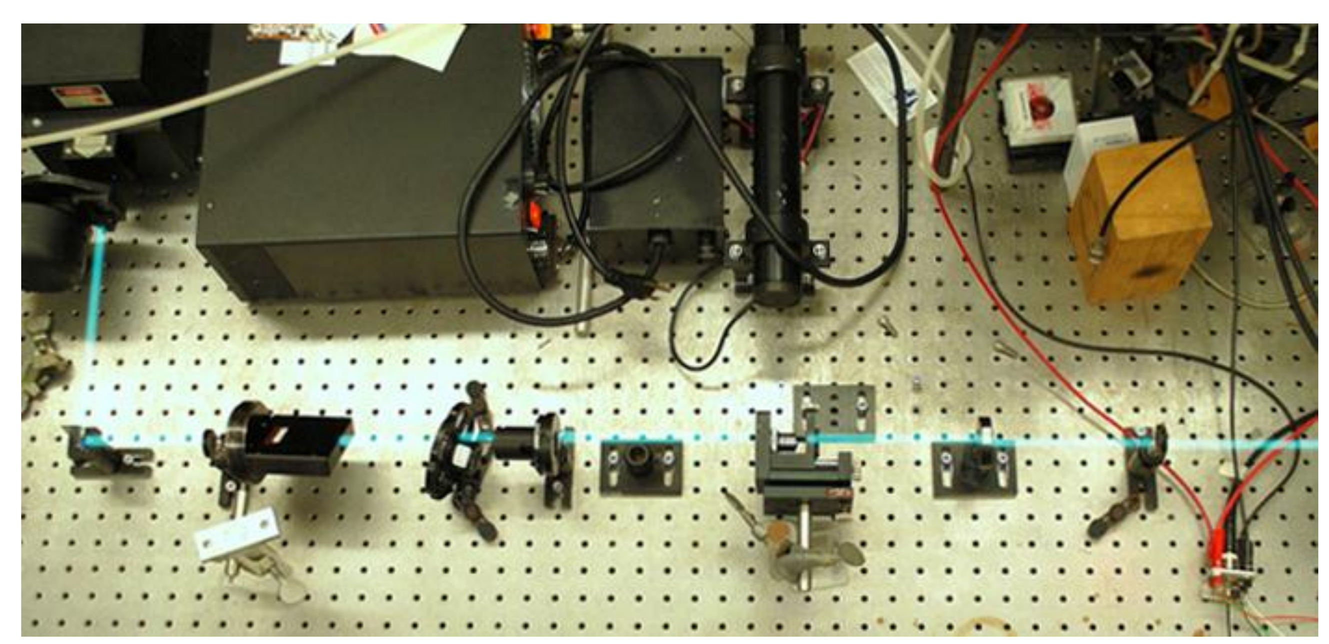




We built the prism coupler system shown at the left using two supsended Newport rotating stages and photomultiplier tube (PMT) operated by LabView. The image at the right is the prism with the pneumatically operated coupling plunger which applies 30-40 psi to the back of the sample onto the back of the prism

Waveguide Modes

Manipulation of Source Beam



488 nm light passing through an iris, Fresnel rhomb, optical density filter, polarizer, spatial filter, + 6 inch lens, and an iris into the PCT system.

By changing the incoming angle α, the angle at the prism base θ changes too. At certain angles θ the wave vector parallel to the interface matches (x-direction) the supported waveguide modes (Fig. 5). The evanescent wave “tunnels” through the air barrier.

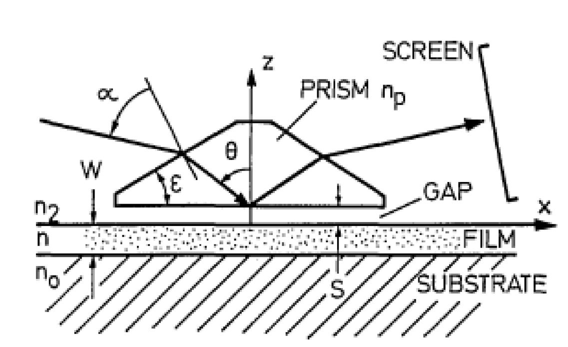


Fig. 3 Waveguide PCT[3]

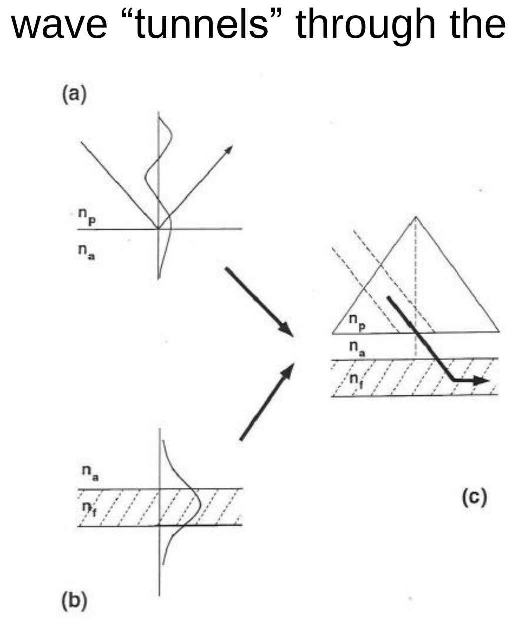


Fig. 4 Optical tunneling in a waveguide PCT[4]

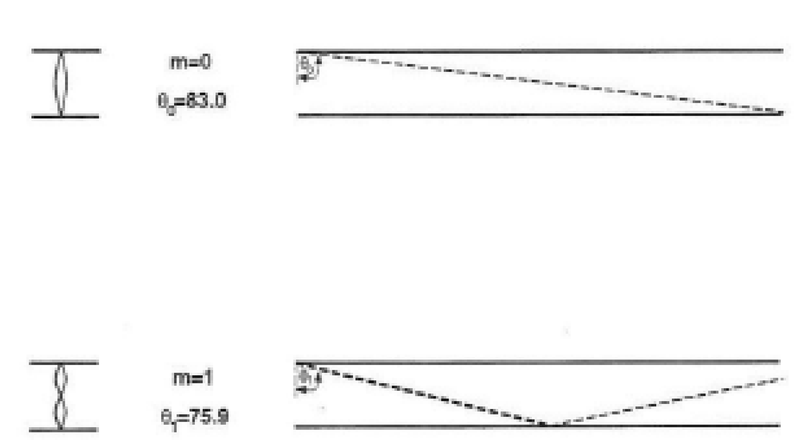


Fig. 5 The supported modes in a thin film[4]

$$\vec{k}_p sin(\theta) = \vec{k}_f sin(\theta_m)$$

Eq. 2 The conditions for matching wavevectors in the x-direction.[4]

Results

When the source beam is coupled into the waveguide modes of a thin film, the intensity of the output beam is reduced and we observe dips.

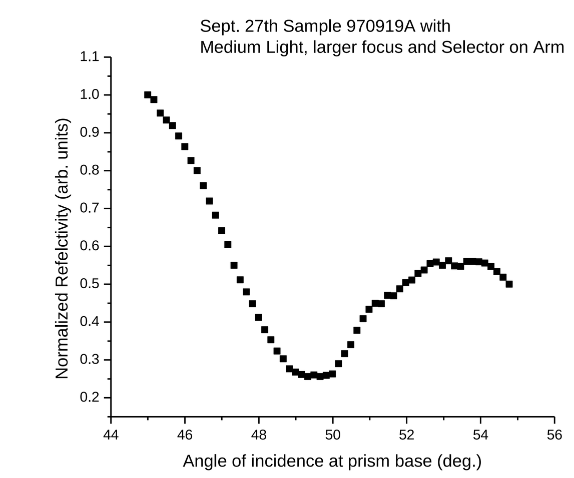


Fig. 6 The first observed dip in a 2.300 μm $Zn_{.91}Cd_{.09}Se$ thin film with 488 nm HeNe laser. Subsequent dips were difficult to observe.

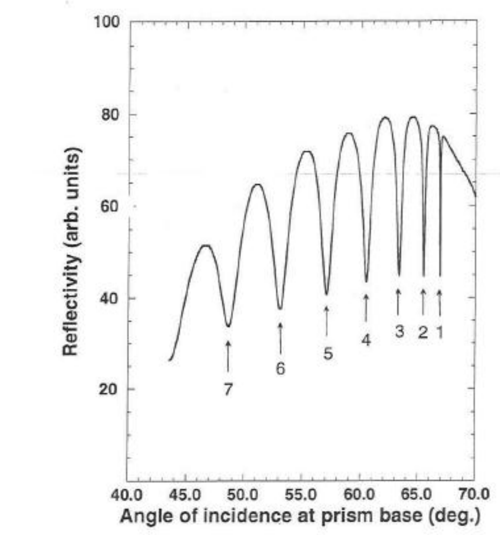


Fig. 7 The spectrum of a 1.400 μm $Zn_{.725}Cd_{.275}Se$ thin film with a 632.8 nm HeNe laser from Peiris’s work[4].

Discussion and Future Work

$$n_{mode} = n_p sin\theta$$

Eq. 3 The effective index of refraction of each waveguide mode is given by n_{mode}.[5]

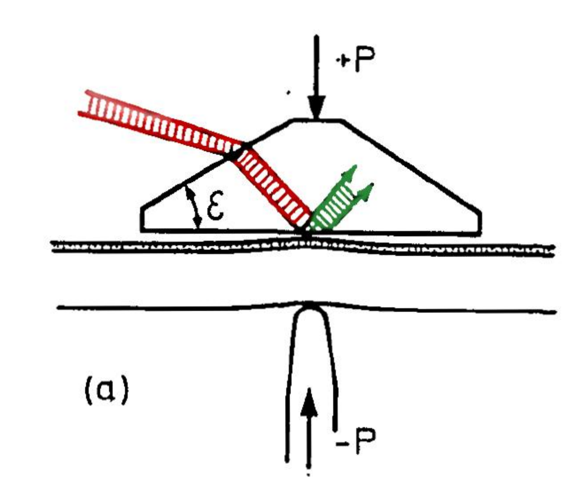


Fig. 8 Localized pressure causing deformation of the film[3].

The number of modes that can be observed is dependent upon the thickness of the film and the index of refraction of the prism (Eq. 3). Because we used a prism with a small index of refraction (1.5) we could only see one or possibly two modes. In order to observe more modes (we need at least two) we plan to use a rutile (TiO_2, n = 2.86) prism for future experiments. In addition we hope to localize pressure as seen in Fig. 8 as the literature has suggested.

The observed second harmonic will be measured by either observing the doubled light coming back though the prism (Fig. 3), or by detecting in the plane of the thin film. We will use a pulsed 1024 nm laser because the second harmonic requires more power.

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