Optical Bistability in Nonlinear Surface-Plasmon Polaritonic Crystals

G. A. Wurtz, R. Pollard, and A. V. Zayats*

School of Mathematics and Physics, The Queen's University of Belfast, Belfast, BT7 1NN, United Kingdom

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Nonlinear optical transmission through periodically nanostructured metal films (surface-plasmon polaritonic crystals) has been studied. The surface polaritonic crystals have been coated with a nonlinear polymer. The optical transmission of such nanostructures has been shown to depend on the control-light illumination conditions. The resonant transmission exhibits bistable behavior with the control-light intensity. The bistability is different at different resonant signal wavelengths and for different wavelengths of the control light. The effect is explained by the strong sensitivity of the surface-plasmon mode resonances at the signal wavelength to the surrounding dielectric environment and the electromagnetic field enhancement due to plasmonic excitations at the controlled light wavelengths.

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All-optical signal processing in integrated photonic circuits and its applications in optical communications and computing require the ability to control light with light [1,2]. All-optical devices based on various types of optical nonlinearities have been considered during past years. The main drawbacks of the majority of such devices are the limitation on their minimum size needed to provide a light pass sufficient to achieve a sizeable nonlinear response, and relatively high operational light intensities. Very recently, nonlinear optical devices based on photonic crystal defects have been proposed that utilize the electromagnetic field confinement and enhancement at the photonic defect location to enhance nonlinear effects [2–5].

A significant field enhancement can also be obtained in plasmonic devices as has been demonstrated in experiments on surface enhanced Raman scattering, second-harmonic generation, and photon tunneling [6]. This enhancement is the result of the increased density of electromagnetic states near a metal surface in the frequency range of surface-plasmon excitations. Coupling a control light to such surface modes can be used to enhance the effective nonlinearity of the system if a nonlinear material is placed near the metal surface [7].

Concurrently, if the surface plasmons in the form of surface-plasmon polaritons (SPPs), propagating surface waves, are chosen to be a signal carrier, their high sensitivity to surface conditions could bring a viable solution to the problem of their all-optical control [6,8]. In this scheme, minute changes induced in the refractive index of the nonlinear material placed on the metal surface would significantly influence the surface-plasmon resonant conditions and surface polariton propagation along the surface.

To combine the above described two requirements in one all-optical device, we have used a periodically nanostructured metal film that converts incident signal and control photons into plasmonic excitations. Such a SPP crystal [6,9,10] behaves as a two-dimensional photonic crystal for surface polaritons and its nonlinear response can be achieved via hybridization with a nonlinear environment. This configuration is simultaneously taking advantage of both the field enhancement at the control-light frequency and the surface sensitivity of SPPs at the signal light frequency to the permittivity changes induced by the control light.

The first experiments on all-optical control of the transmission of SPP crystals have been performed using a free-standing metal film perforated with cylindrical channels hosting a nonlinear polymer [11]. In these experiments, the transmission of the signal light, coupled to surface polaritons on the metal-polymer interfaces, was modulated by the on-off switching of the control light which was coupled to cylindrical surface plasmons in the channels. This coupling provides optical switching at very low controlled light intensities. Even lower light intensities, on the level of single photons, were required to induce nonlinear effects when light was coupled to localized surface plasmons supported by nanopores in a metal film [12,13]. Theories of nonlinear effects in one-dimensional SPP crystals were developed considering the excitation of waveguided modes in subwavelength slits in the metal film filled with nonlinear polymer [14] or the nonlinear behavior induced by SPP at the metal-dielectric interfaces [15].

In this Letter we demonstrate that the transmission of a nonlinear surface polaritonic crystal exhibits bistable behavior that depends on both control-light intensity and wavelength. We study the spectral dependencies of this bistability, the nonlinear transfer function, and discuss its physical origin.

The SPP crystals used in the experiments were fabricated in a 220 nm thick gold film sputtered onto a glass substrate using rf-magnetron sputtering. A square array of circular holes (hole diameter 200 nm, period 600 nm, array size 12×12 μm^2) were milled in the metal film using a focused ion beam microscope [Fig. 1(a)]. The nanostructures were covered with a nonlinear polymer (poly-3BCMU: poly-3-butoxy-carbonyl-methyl-urethane polydiacetylene) obtained by spin coating the monomer solution in chloroform on the surface of the nanostructure. The hybridized nanostructure was then exposed to UV radiation for photopolymerization. The thickness of the polymer

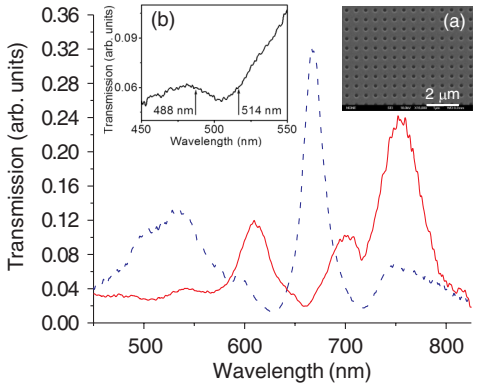

FIG. 1 (color online). Normal incidence transmission spectra of the SPP crystal (dashed line) before and (solid line) after deposition of the nonlinear polymer. (a) Scanning electron microscope image of the SPP crystal. (b) 45° transmission spectrum in the spectral range of the control-light wavelengths (488 nm and 514 nm wavelengths are indicated by the arrows).

film was estimated to be in the 200–250 nm range on the basis of calibration measurements performed with atomic force microscopy and absorption spectroscopy. The polymer film thickness is of the same order of magnitude as the SPP field extension in the spectral range under consideration.

The transmission spectra of the SPP crystal were obtained at normal incidence using a collimated light from a white-light tungsten-halogen source linearly polarized in the direction of one of the main axes of the SPP crystal. The zero-order transmitted light of the same polarization state was then focused onto a fiber-coupled liquid-nitrogen-cooled CCD spectrometer. The control light was generated by an Ar-ion laser and slightly focused on the structure at an angle of incidence of about 45°. The linear polarization of the control light (488 nm or 514 nm wavelength) was set perpendicular to the polarization of the signal light. Both white-light signal beam and control light illuminated the crystal from the substrate side.

The transmission spectrum of the bare (uncoated) structure (Fig. 1) consists of a set of peaks corresponding to resonances in the spectrum of the electromagnetic modes of the SPP crystal [8,9,15,16]. These modes can be related to surface-plasmon excitations localized at the holes in the metal film, surface-plasmon polariton Bloch modes on the metal film interfaces, and to the coupling between these two kinds of modes. As a consequence of the polymer coating, both SPP Bloch modes and localized plasmon modes of the holes are modified due to the changed dielectric constant of the hole filling and interface-adjacent medium. This results in the changes in the transmission spectrum (Fig. 1).

Taking advantage of the nonlinear response of the polymer, the control illumination is used to modify the density of states of the nonlinear SPP crystal and induce the intensity-dependent changes in its optical response at the signal light wavelength. These changes are monitored in the zero-order transmitted light as a function of wavelength using the white-light signal beam. Under the control-light illumination ($\lambda = 488$ nm, $P = 60$ mW), the transmission of the nonlinear SPP crystal is different [Fig. 2(a)]. The observed transmission changes are up to 60% of the initial transmittance of the SPP crystal in some transmission resonances [Fig. 2(b)]. The nonlinear changes in transmittance are observed at various wavelengths of the control light and demonstrate a similar behavior for different peaks exhibiting suppression (negative differential transmittance) or enhancement (positive differential transmittance) of the transmission. In addition to the changes in the transmission magnitude, the position of minima and maxima in the transmission spectra are also affected by the control light. This is an indication of the modification of the surface-plasmon mode spectrum via the control light induced changes in the polymer dielectric constant.

In the control experiments performed under the identical conditions but using bare SPP crystals and homogeneous polymer films deposited onto smooth glass or metal surfaces, no optical nonlinearity has been observed. Nonlinear transmission was also absent in the hybritized SPP crystals with different metallic nanostructure parameters at the same control-light wavelengths. Under very high control-light intensities, the transmission in all these cases exhibits irreversible changes related to polymer damage.

For low intensity signal light, and neglecting the nonlinear response of the metal, we can consider the variations of the dielectric constant of the nonlinear material ε as solely induced by the control light of frequency ω_c:

$$\varepsilon(\vec{r}, \omega) = \varepsilon^{(0)}(\omega) + 4\pi\chi^{(3)}|E_L(\varepsilon(\vec{r}, \omega), \omega_c, \vec{r})|^2, \quad (1)$$

where $\varepsilon^{(0)}$ and $\chi^{(3)}$ are the linear dielectric constant and

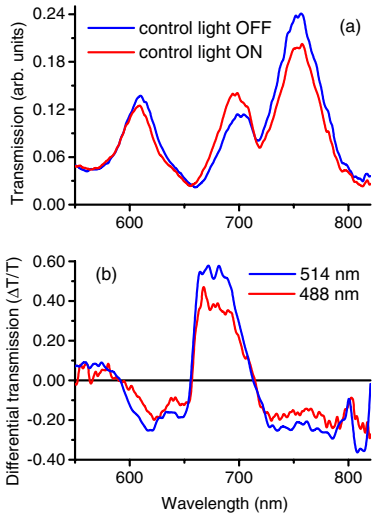

FIG. 2 (color online). (a) Normal incidence transmission spectra of the nonlinear SPP crystal measured with the control light ($\lambda = 488$ nm, $P = 60$ mW) "on" and "off." (b) The induced differential transmittance spectra for the same intensity of the 488 nm and 514 nm control light as in (a).

third-order nonlinear susceptibility of the polymer, respectively, and $E_L(\varepsilon(\vec{r}), \omega_c, \vec{r})$ is the local, position dependent $[\vec{r} = (x, y, z)]$ electric field of the control light, which is determined by the SPP crystal parameters. Thus, the field distribution described by $E_L(\varepsilon(\vec{r}), \omega_c, \vec{r})$ depends on the induced permittivity changes in the nonlinear SPP crystal. As a consequence of this self-consistent process, the field distribution for a given control intensity and wavelength corresponds to a unique spatial distribution of $\varepsilon(\vec{r})$. The signal light then interacts with the SPP crystal which eigenmodes, and, therefore, optical properties are determined by the distribution of the dielectric constant around the nanostructured system [16]. If the control intensity is changed, the changes in the induced permittivity of the polymer result in the modifications of the spatial distribution of $\varepsilon(\vec{r})$ in addition to its magnitude and, therefore, a different optical response of the SPP crystal. This provides a mechanism for bistable behavior of the optical transmission with the intensity of the control light. The nonlinear transmission and its bistability are determined by both the value and spatial variations of $\varepsilon(\vec{r})$, both being responsible for the SPP crystal eigenmodes. Thus, as in a typical configuration for optical bistability, the above described process requires a nonlinear transmission dependence on the control light to achieve a transistor-type effect [1,2] as well as a "built-in" feedback mechanism to enable bistability [17,18].

For the control wavelength of 488 nm, and in the range of the intensities where only reversible changes of the transmission occur (below 5 kW/cm^2), the bistability in the SPP crystal transmission has been observed for all main resonances of the transmission spectrum (Fig. 3). However, the behavior of the hysteresis loops with control intensity is different at different probe wavelengths: both increased and suppressed transmission is observed. The complete switching occurs at similar control intensities for the transmission resonances considered, and the transmission returns to its original values when the control light is off.

The analysis of the band-gap structure and SPP Bloch modes of the nonlinear SPP crystal shows that the different behavior of the bistability loops observed at different wavelengths is related to the origin of the transmission resonances at these wavelengths. The peaks at around 750 nm and 620 nm exhibit a similar bistable behavior and correspond to the excitation of SPP Bloch modes ($\Gamma - M$ and $\Gamma - X$ directions, respectively) on the polymer-metal interface. For these wavelengths the influence of the nonlinearity is chiefly a modification of the absolute value of the transmittance at the resonance. The behavior of the peak at around 700 nm is more complicated: with an increased control intensity, not only does the transmittance change, but the peak shape changes. This highlights the complex origin of this peak corresponding to the coupling of localized surface-plasmon resonances from the holes with SPP Bloch modes at the metal-polymer interface.

At the control wavelength of 514 nm [Fig. 3(d)–3(f)], the nonlinear transmission is also observed for all resonances. Compared to the experiments with the 488 nm control wavelength, the observed relative transmission changes in this case are up to 50% stronger at some signal wavelengths. However, the bistable effect is strongly present in the 690 nm resonance [the bistability is in this case observed at higher control intensities with an unusual "mini-loop" that may give some indication of two resonances involved [1]]; in the other resonances the bistability is weaker if any. The observed control wavelength dependence underlines the importance of both nonlinearity and feedback mechanism for the bistability to occur. At the 488 nm control wavelength, the nonlinear changes are smaller than at 514 nm but the feedback is stronger leading to bistability in the nonlinear transmission at all resonances of the SPP crystal. At the same time, in spite of the stronger nonlinear dependence of the transmission, bistability is not always present under the 514 nm control illumination due to a weaker feedback.

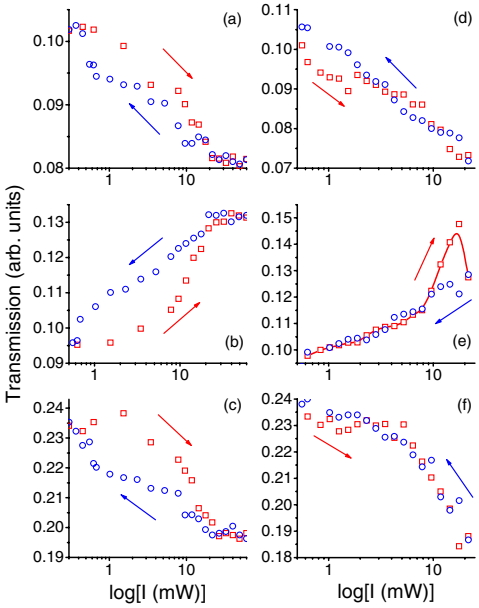

FIG. 3 (color online). Transmission dependence of the nonlinear SPP crystal at (a), (d) 620 nm, (b), (e) 690 nm, and (c), (f) 750 nm wavelengths for the increasing (squares) and decreasing (circles) control-light intensity at (a)–(c) 488 nm and (d)–(f) 514 nm wavelengths.

The dependence of the bistability on the control-light wavelength can be understood taking into account that the SPP field distribution plays a crucial role in triggering bistability as discussed above [Eq. (1)]. In particular, under the 488 nm and 514 nm illumination, different surface-plasmon resonances of the metal-dielectric SPP crystal are excited [Fig. 1(b)]. The analysis suggests that both the near-field intensity and its spatial distribution close to the metal-polymer interface, where nonlinear processes are predominant, are responsible for the differences in the nonlinear properties at the control wavelengths used. These cannot be judged from far-field transmission spectra.

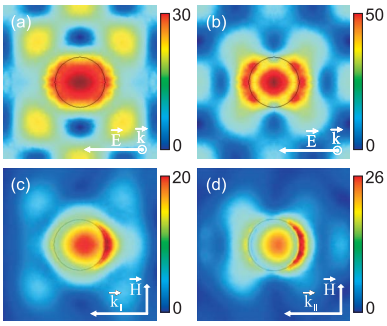

FIG. 4 (color online). The control-light intensity distributions over a unit cell of the nonlinear SPP crystal as in Fig. 1 calculated using finite-element modeling at 10 nm distance above the metal surface on the nonlinear polymer side for (a), (c) $\lambda = 488$ nm and (b), (d) $\lambda = 514$ nm and the angle of incidence of (a), (b) 0° and (c), (d) 45°. $\vec{k}_{\|}$ indicates the projection of the wave vector of the incident TM-polarized light.

We used 3D finite-element calculations to model a system exhibiting a zero-order transmission similar to one measured for the bare SPP crystal. The model predicts that for the SPP crystal hybridized with nonlinear polymer, the electromagnetic field distributions above the metal-polymer interface are different at the 488 nm and 514 nm wavelengths with a higher field between the holes in the case of the 488 nm control-light (Fig. 4). Since the field in the holes is similar for both control-light wavelengths, the SPP Bloch modes can be considered as the dominant origin of the observed bistability. Despite the spectral position of the 488 nm control light above the surface-plasmon resonance of the polymer-covered Au surface, it can excite the SPP modes if the polymer layer is thin enough [19]. In the case of the 488 nm illumination, although the far-field transmission is smaller than at 514 nm, more energy is coupled to the SPP modes at the metal-polymer interface. The changes in this coupling with the incident light intensity at the two control wavelengths may explain the feedback differences in the two cases. Therefore, the different nonlinear behavior triggered by the light of 488 nm and 514 nm originates from the different spatial distributions and magnitude of the electromagnetic field at the metal-polymer interface.

It is important to note that the thermal contribution to the nonlinear response of the polymer can compete with electronic contributions under cw excitation. The dependence of the nonlinear behavior on the control-light wavelength strongly indicates that thermal effects, which are wavelength independent in the range of the control wavelengths used, cannot explain our observations.

In conclusion, we have shown that the nonlinear optical transmission through hybridized surface-plasmon polaritonic crystals exhibits optical bistability with varying control-light intensity. The effect is explained by the strong sensitivity of surface-plasmon resonances to their surrounding dielectric environment as well as to the electromagnetic field enhancement due to plasmonic excitations at the controlled light wavelength. The nonlinear transmission of SPP crystals has been previously observed by stimulating the resonances of cylindrical surface plasmons for the control-light enhancement [11]. Cylindrical surface plasmon provide stronger field enhancement and therefore lower control-light intensities. The approach based on the SPP Bloch modes described in this Letter allows generally lower field enhancement than cylindrical surface plasmons but provides more flexibility for optimizing the spectral response of the structure and higher signal throughput. It should be noted that the bistability observed in transmission through SPP crystals is a consequence of the bistability of SPP Bloch modes and thus SPP excitations propagating on the metallic structure. Therefore, nonlinear (including bistable) behavior of SPP waves guided through the SPP crystal should also be expected.

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*Email address: a.zayats@qub.ac.uk

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