Optical modulation of surface plasmon polariton coupling in a gallium/aluminium composite

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We report on the use of a reversible nanoscale light-induced structural transformation in a new type of gallium/aluminium composite material interfaced with a dielectric to control the efficiency with which light is coupled to a surface plasmon-polariton wave at the interface, and thereby the optical reflectivity of the interface. An optical fluence of just a few mJ/cm^2 is sufficient to significantly modulate the composite's dielectric properties and thereby to provide high-contrast nonlinear optical and plasmonic switching functionality on a nano- to microsecond timescale, for signals in the visible and near infrared wavelength ranges.

There is currently great interest in the possibility of constructing highly integrated photonic devices with sub-wavelength structural elements by using metal/dielectric nanostructures to guide and manipulate signals in the form of surface plasmon-polariton (SPP) waves (i.e. optical excitations coupled with collective electronic excitations on metal/dielectric interfaces [1, 2]).

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Numerous structures for generating and passively manipulating SPP signals have been demonstrated [3, 4] but we will only be able to speak about 'plasmonics' as we do 'photonics' when efficient active SPP signal manipulation techniques are developed. There is also growing interest in the use of materials that undergo structural transformations between forms with differing optical and electronic properties for nanoscale memory and photonic switching applications.[5-7] We report here on the use of a reversible light-induced structural transformation in a Ga/Al composite material interfaced with a dielectric to control the efficiency with which light is coupled to an SPP wave at the interface and thereby to modulate the optical reflectivity of the interface with very high contrast.

Due to the mismatch between their wave vectors light cannot be coupled directly to an SPP wave on a smooth interface. The discrepancy can be compensated for by a grating on the interface or via the attenuated total reflection (ATR) of incident light inside a dielectric-coated prism adjacent to the interface.[1] The efficiency with which SPP waves can be generated in the ATR coupling scheme (and their subsequent propagation the metal/dielectric interface) depend strongly on the dielectric characteristics of the materials within a few tens of nanometers of the interface. These processes may thus be controlled by changing the dielectric properties of the metal at the interface.[8] Gallium is particularly suited to this application because there is a marked difference, across a broad visible to near-IR wavelength range, between the dielectric properties of its assorted structural phases:[9, 10] The SPP decay length in its almost ideally metallic liquid and metastable solid phases (5–20 μm at wavelengths between 700 and 1500 nm) is approximately ten times longer than that in the stable solid α-phase (a partially covalently bound crystalline form). Furthermore, a surface-driven transition between the solid and metallic phases can be reversibly stimulated at an interface by low power optical excitation.[11]

The Ga/Al composite used in the present study comprises a polycrystalline aluminium film wherein the grain boundaries and the substrate interface are infiltrated with nanolayers of gallium. It retains the optical switching functionality of gallium, while the framework of aluminium domains supports the formation of a dielectric interface that is more robust and of more reliable uniform quality than can be achieved with pure gallium using comparably simple production techniques. It was prepared on the hypotenuse face of a BK7 glass prism previously coated with a 185 nm MgF_2 layer (see Fig. 1a) by first evaporating a 250 nm aluminium film onto the prism. Then while maintaining the prism at a temperature just above gallium's melting point (29.8°C), a drop of liquid gallium was applied to the surface of the aluminium. The composite structure is formed as liquid gallium breaches the oxide layer on the aluminium film, spreads across its surface and penetrates its grain structure through to the MgF_2 interface, and is fixed by cooling it to solidify the gallium component. The metals have limited mutual solubility [12] but the Ga/Al system is known for the rapidity of its 'grain boundary penetration' process.[13-16] The optical quality of the silica-metal interface is very high but the presence of gallium is easily detected because the silver/grey tone of the Ga/ Al-silica interface is slightly darker than that of the Al-silica interface. Electron backscatter diffraction imaging indicates that during composite formation the liquid gallium fractures the oxide layer as it spreads across the surface of the aluminium, and that it solidifies primarily in the α phase.

At angles of incidence greater than ~60°, *p*-polarized probe light from a He-Ne laser (λ = 633 nm) is internally reflected at the glass/MgF_2 interface, producing an evanescent wave in the MgF_2 layer. At an incident angle $\theta \sim 65°$ the interface projection of the wave vector of the evanescent wave is equal to the SPP wave vector for the glass/MgF_2/composite structure and the energy of the incident beam is efficiently (resonantly) coupled to an SPP wave. Under these conditions the reflectivity of the structure is very low (<0.5%) but if the gallium component of

the composite is converted from the α phase to the liquid state (either by changing the temperature or applying an external optical excitation [11]) the system is driven away from resonance, there is a significant increase in the reflectivity of the interface, and a corresponding decrease in SPP coupling efficiency. Fig. 1b shows how the contrast between the α and liquid state reflectivity levels depends critically on the angle of incidence.

For the purposes of the present study, reversible transitions to the metallic state were induced using 6 ns pulses from a Nd:YAG pump laser (λ = 1064 nm, rep. rate = 20 Hz). The pump and probe laser spots (with diameters of ~4 and ~1 mm respectively, continuous-wave probe power ~0.27 mW, angle of probe incidence $\theta_c \sim 65°$) were overlapped on the dielectric/composite interface and the reflected probe signal was monitored using a digital oscilloscope. The dynamics of the pump-induced probe reflectivity modulation are illustrated in Fig. 2 - the reflectivity increases sharply on application of a pump pulse, indicating an abrupt decrease in the efficiency of SPP coupling, and then recovers its pre-excitation level on a nano- to microsecond timescale.

For a given level of pump excitation, the magnitude ΔR of the induced reflectivity change and the half-maximum relaxation time τ following termination of the pump pulse are both functions of temperature (see Fig. 3). With increasing temperature, ΔR peaks several degrees below T_m, the melting point of the composite's gallium component, and falls to zero at T_m. The corresponding relaxation time increases rapidly, from substantially less than a microsecond to several microseconds, as ΔR peaks and begins to fall, then falls abruptly itself as the response disappears at T_m. ΔR increases with pump fluence (Fig. 4a) to a temperature dependent saturation level at ~2 mJ/cm^2. The relaxation time τ also increases with fluence, and continues to do so beyond the point where ΔR saturates (Fig. 4b).

These response characteristics are similar to those of pure gallium [11, 17] and confirm that the composite's optical and plasmonic response to laser excitation is underpinned by a reversible, surface-driven structural transition between the α and metallic phases of the gallium component: Gallium exhibits surface melting,[18] so for reasons of surface energy several atomic layers of the metal adjacent to an interface exist in a metallic state even at temperatures below the melting point. With increasing temperature or level of external excitation, the thickness of this metallic layer increases, causing the optical and plasmonic properties of the composite/dielectric interface to change. At a given temperature, higher excitation fluences create a thicker metallic layer and give rise to a larger reflectivity change (Fig 4a). This response saturates as the thickness of the metallized layer exceeds the probe beam skin depth δ. At fixed excitation fluence (Fig. 3), the magnitude of the induced reflectivity change initially increases with temperature as the energy/atom required for metallization decreases. It then decreases at temperatures within a few degrees of T_m because the volume fraction of metallized gallium present within the composite *in the absence* of external excitation increases as part of the normal melting process. The relaxation time required for recrystallization of metallized gallium following the withdrawal of pump excitation increases with both pump fluence and temperature (Fig.'s 3 and 4b) because aside from the fact that (all other factors being equal) a thicker metallic layer takes longer to recrystallize, the recrystallization velocity v is proportional to $|T\text{-}T_m|$, i.e. it decreases as the temperature approaches T_m.[19]

In contrast to pure gallium,[17] the pump fluences required to saturate the response of the composite medium are somewhat lower (~2 mJ/cm^2 instead of ~15 mJ/cm^2). A number of factors may contribute here: For example, the thickness of the gallium nanolayer adjacent to the MgF_2 interface may be smaller than the metal's skin depth ($\delta_{\text{Solid Ga}}$ = 13–20 nm at visible wavelengths depending on crystalline orientation). Furthermore, because it exists as a network of thin films

between aluminium grains, the gallium component of the composite has a much larger surface/interface area than a body of pure gallium. Surface melting may thus occur throughout the gallium structure and not just at the MgF_2 interface, reducing the average energy per atom required for metallization.

To summarize, it has been shown that the efficiency with which light is coupled into an SPP wave at an interface between a Ga/Al composite and a dielectric can be controlled using a reversible nanoscale structural transformation, induced by laser fluences of just a few mJ/cm^2, in the gallium component of the composite. At the same time, by virtue of the fact that resonant probe beam to SPP wave coupling in the ATR configuration reduces the reflected probe intensity to almost zero, the light-induced structural transition also provides very high contrast optical switching.

The authors would like to acknowledge the assistance of M. V. Bashevoy and F. Jonsson in characterizing the composite material by electron backscatter diffraction, and the financial support of the Engineering and Physical Sciences Research Council (UK).

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Figure Captions:

Fig. 1. (a) The arrangement for optical modulation of light-to-SPP wave coupling in the ATR configuration. (b) Angular dependence of reflectivity contrast (defined as $[R_\alpha - R_{liquid}]/R_\alpha$ where R_α and R_{liquid} are the α and liquid state reflectivity levels respectively) for the glass-MgF_2-Ga/Al composite structure shown in (a). Measurements were made using a *p*-polarized 633 nm probe beam. The peak position $\theta_c \sim 65°$ is given by that of the minimum in R_α.

Fig. 2. Dynamics of the change induced in the probe reflectivity of the glass-MgF_2-composite structure by 6 ns pump pulses ($T = 14°C$, angle of probe incidence $\theta_c = 65°$).

Fig. 3. Temperature dependence of the maximum pump-induced change ΔR in the glass-MgF_2-composite structure's reflectivity (closed symbols) and corresponding relaxation time τ (open symbols) at a pump fluence of 2 mJ/cm^2.

Fig. 4. Dependences on pump fluence of (a) the maximum pump-induced change ΔR in the reflectivity of the glass-MgF_2-composite structure, and (b) the corresponding relaxation time τ, at different sample temperatures.

Figure 1

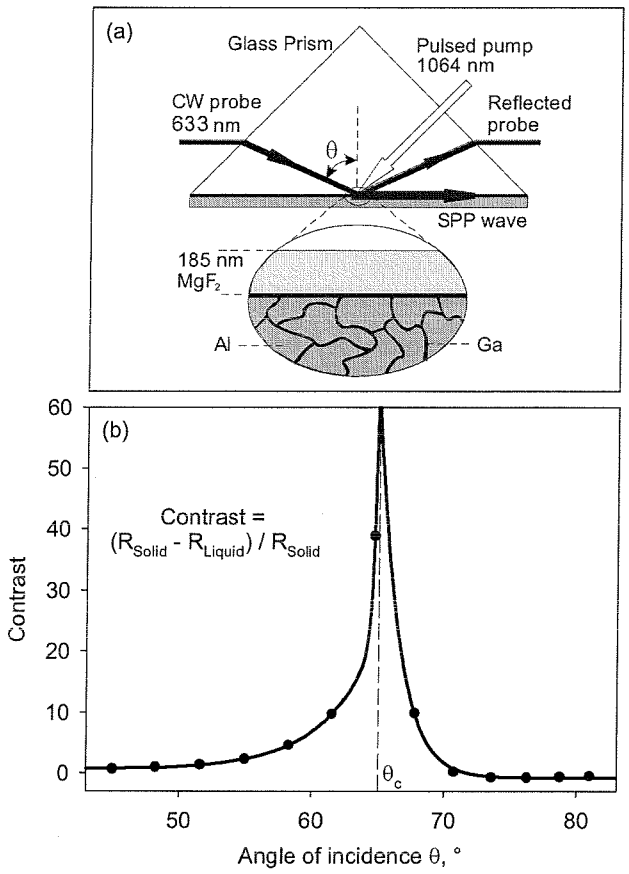

(a)
Glass Prism
Pulsed pump
1064 nm
CW probe
633 nm
Reflected
probe
θ
SPP wave
185 nm
MgF2
Al
Ga
(b)
Contrast =
(RSolid - RLiquid) / RSolid
Contrast
θc
Angle of incidence θ, °

Figure 2

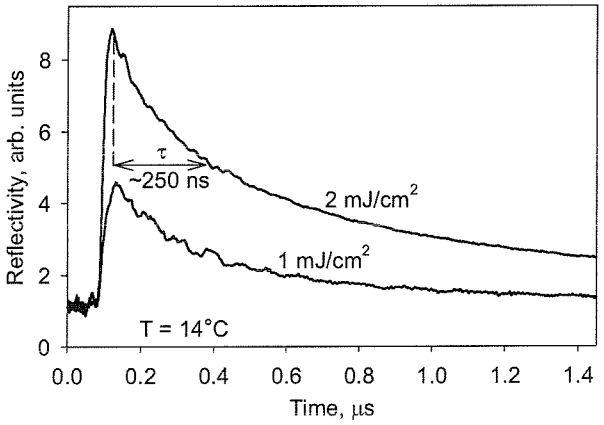

Reflectivity, arb. units
Time, μs
τ
~250 ns
2 mJ/cm2
1 mJ/cm2
T = 14°C
0
2
4
6
8
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4

Figure 3

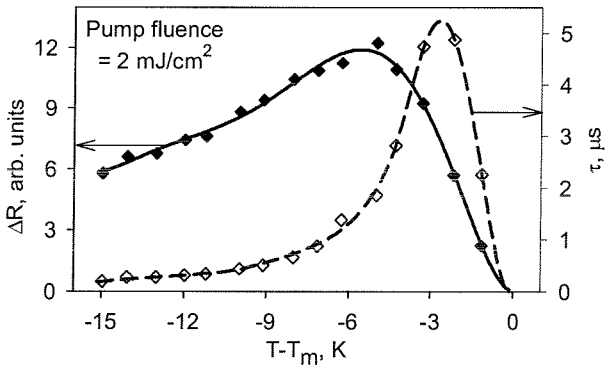
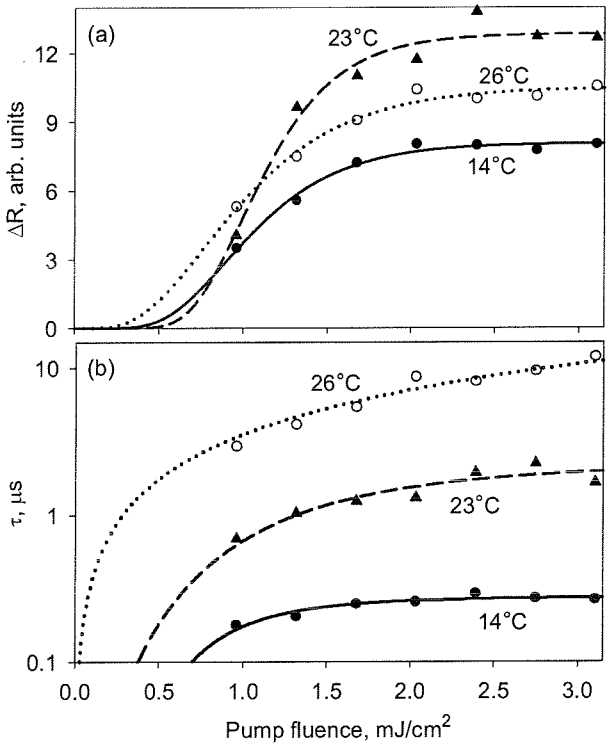

Pump fluence
= 2 mJ/cm2
ΔR, arb. units
τ, ps
T-Tm, K
12
9
6
3
0
5
4
3
2
1
0
-15
-12
-9
-6
-3
0

Figure 4


(a)
23°C
26°C
14°C
ΔR, arb. units
12
9
6
3
0
(b)
26°C
23°C
14°C
τ, μs
10
1
0.1
0.0
0.5
1.0
1.5
2.0
2.5
3.0
Pump fluence, mJ/cm2