



Chalcogenide glasses in active plasmonics

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The phase-change technology behind rewritable optical disks and the latest generation of electronic memories can also offer high contrast plasmonic switching functionality. Numerical simulations illustrate the extent of this potential while pre-

liminary experiments show that a silver/gallium-lanthanum-sulphide interface can support surface plasmon-polaritons and demonstrate the principle of plasmonic modulation through reversible photo-induced changes in the chalcogenide.

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1 Introduction Much recent interest in the field of plasmonics has been driven by the prospect of high-bandwidth, nanoscale data transport and processing systems based on surface plasmon-polaritons (SPPs) as information carriers and an enhanced plasmonic synergy between today's microelectronic and photonic technologies. 'Active plasmonic' devices able to dynamically switch and modulate SPP signals will be crucial to such applications and a range of functional media have been investigated for this purpose in recent years [1]. With proof-of-principle demonstrations pushing performance limits into technologically competitive terahertz modulation frequency [2] and femtojoule switching energy [3] domains, increasing attention is being given to practical issues like CMOS (complementary metal-oxide semiconductor) and/or SOI (silicon-on-insulator) process compatibility and the long-term performance characteristics of switching media.

2 Concept Here we consider the use of chalcogenide glass media in active plasmonic switching devices and present a proof-of-principle demonstration based on reversible photo-induced changes in the optical properties of a chalcogenide thin film. The chalcogenides – compounds containing heavier Group-16 elements (S, Se, Te) – are a remarkable family of materials, displaying an extensive range of technologically relevant optical, electronic, thermal and mechanical effects [4], including the ability to be

reversibly switched on the nanosecond timescale between structural phases with markedly different optical properties [5]. They are also physically robust and their properties (e.g. refractive index, resistivity) can be tuned substantially through compositional variation [6]. As such, chalcogenide glasses have been in widespread commercial use for many years in re-writable optical discs (where data is routinely encoded at more than 200 Mbit/s) [7] and are emerging as the basis of next-generation electronic memories, where bit sizes may be as small as a few tens of nanometers and switching energies are on the order of picojoules [8]. These properties are also well suited to nanophotonic switching applications in nanoscale volumes of material [9]. Indeed, a metamaterial electro-optic switch exploiting chalcogenide phase-change properties in a device of sub-wavelength thickness has recently been demonstrated [10].

3 Chalcogenide plasmonics: Modeling To understand how photo-induced changes in chalcogenides affect plasmonic functionality, we consider the Kretschmann configuration for the excitation of SPPs on a chalcogenide-metal interface. Germanium antimony telluride (GST) is by far the most extensively studied and well-characterized chalcogenide system. However, a high index of refraction and substantial absorption in the visible to near-IR part of the spectrum complicate its application to

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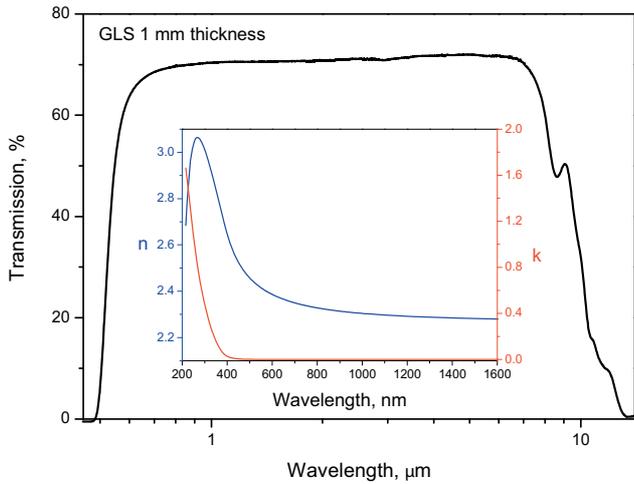


Figure 1 (online colour at: www.pss-rapid.com) Optical transmission of a 1 mm thick amorphous GLS glass sample in the VIS–IR range. The inset shows the real (n) and imaginary (k) parts of the refractive index in the VIS–NIR range, derived from ellipsometric measurements.

plasmonics. We turn therefore to gallium lanthanum sulphide (GLS), a semiconducting chalcogenide with a transparency band extending from 500 nm to $\sim 10 \mu\text{m}$ [11] (see Fig. 1). Bulk samples and sputtering targets in this novel glass composition are prepared by melt quenching and annealing a mixture of high-purity gallium sulphide and lanthanum sulphide precursors.

The transfer-matrix method (TMM) [12] was employed initially to analyze a three-layer Kretschmann system of rutile (TiO_2), silver and GLS as shown in Fig. 2a at the telecommunications wavelength of 1550 nm. Refractive index data for amorphous GLS were derived from ellipsometric measurements (inset to Fig. 1, $n_{1550\text{nm}} = 2.281$, $k_{1550\text{nm}} = 0$); values for TiO_2 and silver were taken from Refs. [13] and [14] respectively. Figure 2b shows reflectivity R as a function of incident angle θ and silver film thickness t for p- and s-polarized light at a wavelength of 1550 nm. The pronounced reflectivity dip associated with the excitation of SPPs on the metal/GLS interface is present only in the p-polarized case for silver thicknesses of a few tens of nanometers. Figure 2c shows cross-sections of these plots at $t = 45 \text{ nm}$ and illustrates the quality of the coupling resonance ($R \rightarrow 0$ over a narrow angular range).

For practical applications one must consider finite GLS film thicknesses. To a point, thinner (sub-micron) films are easier to fabricate and show better switching attributes (faster, lower energy). At the same time, as Fig. 3 shows, the resonant SPP coupling characteristics in the Kretschmann geometry depend strongly on chalcogenide film thickness t_2 . For a given silver film thickness, the coupling angle for SPP excitation decreases sigmoidally with GLS thickness between about 500 nm and 50 nm as shown in Fig. 3c. Thus, by controlling the chalcogenide film thickness one may substantially tune the plasmonic properties of the structure.

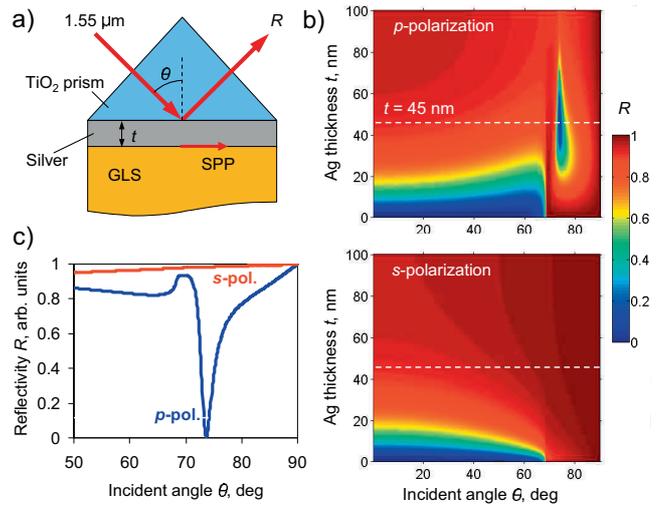


Figure 2 (online colour at: www.pss-rapid.com) (a) Modeled Kretschmann geometry for SPP excitation on a silver/GLS interface; (b) reflectivity R of the structure shown in part (a), for p- and s-polarized light at 1550 nm as a function of incident angle θ and silver thickness t ; (c) cross-sections of the plots in part (b) showing R as a function of θ at $t = 45 \text{ nm}$.

4 Chalcogenide plasmonics: Experiment To confirm that high-quality, stable GLS/silver interfaces can be formed and that they support SPP propagation, the hypotenuse face of a right-angle TiO_2 prism was sputter-coated with 30 nm of silver and 160 nm of amorphous GLS. The reflectivity of this structure was measured via the prism as a function of incident angle using both p- and s-polarized

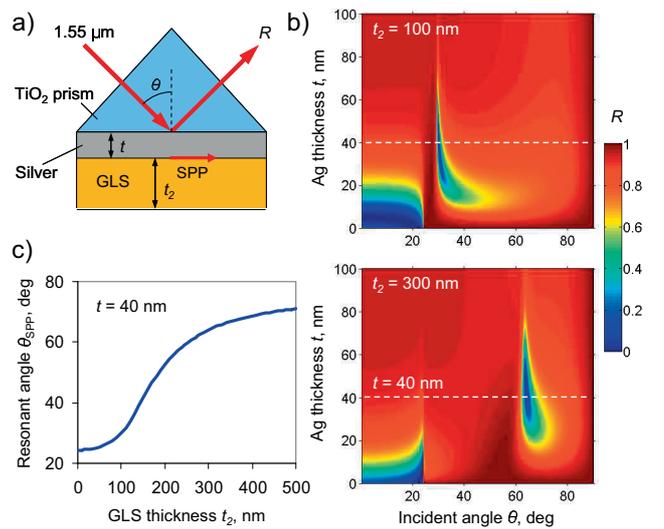


Figure 3 (online colour at: www.pss-rapid.com) (a) Modeled SPP coupling geometry with finite GLS thickness t_2 ; (b) reflectivity R of the structure shown in part (a) for p-polarized light at 1550 nm as a function of incident angle θ and silver thickness t for different thicknesses t_2 of GLS (as labeled); (c) angular position of SPP coupling resonance (min. R) as a function of t_2 at a fixed silver thickness $t = 40 \text{ nm}$.

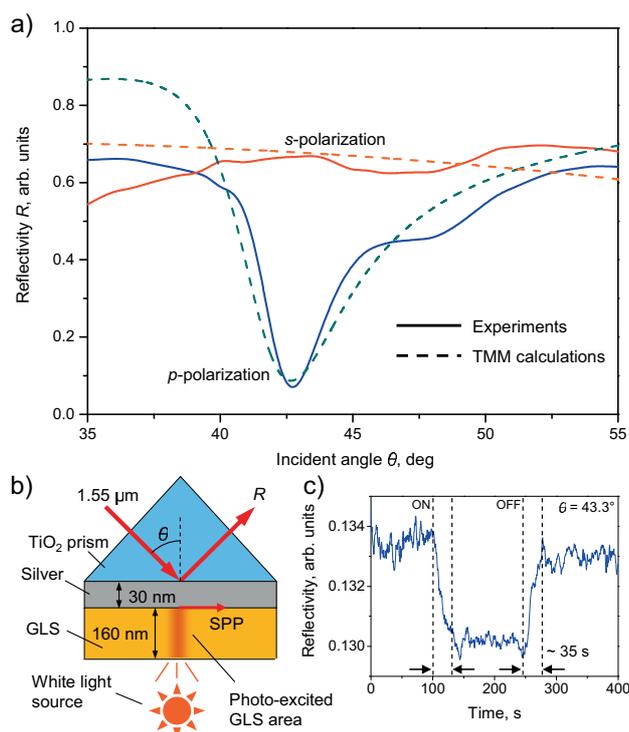


Figure 4 (online colour at: www.pss-rapid.com) (a) Experimentally evaluated reflectivity R of a (TiO₂ prism – 30 nm silver – 160 nm GLS) Kretschmann SPP-coupling configuration as a function of incident angle for p- and s-polarized 1550 nm light (solid lines) overlaid with curves derived analytically using the transfer matrix method (GLS refractive index 2.2, dashed lines); (b) experimental configuration for plasmonic modulation via photo-excitation of GLS; (c) at off-resonance incident angles, white light illumination of GLS produces a modulation of SPP coupling efficiency manifested as a modulation of R .

incident beams at a wavelength of 1550 nm. These data are shown in Fig. 4a alongside computational results: As expected, the reflectivity dip associated with SPP coupling is only observed for p-polarized light, in the present case at an incident angle $\theta_{\text{SPP}} = 42.7^\circ$; good agreement is achieved between experiment and analytical model with a GLS refractive index of approximately 2.2.

Photo-induced changes in the chalcogenide excited by low-intensity white light (via one or a combination of photo-refractive, -darkening and -expansive effects [15, 16]) provide an accessible mechanism for low frequency transient plasmonic modulation in the experimental Kretschmann geometry. With the incident angle for the probe beam detuned from resonance (i.e. to a point where $|dR/d\theta|$ is high), the GLS from the back was subjected to white light illumination (halogen source via a fiber-bundle) at intensities up to 630 mW/cm² (Fig. 4b, c). This brings about a reversible change in the reflected probe intensity, indicating a photo-induced change in the efficiency of plasmonic coupling to the silver/GLS interface. Relative reflectivity changes of up to 3% were observed for detunings of $\pm \sim 0.6^\circ$ from θ_{SPP} . The changes are slow (saturating

over ~ 35 s) and maintained only as long as the illumination was present. The sign of the induced change is negative for $\theta > \theta_{\text{SPP}}$ and positive for $\theta < \theta_{\text{SPP}}$, suggesting that the photo-excitation of GLS shifts the SPP coupling resonance to larger angles. A TMM fitting indicates an induced refractive index change in the GLS of $\sim 10^{-3}$ (assuming no change in absorption or GLS thickness).

When one considers that amorphous-crystalline phase transitions in chalcogenides may bring about a relative index change approaching 0.1 [6] on nanosecond timescales, the great potential held by chalcogenides in the plasmonic domain becomes clear: In the Kretschmann geometry considered above, a change of this magnitude would dramatically shift the angular position of the SPP coupling resonance and provide a switching contrast ratio in excess of 100:1 (depending on GLS thickness). For device applications in plasmonic waveguide architectures one would, for example, only need to transform GLS over a path length of $\sim 1.5 \mu\text{m}$ (about one wavelength) within a Mach-Zender interferometer operating at 1550 nm to achieve the π phase-shift required for high contrast output modulation/switching (assuming an SPP refractive index for a planar gold/GLS interface of $\sqrt{\epsilon_{\text{Au}} \epsilon_{\text{GLS}} / (\epsilon_{\text{Au}} + \epsilon_{\text{GLS}})}$, where ϵ_{Au} and ϵ_{GLS} are the complex relative permittivities of gold and GLS, respectively).

5 Conclusions In summary, analytical modeling and preliminary experimental demonstrations employing gallium lanthanum sulphide as a photo-active medium illustrate that chalcogenide glasses – robust, commercially established, CMOS/SOI-compatible media – may offer practical active plasmonic switching/modulation functionality for future nanophotonic device applications.

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