CHALCOGENIDE MATERIALS FOR NONLINEAR PHOTONICS IN THE NEAR AND MIDDLE INFRARED

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Paper Summary
Chalcogenide glasses have amongst the highest third order nonlinearity of any amorphous material, negligible nonlinear absorption and excellent mid infrared transparency. This talk summarizes recent progress on nonlinear devices based on chalcogenide glass waveguides.

Introduction
The third order nonlinearity of an optical waveguide has been extensively employed for high speed, high bandwidth signal processing in telecommunications [1]; in quantum optics, for example, for the generation of correlated photon pairs [2]; and to create novel broadband sources via supercontinuum generation [3]. A major challenge for such applications is to create waveguides that have very large nonlinear response combined with negligible linear and nonlinear losses since this will lead to high efficiency and low operating powers in devices that can be integrated into photonic "chips".

The real and imaginary parts of the ultra-fast third order nonlinearity of any material are related via a Kramers-Kronig relation which means that in many materials the real part of the nonlinearity that is required for the applications is always accompanied by a potentially detrimental imaginary part associated with two-photon absorption (2PA). In the 1990s various figures of merit were introduced to characterize materials for waveguide nonlinear optics, with $FOM_{2PA}=n_2/\beta\lambda$ used to characterize the influence of two-photon absorption [4]. Here n_2 is the nonlinear refractive index; β is the two-photon absorption coefficient; and λ is the wavelength. For 2PA to be negligible the condition $FOM_{2PA}>>1$ is required. Unfortunately, in many materials used to create compact waveguides for optical signal processing FOM_{2PA} *is* ≤ 1, and this means that nonlinear losses effectively reduce the nonlinear phase change that can be achieved in the device. For example, crystalline silicon (c-Si) at 1550nm has $FOM_{2PA}=0.4$, whilst the values for amorphous silicon (a-Si:H) are significantly better at ≈5.

Silicon, however, generally suffers from an additional problem that nonlinear absorption creates free carriers that themselves increase the losses. According to models, nonlinear absorption will be present when the photon energy is more than half the bandgap energy of the material: $h\nu \geq 0.5\ E_g$ where E_g is the bandgap energy and ν the laser frequency. In the case of c-Si with a bandgap of 1.15eV, the telecommunications band clearly falls between the single and two-photon absorption edges. According to the predictions of authors such as Dinu [5], two photon absorption will only vanish when $h\nu \leq 0.5\ E_g$ which corresponds to wavelengths beyond 2.2μm for c-Si. Thus, to improve FOM_{2PA} requires materials with a wider bandgap. However, the nonlinear polarizability of any material is proportional to its linear polarizabilty, which is manifested through the semi-empirical Miller's rule which related the n_2 to the linear index, n_o, by the relation $n_2 \propto (n_o^2-1)/n_o^2$. Thus, high index materials with bandgaps > 1.6eV are desired for signal processing at 1550nm.

One material system that satisfies these requirements are the so-called chalcogenide glasses which contain one or more of the chalcogen elements, S, Se and Te, covalently bonded to network-formers such as As, Sb, Si, Ga, Ge, La, etc. Typically, these glasses have refractive indices in the range 2-3 and bandgaps in the 1.4-2eV range. Third order nonlinearities similar to those of c-Si have also been reported. This combination of properties leads to FOM_{2PA}≈100 for many glass compositions. Being composed of covalently-bonded heavy elements also means that these glasses are transparent far into the mid-infared because the vibrational frequencies of the chemical bonds are low. Typically sulphides transmit to around 8μm, selenides to around 14μm and tellurides to beyond 20μm. Thus, the chalcognides appear uniquely suitable for nonlinear photonic in the near- and mid-infrared [6].

Uses of chalcogenides
Over the past decade the teams in CUDOS have become world-leaders in the use of the nonlinear properties of chalcogenide glasses for signal processing in the telecommunications band. The absence of nonlinear and free carrier absorption combined with waveguide nonlinearities of the same order as those achievable in c-Si have allowed, for example, all-optical processing with THz bandwidths [7,8]. In addition other unique properties of the chalcogenides have been being exploited such as their large Brillouin coefficients [9].

In spite of these impressive results, chalcogenide suffer some limitations. In particular, they are notorious for their photosensitivity – the tendency for their optical properties to change when exposed to intense light [6]. Although this is primarily a problem when exposed to wavelengths near their band-edge, photosensitivity has

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also reported in materials exposed in their transparencey window in the infrared [10]. In addition the most widely used chaclogenides contain Arsenic as a network-former which is unacceptable for some applications. Finally their smaller refractive index relative to silicon means that the mode area of the most chalcogenide nanowires is larger than in devices made from c-Si or a-Si:H and this can only be compensated by using glasses with extremely high nonlinearity and high refractive index.

In this context, we have undertaken a wide study of structure–property relations for chalcogenide glasses from which we attempted to identify the best glasses for nonlinear photonics. An example of our results is presented in Fig 1, where we plot n_2 vs n_o for a wide variety of glasses. These results overwhelmingly demonstrate the relevance of Miller's rule in determining glass nonlinearity.

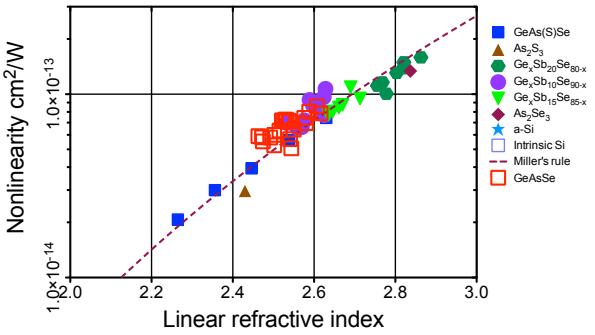

Fig 1: Third order nonlinearity of a wide range of chalcogenide glasses at 1550nm vs linear index. The dashed line is a fit to the semi-empirical Miller's rule. The most nonlinear glasses in this case belong the the family of GeSbSe materials which have n_2 values ≈500 fused silica.

Whilst on the basis of these data one might be tempted to chose the most nonlinear glass, other factors come into play. In fact as n_o increases in general E_g decreases and hence $h\nu \approx 0.5E_g$ and 2PA can become an issue. Furthermore, the photosensitivity of the glasses is affected by composition. Hence for any given glass system, photosensitivity is only small for some compositions. Low photosensitivity correlates with the "fragility" of the particular glass-former. The fragility index, m, is determined from the thermal properties of the glasses and, as shown in Fig. 2, varies with compsosition [11]. Here composition is characterized by a single parameter known as the mean coordination number, MCN. MCN is the sum of the products of the valency of the individual elements in the glass multipled by their abundance. Broadly speaking glasses with the minimum fragility index are the most stable, and Fig. 2 shows that several glasses with MCN around 2.5 are favoured.

Based on results such as these we have chosen a few glasses for device fabrication. In particular, we chose a somposition $Ge_{11.5}As_{24}Se_{64.5}$ ($Ge_{11.5}$) as an example of a glass with near minimum fragility and low photosensitivity. This glass has an linear refractive index of around 2.62 and corresponds to the most right hand of the red squares in Fig. 1.

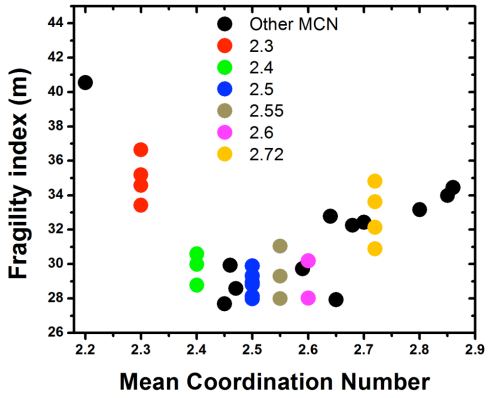

Fig. 2. Measured fragility index, m, of glasses in the GeAsSe system as a function of MCN. Smaller values of m indicate the strongest glass formers in the system [11].

Optical devices based on $Ge_{11.5}$

We have fabricated a range of devices from $Ge_{11.5}$ glass including polarization independent nanowires [12] and high-Q photonic crystal microresonators which could confirm the absence of infrared photosensitivity in this glass composition at the few ppm level [13]. The nanowires have been used to demonstrate correlated photon pair generation [14]. More recently we have been using $Ge_{11.5}$ as a waveguide core material combined with a $Ge_{11.5}As_{24}S_{64.5}$ cladding to create linear and nonlinear devices for the mid-infrared. We recently reported the low loss waveguides for the full functional group band of the mid infrared designed for evanescent field sensing [15] and have now used these same waveguide for mid infrared supercontinuum generation (SC).

Some results on SC generation are shown in Fig. 3. These illustrate the best long wavelength extension reported so far from any material with the spectrum extending over more than two octaves from ≈1.8μm to beyond 7.5μm. The average power in this SC was ≈20mW when about 25mW of average pump power consisting of a 21MHz train of 320fsec pulses at 4μm was coupled into a 1cm long $Ge_{11.5}$ rib waveguide. At the long wavelength end the spectrum is truncated by the cut-off of this particular waveguide that occurs just beyond 8μm as well as increases losses due to the existence of a thin fluoro-polymer top cladding on the waveguide. Nevertheless this is an encouraging results since this source is around 100 x brighter than current synchrotrons with a similar average power. We have developed waveguide designs that should allow the SC spectrum to be extended to around 11μm and average power improved to around 60mW. Such a source would certainly be a candidate for the sort of diagnostic imaging based on microspectroscopy that can currently only be performed using synchrotrons.

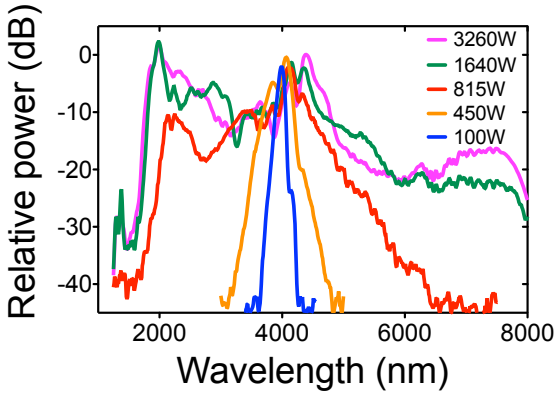

Fig. 3. Supercontinuum spectra obtained from a 2.5µm thick x 4µm wide $Ge_{11.5}$ rib waveguide pumped with 320fs pulses at 4µm. More than two octaves spectral coverage was obtained with the emission extending to beyond 7.5µm. The different curves represent different peak powers coupled into the waveguide

Conclusions

The optical properties of chalcogenide glasses make them very suitable for waveguide nonlinear optics in the near and middle infrared. However, care must be taken to choose the right glass composition for waveguide applications if a combination of high nonlinearity and good stability is required.

Acknowledgements

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