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Optically switchable photonic metasurfaces
R. F. Waters, P. A. Hobson, K. F. MacDonald, and N. I. Zheludev

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Optically switchable photonic metasurfaces

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We experimentally demonstrate an optically switchable gallium-based metasurface, in which a reversible light-induced transition between solid and liquid phases occurring in a confined nanoscale surface layer of the metal drives significant changes in reflectivity and absorption. The metasurface architecture resonantly enhances the metal’s “active plasmonic” phase-change nonlinearity by an order of magnitude, offering high contrast all-optical switching in the near-infrared range at low, $\mu$W $\mu$m$^{-2}$, excitation intensities. © 2015 AIP Publishing LLC.

Photonic metamaterials were conceived and first realized as a means of achieving exotic optical properties, such as negative refractive index and optical frequency magnetism, not available in nature. They have evolved rapidly into a functional platform for the engineering of nanoscale photonic “meta-devices” offering a wide range of switchable, tuneable, and nonlinear properties, typically achieved by hybridizing a plasmonic metal resonator framework with an active medium. Among the latter, phase-change media such as chalcogenide glasses,2,3 vanadium dioxide,4,5 and liquid crystals6–8 have been shown experimentally to offer substantial excitation-induced changes in optical properties, including nonvolatile all-optical switching functionality in the chalcogenide case.9 In the context of nonlinearity, metamaterial and metasurface nanostructures have been employed to resonantly amplify the response of conventional intrinsically nonlinear media via local field enhancement effects and/or arrangements of constituent elements that enhance efficiency (esp. phase-matching for harmonic generation).10 But they also provide mechanisms, via reversible mechanically-, thermally-, electrically-, magnetically-, and optically-induced changes in the composition and/or configuration of unit cells, which can deliver effective optical nonlinearities orders of magnitude larger than those found in bulk media.11 Here, we harness an optically induced interfacial phase transition in the plasmonic metal framework of a planar metamaterial—the gallium backplane of a metamaterial “perfect absorber”—to drive reversible changes in its spectral response and thereby to provide a large, resonantly enhanced, effective optical nonlinearity.

Gallium is a remarkably polymorphic element, existing in up to nine different phases with properties ranging from those of the liquid, which is a highly reflective, near-ideal free-electron metal at optical frequencies,12,13 to those of the $\alpha$ phase—the stable bulk solid form, which is less “metallic” in character (less reflective, more absorbing) as a consequence of the low free-electron density in its partially covalently bound orthorhomic crystal structure.14,15 An unusually large change in optical properties is thus associated with solid-liquid transitions in gallium ($|\epsilon_i - \epsilon_s| \approx 90$ at a wavelength of 1 $\mu$m, $\epsilon_i$ and $\epsilon_s$, being, respectively, the complex relative permittivities of liquid and polycrystalline solid gallium), which has a bulk melting point $T_m$ of 29.8 °C. Gallium also exhibits strong surface melting behavior, whereby at an interface between the solid $\alpha$-phase and a dielectric, surface energy considerations dictate the formation of a few-nm thick liquid layer even at temperatures several degrees below $T_m$.16 The thickness of this interfacial layer, and thereby the optical properties of the metal/dielectric interface are highly sensitive to both temperature and incident light intensity—in the latter case via both thermal and non-thermal transition mechanisms.17,18 These characteristics have seen gallium previously exploited in nonlinear mirrors for laser cavity Q-switching19 (cf. saturable absorbers), and for all-optical and “active plasmonic” signal modulation,20–22 including in the form of a gallium/aluminum metamaterial composite formed by grain boundary penetration.21 Indeed, the term “active plasmonic” itself was coined to describe functionality based upon gallium surface metallization.23

Gallium’s surface-mediated phase-change nonlinearity can readily be harnessed and resonantly enhanced in a photonic metamaterial “perfect absorber.” Such structures, realized over recent years at progressively higher frequencies from the microwave24 to near-infrared25,26 and optical domains,27 generically comprise a planar array of subwavelength plasmonic metal resonators and a continuous metallic (mirror) backplane, separated by a thin dielectric spacer—the resonant absorption frequency being set by the relative permittivities of the constituent media, the geometry of the nanostructured metal layer, and thickness of the spacer.28 Incorporating a gallium backplane mirror, as illustrated in Fig. 1(a), provides a mechanism for dynamically controlling the resonant response of the plasmonic metasurface with light, and delivers an order of magnitude enhancement of the metal’s optical nonlinearity.

Experimental samples consist of a gold nano-disc array and silicon nitride spacer layer over the elemental gallium backplane. Gold disc arrays (Fig. 1(b)) (typically covering a 25 $\mu$m x 25 $\mu$m area; ~5400 unit cells) are fabricated by...
focused ion beam milling in a 30 nm film of gold evaporated onto one side of a 50 nm thick 500 \(\mu\)m x 500 \(\mu\)m silicon nitride membrane suspended in a silicon frame. The opposing side of the membrane is subsequently pressed into contact with a bulk liquid gallium droplet (6N purity), which is then cooled to produce an optically thick solid gallium/silicon nitride backplane mirror. Normal-incidence metasurface optical reflectivity spectra for the solid and liquid phase states of the gallium backplane (at sample temperatures \(T\) of 23.5 and 31.5 °C, respectively) were measured experimentally using a microspectrophotometer, and evaluated computationally in 3D finite element simulations. The numerical model employed material parameters for gold, liquid, and solid gallium from Refs. 29, 30, and 15, respectively, and a non-dispersive refractive index of 2.0 for the silicon nitride backplane/Si\(_3\)N\(_4\)/gold-disc metasurface absorber with a resonant wavelength of \(\sim 1310\) nm. (b) Plan view scanning electron microscope image of a section of the fabricated gold disc array on silicon nitride.

Reversible optical tuning of metasurface absorption underpinned by light-induced interfacial metallization of gallium was characterized using a near-infrared microscope configured, via a single laser input port, for dual-wavelength (1310 nm pump; 1550 nm probe) reflectivity measurements. A metasurface sample was engineered, with dimensions as presented in Fig. 1, to provide strong absorption at the 1310 nm pump wavelength and good reflectivity contrast between gallium phase states \((C = (R_{\text{max}} - R_{\text{min}})/R_{\text{min}})\) at 1550 nm, and mounted in a low pressure thermostatic stage (with sample temperature \(T\) being calibrated against the bulk melting point of gallium \(T_m = 29.8\) °C). The two input beams, originating from single-mode fibre-coupled diode lasers, were focused to concentric spots on the sample with Gaussian intensity FWHM dimensions of 15.7 \(\mu\)m for the 1310 nm pump and 6.6 \(\mu\)m for the 1550 nm probe. The probe was maintained at a fixed CW intensity of 1.0 \(\mu\)W \(\mu\)m\(^{-2}\), while the pump was modulated at 500 Hz with 25% duty cycle (rise/fall times <1 \(\mu\)s) at peak intensities up to 16.8 \(\mu\)W \(\mu\)m\(^{-2}\). For a selection of fixed pump peak intensities, the time dynamics of the metasurface’s non-linear 1550 nm reflective response were recorded while ramping the sample temperature, at a rate of 0.5 °C min\(^{-1}\), from 0 to 32 °C (Fig. 3(a)). Under pump illumination the nanoscale layer of metallic gallium at the metal’s interface with silicon nitride grows to
FIG. 3. (a) Absolute 1550 nm reflectivity of the gallium metasurface as a function of time during and after excitation with a 500 μs, 9.5 μW μm\(^{-2}\) pump pulse at 1310 nm, for a selection of sample temperatures (as labelled) approaching the metal’s bulk melting point (pump modulation frequency 500 Hz; traces averaged over 32 cycles). (b) Maximum induced 1550 nm reflectivity change for a selection of 1310 nm pump intensities (as labelled) as a function of temperature. The inset shows reflectivity relaxation time as a function of temperature and pump intensity.

against the conductive removal of heat from the skin layer predominantly into the gallium bulk. At higher intensities and temperatures close to \(T_m\), the surface metallization observable as a change in 1550 nm reflectivity proceeds more rapidly and the reflectivity level saturates within the pump pulse duration.

Relaxation time, defined as the interval between withdrawal of the pump excitation and recovery of the photoinduced reflectivity change to below 1/e of its maximum value, as the metallized surface layer of gallium reverts to the \(\alpha\)-phase, increases critically towards \(T_m\) as shown in the inset to Fig. 3(b).

The phase transition-mediated nonlinear dependence of gallium metasurface reflectivity on incident light intensity cannot be quantified in terms of a conventional \(\chi^{(3)}\) nonlinear susceptibility value nor indeed can one readily be approximated on the basis of the induced change in the metal’s relative permittivity alone. However, a meaningful figure of merit \(\gamma\) may be obtained by considering the induced change in reflectivity per unit illumination intensity

\[
\gamma = \frac{dR/R_0}{dT},
\]

where \(R_0\) and \(R\) are the probe wavelength reflectivities at pump illumination intensity levels of zero and \(I\), respectively. \(\gamma\) reaches a peak value of order 120 \(\mu m^2\) mW\(^{-1}\) (a relative reflectivity change of 38% at an incident intensity of 3.8 \(\mu W\) \(\mu m^{-2}\)) for the gallium metasurface at a temperature of 28.8°C, one degree below \(T_m\). This should be compared with the values of 17 \(\mu m^2\) mW\(^{-1}\) for a simple planar mirror interface between gallium and a 50 nm silicon nitride membrane (evaluated as part of the present study) and a value of 24 \(\mu m^2\) mW\(^{-1}\) for a planar gallium/silica interface at a wavelength of 810 nm (Ref. 17), both at \(T = T_m = 1 = 28.8^\circ\)C.

Photonic metasurfaces with actively controllable, adaptive, and nonlinear spectral response functions offer applications potential in fields ranging from radiation emitters and sensors to spatial light modulators. The present study shows that gallium, as an active plasmonic medium undergoing optically induced surface metallization at low light intensities, provides unique functionality for cross-wavelength continuous tuning and switching of the spectral response of such structures, delivering in the present case a >50% relative change in 1550 nm reflectivity under \(<20 \mu W\) \(\mu m^{-2}\) illumination at 1310 nm. Indeed, the gallium metasurface achieves a nonlinear reflection coefficient, for pump and probe wavelengths selected by design, which is an order of magnitude larger than that of the corresponding unstructured gallium/dielectric interface.

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