High-contrast modulation of light with light by control of surface plasmon polariton wave coupling

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We have demonstrated a mechanism for modulating light with light by controlling the efficiency with which light is coupled into a plasmon polariton wave. An optical fluence of 15 mJ/cm^2 in the control channel is sufficient to achieve nearly a ten-fold intensity modulation of the signal beam reflected from a Glass /MgF$_2$/Ga structure. The mechanism depends on a nanoscale light-induced structural transformation in the gallium layer and has transient switching times of the order of a few tens of nanoseconds. It offers high modulation contrast for signals in the visible and near infrared spectral ranges. © *2004 American Institute of Physics*. [DOI: 10.1063/1.1808240]

Surface plasmon polariton (SPP) waves, i.e., surface electromagnetic excitations coupled with electrons at a metal–dielectric interface,[1–4] are attracting attention as a potential type of information carrier for future highly integrated photonic devices. A range of very promising nanostructures that direct and guide SPP waves and allow for subwavelength structural elements on plasmonic "chips" are now being investigated.[5,6] However, it will not be possible to speak about "plasmonics" in the same way that we speak about "photonics" until techniques for active manipulation of SPP signals are developed. Recent theoretical analysis shows that active switching of plasmonic signals should be possible through a stimulation-induced nanoscale structural transformation in the waveguide material.[7] Here, we report that a light-induced nanoscale structural transformation can be used to control the efficiency with which electromagnetic radiation is coupled into SPP waves, and thus modulate the intensity of optical signals.

SPP waves propagate along the interface between a metal and a dielectric. As the dispersion relation for SPPs is different from that for light, it is only possible to couple light into a SPP wave on a smooth surface by using a matching device such as a grating, or a prism placed at the interface.[1] Effective coupling is possible only in an optimized regime, i.e., for a particular grating period or for a particular angle of incidence through the prism. The coupling efficiency also depends on the dielectric characteristics of the materials in a very thin region around the interface. That part of the light wave which is not coupled into the SPP wave is reflected from the interface. Our modulation concept is based on the idea that by changing the dielectric characteristics of the metal at the interface through a light-induced structural transformation, one can drive the system away from the resonant coupling conditions and thus exercise control over the intensity of the wave reflected from the interface.

Gallium is a uniquely suitable material to realize this concept. It is known for its polymorphism[8] and α-gallium, the stable "ground-state" phase,[9] has a very low melting point, 29.8 °C, and is partially covalent bound. The optical properties of α–Ga and those of the more metallic phases, which are metastable under normal conditions, are very different. The properties of the metastable phases are similar to those of the highly metallic liquid phase. In terms of the dielectric coefficients at a wavelength of 780 nm, $|\varepsilon_{\text{liquid}}|/|\varepsilon_\alpha| \sim 5$. A metastable metallic phase (quasi-melt) may be achieved at the interface by simple heating, or by light absorption through a nonthermal "optical melting" mechanism based on the destabilization of the optically excited covalent bonding structure, which only affects a few atomic layers of the material at an interface.[10] Such structural transformations have already been shown to provide photonic functionality, offering, for example, all optical switching at normal reflection from bulk interfaces[11] and nanoparticle films.[12] Here it is demonstrated experimentally that the use of a stimulated transformation to control the coupling between light and SPP waves provides modulation contrast more than one order of magnitude higher than at bulk interfaces (at normal incidence) and about two orders of magnitude higher than for nanoparticle films.

In our experiments we used the Otto configuration[1] for coupling light to a SPP wave. Here gallium is interfaced with a BK7 glass prism covered with a MgF$_2$ film of thickness D=185 nm [Fig. 1(a)]. A light wave undergoing total internal reflection on the glass/MgF$_2$ interface is efficiently coupled to a SPP wave at the MgF$_2$/Ga interface at a particular (resonant) angle of incidence where the photon wave vector in glass is equal to the SPP wave vector on the MgF$_2$/Ga interface. A gallium film was prepared on the prism by squeezing a bead of the liquid metal then solidifying it. A continuous wave diode laser operating at λ_p =780 nm was used as a probe source [Fig. 1(a)]. The dependencies of the reflectivity of the glass /MgF$_2$/Ga structure on incident angle for *s* and *p* polarizations are presented in Fig. 2 for two different Ga phases. The reflectivity of the structure has a clear minimum for the *p* polarization, but not for the *s* polarization of the incident light. The reflectivity minimum corresponds to the resonant conditions for coupling light into the SPP wave, which can only be achieved for the *p* polarization.[1]

The resonant SPP coupling is illustrated by numerical modeling of the electromagnetic fields in the interface area, as presented in Figs. 1(b) and 1(c). The gray scale in these

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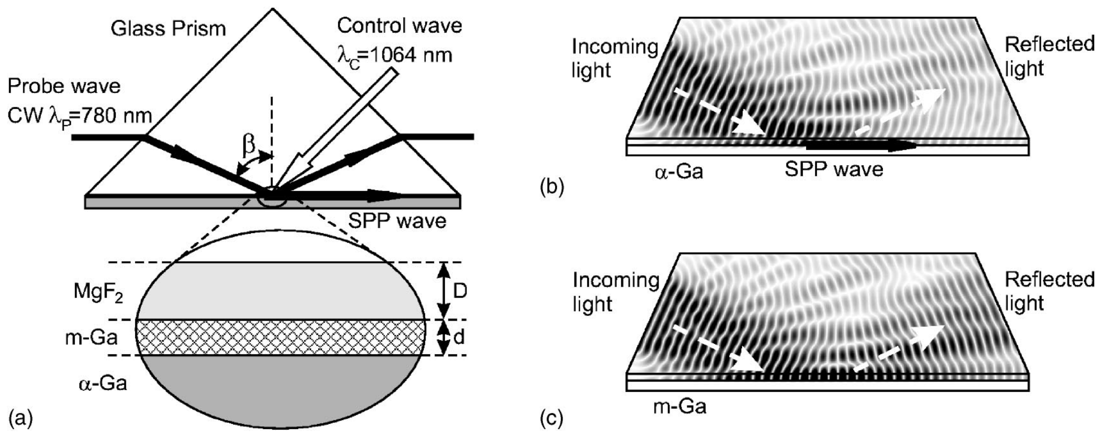

FIG. 1. (a) Arrangements for modulation of light with light and control over light-SPP wave coupling in a glass /MgF_2/Ga structure in the Otto configuration. (b), (c) Field distributions in the vicinity of the glass /MgF_2/Ga structure calculated for (b) α and (c) m phases of gallium.

images represents the amplitude of the magnetic component of the field (darker for higher amplitude). Image (b) shows the case for high coupling efficiency and low reflectance, under resonant conditions of SPP excitation (β=66°), when the energy of the incoming wave is converted into the SPP wave which is rapidly damped because of high SPP wave losses ($\alpha-$Ga phase at the interface). Image (c) shows the case for low coupling efficiency and high reflectance (gallium is in the metallic phase at the interface).

The inset in Fig. 2 shows the interface reflectivity at β =66°, the resonant angle for excitation of the SPP wave on the MgF_2/Ga interface, as a function of increasing thickness d of the metallic layer, calculated using data on dielectric coefficients from Refs. 13 and 14 . From here one can see that by changing Ga from the α phase to the m phase in a layer only a few nanometers thick, the coupling efficiency, and thus the reflected beam intensity at the resonant incident angle, can be changed dramatically.

To demonstrate light-by-light modulation of the reflected probe beam via a structural transformation from the α phase tothe metallic phase in the gallium film, we introduced a channel for optical excitation of the interface with a Nd:YAG laser, generating 6 ns pulses at λ_c=1064 nm with the repetition rate of 20 Hz (Fig. 1). The control and probe laser spots were overlapped on the interface. Stimulation with the control laser leads to an immediate increase in the reflected probe intensity R. At an excitation fluence of Q =15 mJ/cm^2 the obtained reflectivity increase $R_{\text{on}}/R_{\text{off}}$ reaches 9.4, where the subscripts "on" and "off" indicate the state of the pump laser. This significant change in the intensity of the reflected wave corresponds only to about a 20% decrease in the efficiency of coupling into the SPP wave. The magnitude of the effect increases with the fluence up to about 15 mJ/cm^2 and then saturates (insert in Fig. 3). This behavior may be explained as follows: higher fluences of optical excitation create a thicker layer of the metallized phase. The maximum reflectivity increase diminishes with temperature. We studied the transient characteristics of the effect by monitoring the reflected signal with a photodetector and a real time digital scope (see Fig. 3). The overall bandwidth of the registration system was 125 MHz. The transient "switch-on" time has not been resolved in this experiment. It might have been as short as 4 ps, which was the intrinsic transition time for a transformation from the α phase to the metallic phase.[15] For a given excitation level, there is a steep increase in the relaxation time as the temperature of the structure ap-

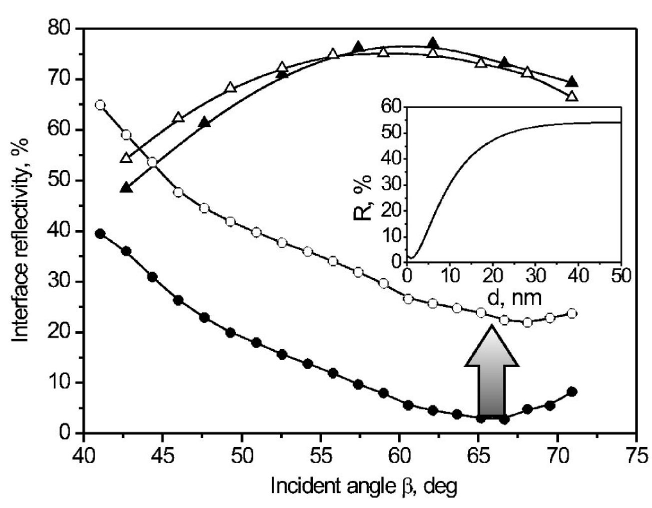

FIG. 2. Angular dependence of reflectivity from the glass /MgF_2/Ga structure. Closed and open symbols represent α and metallic gallium phases, respectively. Triangles and circles denote s and p polarizations of the incident light. The vertical arrow shows the reflectivity change that can be achieved by a complete transformation of the gallium film from the α phase into the metallic phase. The inset shows the theoretical dependence of reflectivity on the thickness d of the metallic m phase of gallium.

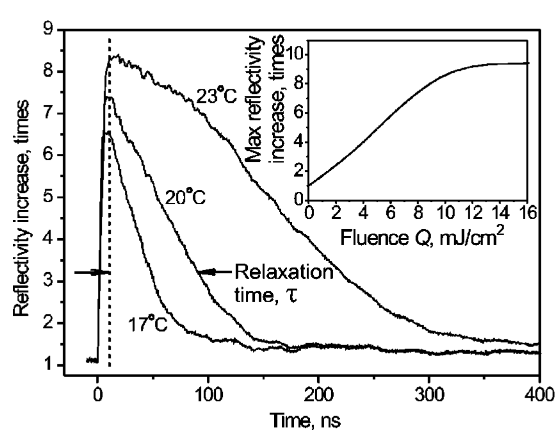

FIG. 3. Transient reflectivity of the glass /MgF_2/Ga structure following 6 ns impulse excitation at a wavelength of 1.06 μm (Q=15 mW/cm^2) for various interface temperatures. The angle of incidence is 66°. The inset shows the dependence of the maximum reflectivity increase on control wave fluence at 28 °C.

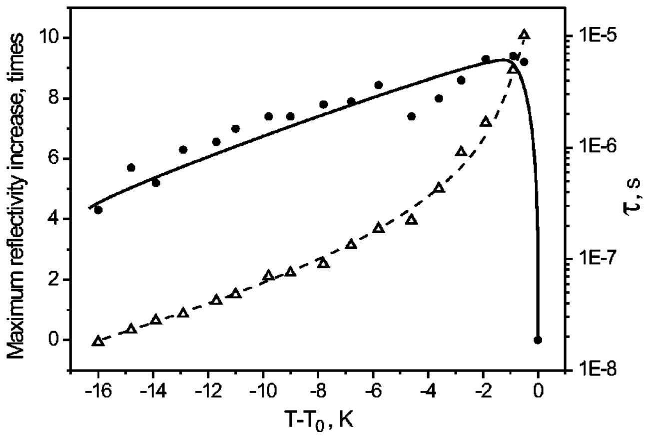

FIG. 4. Temperature dependence of the maximum reflectivity increase (●) and corresponding relaxation time (△) at $Q=15$ mJ/cm^2.

proaches gallium's melting temperature $T_0=29.6\,°C$, while relaxation times as short as 20 ns are observed at temperatures below 14 °C (Fig. 4). This can be explained by the fact that the recrystallization velocity v in turn depends on temperature: $v \propto (T-T_0)$,[16] so the closer the system is to the melting temperature, the longer the time required for the metastable metallized layer to recrystalize back to the α phase. In conclusion, our experiment demonstrates that light-induced metallization of α-Ga under conditions for "resonant" SPP coupling can lead to a very effective modulation of the reflected light intensity. This observation also provides a strong indication that the active plasmonics concept, proposed in Ref. 7, will indeed provide an efficient technique for all-optical modulation of SPP signals with a bandwidth of tens of megahertz. It follows from the previous studies of optically induced metallization of gallium[10] that the modulation mechanism is inherently optically broadband and could provide high modulation contrast for signals in the visible and near infrared spectral ranges.

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