Terahertz Transmission Spectroscopy of Chalcogenide Glasses

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***Abstract*—Terahertz transmission of a chalcogenide family of glasses is reported. The glasses were processed using established methods for non-oxide glasses. Transmission through the samples was measured using a THz spectrometer. The transmission and optical properties of the glasses are summarized. The results show promise of these glasses for application as THz windows.**

I. Introduction And Background

Terahertz (1 THz = 10^{12} Hz) [1] science and technology has emerged promising for potential applications in many scientific and technological fields [2-4]. With advancements in THz radiation sources [5] and detectors [6], there is also increasing need for development of relevant THz optical components for controlling and manipulating THz waves such as lenses, filters, switches, polarizers, and attenuators. This is vital for device development to fill the THz gap existing at ~ 0.3-20 THz [7].

Elements found in group 6 of the periodic table (except oxygen) are commonly termed chalcogens. Chalcogenides contain one or more of sulfur, selenium and tellurium in addition to other elements. In a glass form, the chalcogenides have many interesting and unique properties but most importantly a transmission range that extends to wavelengths far beyond the range of silica and other glasses (e.g., long-wave infrared regime). We report their transmission and optical properties extending well into the THz regime.

II. Methods

We chose five stoichiometries in the simple binary As-S system to study the transmission properties in THz regime. The glasses were processed in Non-Oxide Materials Synthesis Laboratory (NOMSL) at PNNL. All samples were prepared as a window (or lens) with an optical quality polish (1 µm diamond suspension) which were ~ 2-3 mm thick. We used a backward wave oscillator (BWO) based THz spectrometer (Microtech Instruments, Inc.) for transmission measurements were taken in the frequency range 172-506 GHz; see Figure 1 for the full transmission spectrum in this range for $As_{40}S_{60}$.

III. Results & Discussion

A representative spectrum with fitting results is shown in Figure 1. Software was used to fit the Fresnel expressions for complex transmission to the resulting graph to determine the transmission, Tan, refractive index (n), and dielectric properties: extinction coefficient (k), dispersion (E'), loss (E''), and conductivity (S_g) of each sample as a function of frequency. That data is presented in Table 1.

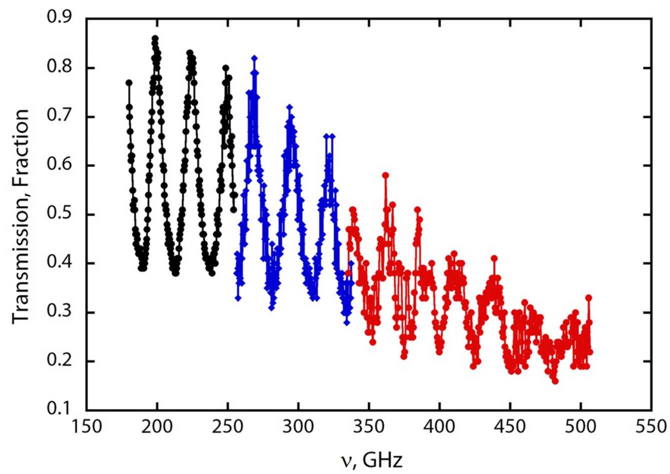

Figure 1. Transmission as a function of frequency of a representative glass ($As_{40}S_{60}$)

Table 1. Summary of properties obtained using THz spectrometer

Chemistry	ν (GHz)	n	k	E'	E''	Tan	S_g $(\Omega\text{-cm})^{-1}$
$As_{30}S_{70}$	193.91	2.424	0.00446	5.886	0.0216	0.00368	0.00233
2.25mm	216.63	2.427	0.00377	5.889	0.0183	0.00311	0.00221
	248.11	2.430	0.00617	5.907	0.0300	0.00507	0.00413
$As_{33}S_{67}$	188.95	2.535	0.00426	6.426	0.0216	0.00336	0.00227
2.82mm	209.02	2.538	0.00479	6.441	0.0243	0.00378	0.00283
	230.12	2.541	0.00559	6.456	0.0284	0.00440	0.00363
	250.88	2.544	0.00659	6.471	0.0335	0.00518	0.00468
$As_{36}S_{46}$	190.91	2.645	0.00348	6.994	0.0184	0.00263	0.00195
2.09mm	214.44	2.649	0.00453	7.015	0.0240	0.00342	0.00286
	244.3	2.654	0.00626	7.043	0.0332	0.00471	0.00451
$As_{40}S_{60}$	199.79	2.795	0.00596	7.813	0.0333	0.00427	0.00370
2.15mm	223.89	2.795	0.00562	7.813	0.0314	0.00402	0.00391
	249.72	2.795	0.00701	7.813	0.0392	0.00501	0.00544
$As_{42}S_{58}$	190.68	2.713	0.00387	7.359	0.0210	0.00285	0.00223
2.04mm	214.09	2.718	0.00567	7.386	0.0308	0.00418	0.00367
	243.84	2.724	0.00764	7.420	0.0416	0.00561	0.00564

The results show a maximum transmission of these glasses in the range of about 20-95% confirming their promise for use in design and fabrication of THz optical components. The general trend being that the transmission peak maxima decrease with increasing frequency. Systematic increase of the refractive index (n) with increasing As-content in the glasses (up to 40 mole %) demonstrates the effect of chemistry on properties however, n decreases as As-loading is increased past 40 mole %. This enables compositional control of THz properties of this family of glasses. These glasses showed more absorption, compared to fused quartz, most likely due to chemical impurities (e.g., water, oxygen) from the starting chemicals.

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The data in Figure 1 was obtained using different frequency multipliers used in THz spectrometer to extend the frequency range covered and therefore the three data sets do not line up exactly with their neighboring datasets but the fit is close aside from a disconnect at ~255 GHz and a decreased amplitude peak maxima at ~345 GHz. At higher frequencies (ν > 450 GHz), the data gets fairly scattered most likely due to low signal (low transmission) to the detector. The highest transmission amongst the glasses tested was with $As_{36}S_{64}$ where the maximum transmission was 0.95 at ~189 GHz.

Figure 2 shows the chemical compositional dependence of refractive index of these glasses. The refractive index increases with As content from 30-40% linearly after which there is a sharp decrease, suggesting a structural change. Within the linear region, these glasses show a good compositional flexibility, demonstrating the ability to change the refractive index by changing the chemistry. This is valuable in designing and building optical components for various THz applications.

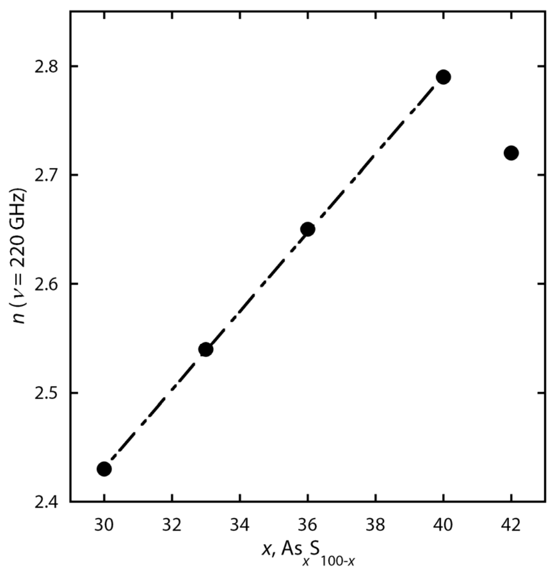

Figure 2. Index of refraction, *n*, correlation to the arsenic content in the glasses.

IV. CONCLUSION

This family of glasses shows promise for applications as passive components in the THz regime. It is believed that the drop in the value, n at $x > As_xS_{100-x}$ seen in Figure 2, is due to a structural transformation that occurs as determined by other means of analyses performed on these specimens. In order to fully understand the structure-property relationship in these glasses, several new glass compositions need to be made throughout the region tested here to fill in the gaps, primarily around $39 < x < 43$.

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