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Relative SHG measurements of metal thin films: Gold, silver, aluminum, cobalt, chromium, germanium, nickel, antimony, titanium, titanium nitride, tungsten, zinc, silicon and indium tin oxide

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ABSTRACT

We have experimentally measured the surface second-harmonic generation (SHG) of sputtered gold, silver, aluminum, zinc, tungsten, copper, titanium, cobalt, nickel, chromium, germanium, antimony, titanium nitride, silicon and indium tin oxide thin films. The second-harmonic response was measured in reflection using a 150 fs p-polarized laser pulse at 1561 nm. We present a clear comparison of the SHG intensity of these films relative to each other. Our measured relative intensities compare favorably with the relative intensities of metals with published data. We also report for the first time to our knowledge the surface SHG intensity of tungsten and antimony relative to that of well known metallic thin films such as gold and silver.

Introduction

Surface SHG is a very useful technique for generating second order nonlinearities in systems with inversion symmetry [1,2]. From the early experimental observation of surface SHG in metals in the 60's and 70's [3–8] to the present day [9–13], a lot of experimental data has been generated for the surface SHG and susceptibility components of many metals. There is a lot of published experimental data on the surface SHG of metallic films such as Au, Ag and Al [14–16]. However, most of these studies typically cover narrow frequency ranges, illumination angles and film thicknesses. Published experimental data on SHG in other metallic thin films is even more scarce. Unlike the ready availability of experimental data on the linear optical properties of metal thin films [17,18], SHG data for these same materials is hard to come by and therefore very much cherished. Thus any new addition to the experimental data of SHG for different thin films, using different sources and illumination configurations is very useful especially for researchers who need some kind of starting reference for studying a given thin film.

In our study, we set out to investigate the reflected SHG from a variety of metal and semiconductor thin films and to develop a simple yet useful relative calibration system for these films. We examined the SHG intensity of Au, Ag, Al, Co, Cr, Ge, Ni, Sb, Ti, TiN, W, Zn, Si and ITO. For an incidence angle of 23.6°and a wavelength of 1561 nm, we were able to calibrate the surface SHG response of these thin films relative to each other and to well studied films such as Au and Ag.

Experimental setup

We irradiate the thin films used in our study using the configuration shown in Fig. 1, where θ_0 is the incident angle. The conducting films are deposited on a glass substrate. All the thin films were grown through magnetron sputtering, with the thickness of the films monitored during growth. The films were grown to thicknesses of either 50 nm or 100 nm.

We used a linearly polarized fiber laser source, with a central wavelength of 1561 nm, and a pulse width of 150 fs. The average incident power range at the sample plane, after propagating through the system optics was 8–70 mW. The incident laser beam was collimated using a broadband collimator, passed through a linear polarizer and rotated to generate p-polarized light. This p-polarized light was focused on the thin film sample using a 0.4 N.A reflective microscope objective at an angle of incidence of 23.6°. The angle of incidence is not very critical in these studies, since surface plasmon coupling is not involved. The reflected linear and second-harmonic beams were collected and collimated by the same microscope objective. They were separated by a dichroic

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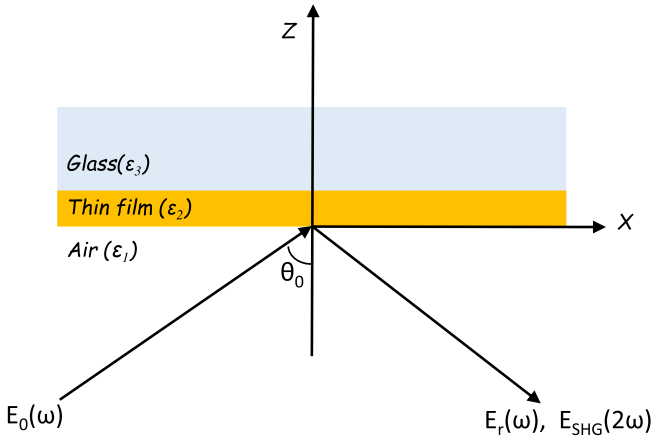

Fig. 1. Irradiation configuration.

mirror, with the second-harmonic wave aligned for p-polarization and detected with a spectrometer. The incident light intensity at the source plane was measured regularly and the variation was less than 4%.

Results

Fig. 2 shows both the incident wave spectrum and the SHG spectrum for a 50 nm thick Au film when irradiated with a 38 mW, 150 fs laser centered at 1561 nm. We use the same source for the rest of our measurements and vary the intensity from 8 mW to 70 mW.

Fig. 3 shows the measured second-harmonic intensity of p-polarized light for 100 nm thick conducting thin films. We observe that the second-harmonic intensity profiles split roughly into four groups. Group A consists of Au, Ag, Al and W and this group has the highest second-harmonic nonlinear response of our system. This group of metals has also being well studied. We can see from [15] that they all have comparable SHG signals for p-polarized light. Group B consists of Cu, Ti, Co and Ni with the signals 30% weaker than those of group A. Group C consists of Cr, Ge, Zn, Sb and TiN, which have 15% weaker SHG intensity compared to group B. Finally, group D films consisting of Si and ITO which are both

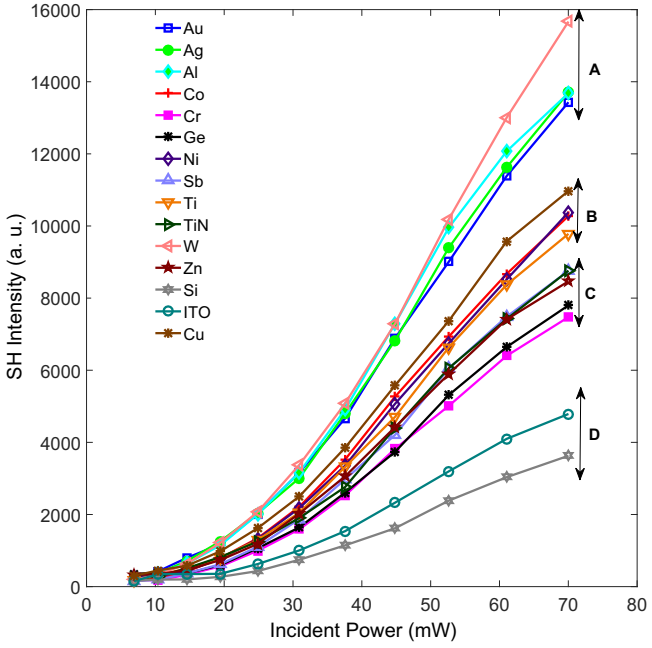

Fig. 3. Measured SHG intensity for 100 nm thick films.

semiconductors have the weakest measured intensity with their SHG intensity being an order of magnitude lower than that of group A.

Some of the materials in Fig. 3 were grown to just 50 nm in order to examine any dependence of thickness on the second-harmonic response of the films. The results are shown in Fig. 4. We do not observe any significant difference between the second order response of the 100 nm films compared to the 50 nm films. This is consistent with the results in [19], where little variation in SHG intensity is observed in films with thickness ranging between 50 nm and 100 nm.

A power dependence test of the reflected second harmonic wave is shown in Fig. 5 for sample 50 nm films from the different groups identified. Linear regression is then applied to the log–log plots revealing a second-order dependence of the SHG intensity to the incident power.

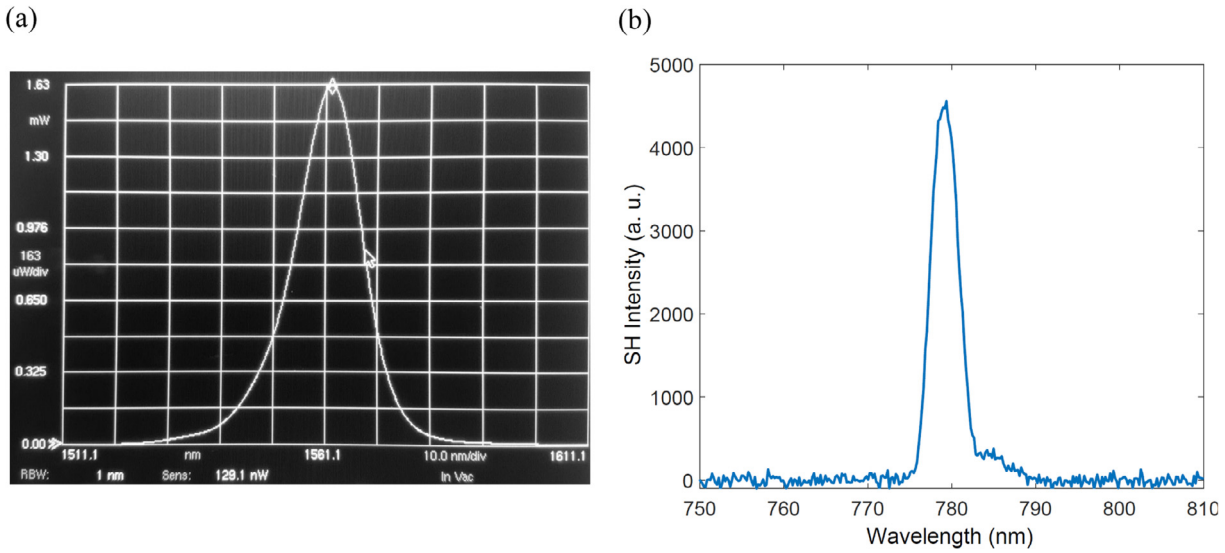

Fig. 2. (a) Incident fundamental spectrum at 1561 nm (b) SHG spectra of a 50 nm thick Au film.

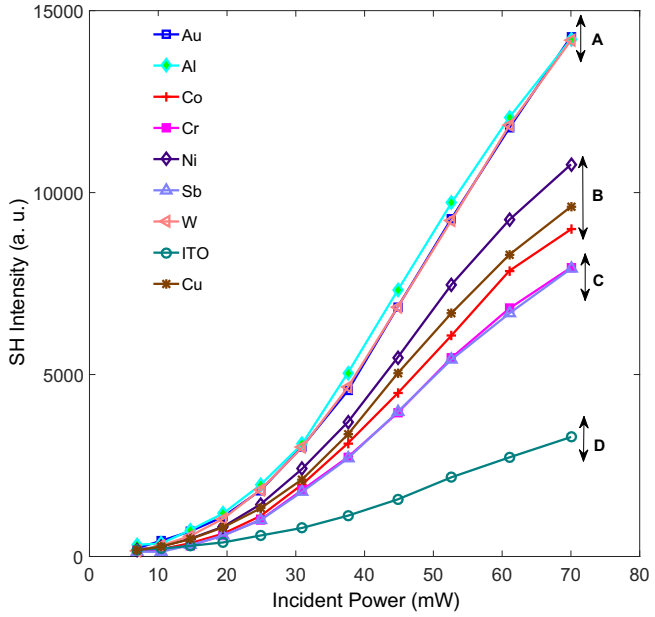

Fig. 4. Measured SHG intensity for 50 nm thick films.

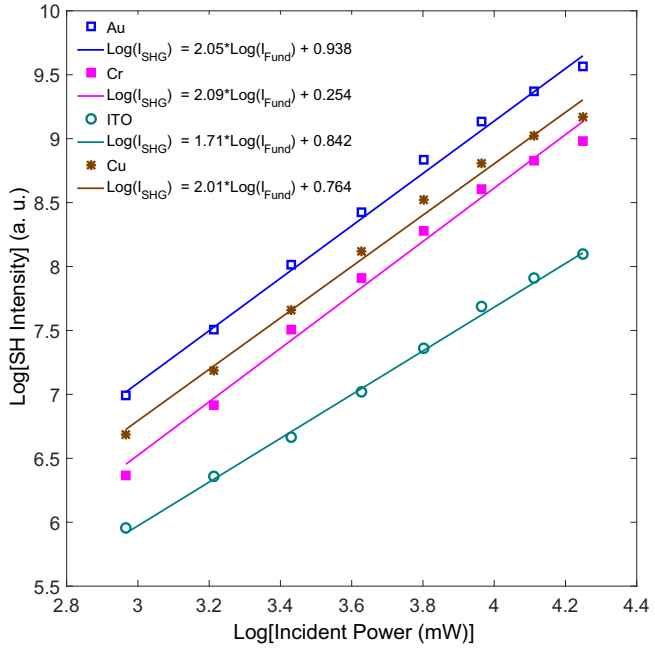

Fig. 5. Log–log plot of the SHG intensity of 50 nm thick Au, Cr, Cu and ITO films. The lines represent the linear regression data fit.

Conclusions

We have characterized the relative SHG intensity of various metal and semiconductor thin films using a 1561 nm p-polarized incident source. In cases where published experimental data for SHG was available, we found that our relative SHG intensities matched the relative intensities from the published data. In the case of W and Sb thin films where little or no experimental data is available for surface SHG, we can draw useful conclusions on the strength of their surface second-harmonic nonlinearities. We can see clearly from Fig. 3 that W has a comparable second-harmonic response to Au and Ag and the response of antimony is closer to that of Cr, Zn and Ge. This experimental data serves as a first point of reference for the surface SHG response of thin films whose experimental data is not readily available.

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