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Active control of surface plasmon–polariton waves

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Abstract
We outline a new concept for active plasmonics that exploits light-induced nanoscale structural transformations in the waveguide material. The concept is illustrated by numerical modelling and test experiments on a gallium–dielectric interface. We also discuss other possible implementations of the concept such as an electro-plasmon modulator, a plasmon detector and a switch that controls one plasmon wave with another.

Keywords: surface plasmon–polaritons, optical switching, gallium, structural transformation

1. Introduction

Propagating plasmon–polariton excitations in nanostructured metal films are clearly emerging as a new information carriers for highly integrated photonic devices for signal and information processing, with structural elements smaller than the wavelength, which enables strong guidance and manipulation of light. The functionality of these devices depends on electromagnetic excitations coupled with electrons at a metal–dielectric interface [1–3].

Ranges of very promising nanostructures that guide surface plasmon–polariton (SPP) waves are currently being investigated [4–7]. SPPs in gold films can propagate for tens of microns and may be guided by structuring the metal film and creating polaritonic bandgap materials [8]. These developments emphasize the need to find efficient techniques for active manipulation and detection of surface polariton waves.

We recently proposed a new concept for active nanoscale functional elements operating with SPP signals [9], that we review and further develop here. In this paper we also report on first experiential demonstrations of the new concept at work [10]. Our approach takes advantage of one of the most characteristic features of SPPs, namely that their propagation depends strongly on the metal's properties in a thin layer near the surface. This characteristic is exploited by manipulating the dielectric properties of the metal film using light-induced structural phase transformation, an idea which has already been shown to offer all-optical switching at milliwatt power levels in thin flat [11] and nanoparticle films [12], and has recently enabled the demonstration of an all-optical nano-aperture gate operating at only a few nanowatts of optical power using a single gallium nanoparticle [13].

Here we show that SPP signals in metallic on dielectric waveguides containing a gallium section can be effectively controlled by switching the crystalline gallium section of the waveguide from one structural phase to another. The switching may be achieved both by changing the waveguide temperature and by external optical excitation. Gallium is a uniquely suitable material to realize this concept. It is known for its polymorphism [14, 15]. In α-gallium, the stable 'ground-state' phase [16, 17], molecular and metallic properties coexist—some inter-atomic bonds are strong covalent bonds, forming well defined Ga_2 dimers (molecules), and the rest are metallic bonds. This so-called boron-like structure is what some authors call an inorganic polymer. The structure is highly anisotropic, with much better thermal and electrical conductivity in the 'metallic planes' than along the covalent bonds. Remarkably, α-gallium has a very low melting point, only 29.8 °C. The covalent bonding leads to energy gaps at the Fermi level, and gives rise to a strong optical absorption peak centred at 2.3 eV and spreading from approximately 0.68 eV (∼1820 nm) to about 4 eV (∼310 nm). The pronounced difference between the electronic structure of α-Ga and the more metallic phases, which are metastable under normal conditions, manifests itself as a marked difference in the optical properties. The properties of the metastable phases are similar to those of the highly metallic liquid phase.

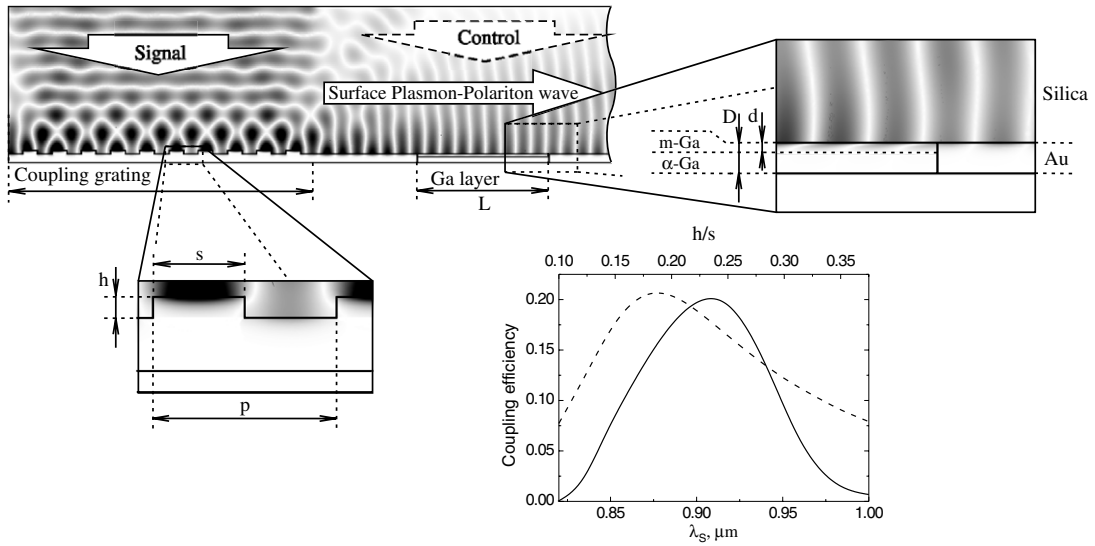

Figure 1. An SPP gold-on-silica waveguide containing a gallium switching section. The metallic film is on the bottom surface of the silica substrate. Field mapping shows the magnitude of the z component of the magnetic field. In the graph, the solid curve shows the dependence of the incident-light-to-SPP-wave coupling efficiency on the wavelength of the incident light in silica (λ_S) for a grating with $p = 2s = 0.9$ μm, $h/s = 1/5$. A peak at the in-silica wavelength $\lambda_S = 0.91$ μm corresponds to a vacuum wavelength $\lambda = 1.31$ μm. The dashed curve represents the dependence of the input coupling efficiency on the ratio h/s for a grating with $p = 2s$ at $\lambda_S = 0.91$ μm.

In terms of the dielectric coefficients at a wavelength of 1.55 μm, $|\varepsilon_{\text{liquid}}|/|\varepsilon_\alpha| \sim 7$. To achieve switching and control functions one need to achieve a controllable and reversible transition from the α-phase to a metastable metallic phase or the melt. The energy required to achieve melting is relatively small, about 60 meV/atom. High quality α-gallium interfaces with silica may be achieved using various techniques, from squeezing molten gallium to ultra-fast pulsed laser deposition [18].

2. Modelling the switching characteristics of an active plasmonic device

To evaluate the potential switching characteristics of the active SPP waveguide we numerically modelled it using the finite element method. We investigated a gold film waveguide containing a gallium section of length L on a silica substrate (figure 1). To couple and decouple optical radiation to and from the waveguide, ten-element gratings were placed at either end of the structure (the decoupling grating is not shown in the figure). In such a waveguide the SPP wave propagates at the interface between the metal film and silica substrate, through the gold and gallium sections. SPP decay length in a continuous gold/silica waveguide is 53 μm for an excitation wavelength of 1.31 μm. In the waveguide containing a Ga section, the transmitted SPP wave is attenuated due to the mismatch of dielectric characteristics at the Au–Ga boarders and losses that depend on the structural phase of gallium.

External stimulation may be used to convert the gallium section of the waveguide from the ground α-phase to the metallic liquid-like phase. This transformation takes the form of an interface metallization: a thin layer of metallic gallium of thickness d develops at the silica interface (see figure 1). As α-gallium is a highly anisotropic material (crystal class mmm), we performed calculations of waveguide transmission (disregarding coupling–decoupling losses) for each of the

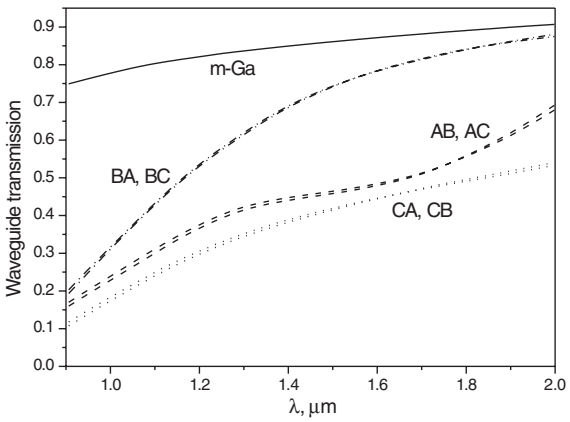

Figure 2. Waveguide transmission as a function of wavelength for different phases and crystalline orientations of the gallium film and the isotropic metallic phase.

main crystalline orientations of the Ga film at the interface, in the range of excitation wavelengths from 0.9 to 2.0 μm. Figure 2 shows the results of these calculations for a waveguide containing a gallium section of length $L = 2.5$ μm, assuming that it was a homogenous crystal of a given orientation or is a entirely molten isotropic metallic phase. The following notation is used: curve AB corresponds to a gallium crystalline structure with the A-axis parallel to the direction of SPP propagation and the B-axis perpendicular to the plane of the interface. Similar notation applies to curves BA, BC, CA and CB. The transmission efficiency was obtained by integrating the power flow over the region above the decoupling grating with the Ga strip present and dividing it by integrated power flow over this region without the Ga strip. The values of the complex dielectric coefficients of α-Ga, m-Ga and gold were taken from [19–21].

Our calculations revealed that the waveguide transmission strongly depends on the structural phase of the gallium section

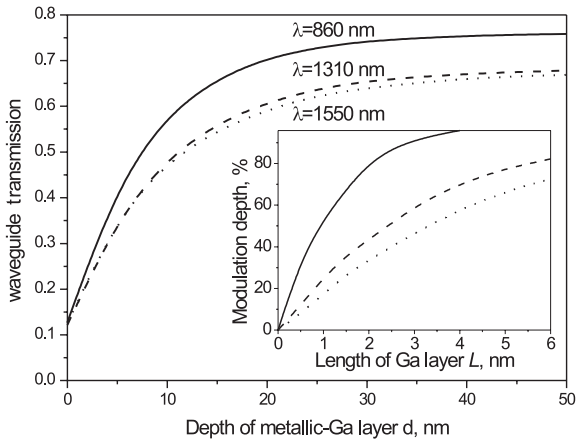

Figure 3. Waveguide transmission as a function of the depth d of the metallic gallium layer. Length L of the Ga section: $\lambda = 860$ nm—$L = 2.2$ μm, $\lambda = 1310$ nm—$L = 5.8$ μm, $\lambda = 1550$ nm—$L = 9$ μm. The inset shows the dependence of modulation depth on the length L of the gallium section (assuming d equals the full thickness of the Ga layer D).

and that a change in the phase can be used to actively controlling the transmission. As the structural transformation in gallium is a surface mediated effect, two phases of gallium may co-exist at the interface, with a thin layer of the metallic phase sandwiched between silica and α-gallium. This gives the possibility of continuous, 'analogue' control of the waveguide transmission. To analyse this functionality we calculated the waveguide transmission for different thicknesses of metallic gallium, up to $d = 60$ nm for a number of incident wavelengths (see figure 3). For illustrative purposes the gallium strip was taken to be polycrystalline α-gallium (isotropic with dielectric constants averaged over crystal directions). In can be seen that the presence of a metallic Ga layer just $d = 30$ nm deep can dramatically change the transmission of plasmons through the waveguide.

3. Optical control functionality

The switching of structural phases in gallium may be achieved by external optical excitation, which is represented in figure 1 by a dashed arrow. This switching may be reached through simple heating of the gallium layer with light. However, in addition, gallium presents another mechanism of obtaining the metallic phase, namely a non-thermal transition [22, 23]. Due to localization of photo-generated electron–hole pairs on the dimers light absorption can excite a dimer from the bonding to the antibonding state, reducing the stability of the surrounding crystalline cell. The α-gallium cell subsequently undergoes a transition to a new configuration without necessarily achieving the melting temperature. Such a transition can be observed if the gallium interface is illuminated with light of a wavelength within the dimers' absorption line. This non-thermal mechanism is important for quasi-cw excitation of gallium interfaces at intensities up to a few kW cm^{-2}. However, with shorter optical pulses thermal diffusion does not have enough time to remove heat from the skin layer during the pulse, and the temperature at the excitation point increases, inevitably leading to thermal melting of the gallium film. Whatever the mechanism of phase transition is, it is a surface mediated effect and develops as a propagation of the new phase from the surface into the bulk of the crystal. The phase transition process is highly reproducible as it only involves a few atomic layers of gallium at the interface.

To evaluate the possibility of optical control of SPP propagation with light we performed reflective pump–probe experiments on an α-Ga interface under resonant conditions for SPP excitation by the probe laser (where reflection of the probe light is low) [10]. When the pump (control) laser initiates a structural transformation in the gallium it drives the system away from resonance. Since the efficiency of SPP coupling depends strongly on the dielectric parameters of the material, such excitation with the pump leads to an increase in probe reflectivity.

As the dispersion relation for SPPs is different from that for light, it is only possible to couple light into an SPP wave on a smooth surface by using a matching device such as the grating described in the previous section. For the purpose of matching in our experiments we used an attenuated total reflection coupling scheme known as the Otto configuration [1]. In our implementation of the Otto scheme gallium is interfaced with a BK7 glass prism covered with a MgF_2 film of thickness $D = 185$ nm (figure 4(a)). A gallium film was prepared on the prism by squeezing a bead of the liquid metal then solidifying it. The probe light wave from a p-polarized laser diode ($\lambda_p = 780$ nm) is then efficiently (resonantly) coupled to an SPP wave at the MgF_2/α-Ga interface at an incident angle $\beta = 66°$. The photon wavevector in glass in this case is equal to the SPP wavevector on the MgF_2/α-Ga interface. To initiate the structural transformation from the α-phase to the 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 $\lambda_c = 1064$ nm with a repetition rate of 20 Hz (figure 4(a)).

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 (figure 4(b), inset). When the excitation is terminated the molten layer rapidly recrystallizes. It returns to the structural configuration and crystalline orientation that it was in before melting due to the presence of the solid gallium crystal behind the metallized layer. The surface mediated phase transition is therefore truly reversible. The magnitude of the effect increases with pump fluence up to about 15 mJ cm^{-2} and then saturates (figure 4(b), solid curve). The dashed curve in the figure represents the dependence of the reflectivity of the structure on the depth d of the metallized layer. From these data we can estimate that a fluence $Q = 15$ mJ cm^{-2} corresponds to a depth $d \approx 30$ nm which is sufficient for about 80% modulation of the SPP wave in the scheme presented on figure 1. This means that for a 2.5 μm $\times$ 2.5 μm section of gallium waveguide the optical energy required for high-contrast switching is of the order of only 1 nJ.

We investigated the temperature behaviour of the reflectivity modulation at a constant fluence of $Q = 15$ mJ cm^{-2}. The reflectivity modulation changes slowly with decreasing temperature. The relaxation time depends more strongly on temperature and may be as small as a few tens of nanoseconds (figure 5). The decrease of the relaxation time can be explained by the fact that the recrystallization velocity v depends on the temperature: $v \propto (T - T_0)$, where

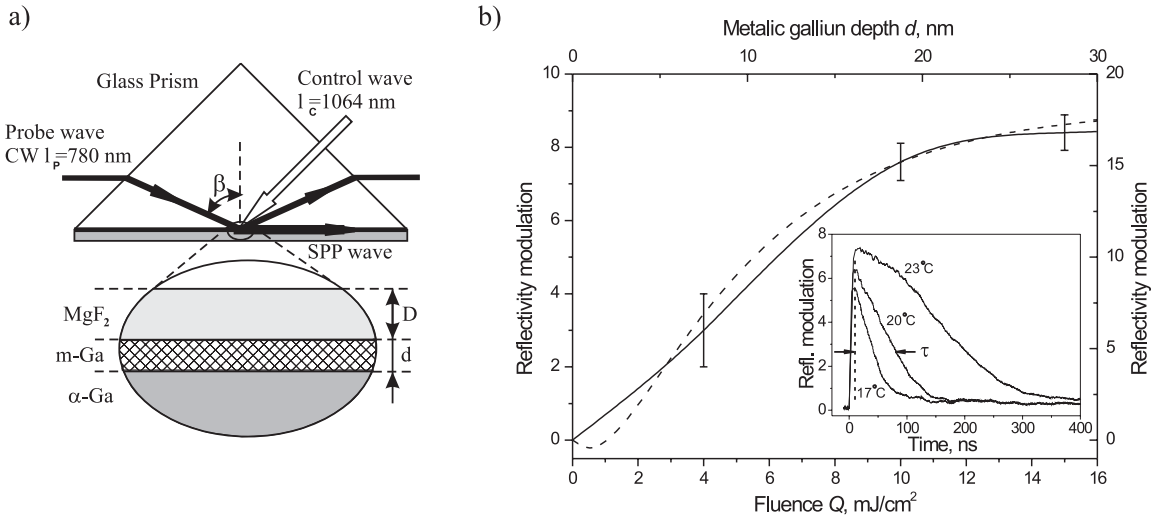

Figure 4. (a) Arrangements for the generation of a metallic gallium layer and the monitoring of its depth. (b) Dependence of maximum reflectivity modulation of the structure on the fluence of the pump light at $T = 28\,^{\circ}\mathrm{C}$ (solid curve), and the theoretical dependence of the reflectivity on the thickness d of the metallic m-phase of gallium (dashed curve). The inset presents the reflectivity modulation of the structure following 6 ns impulse excitation at a wavelength of 1.06 μm ($Q = 15$ mW cm^{-2}) for various structure temperatures.

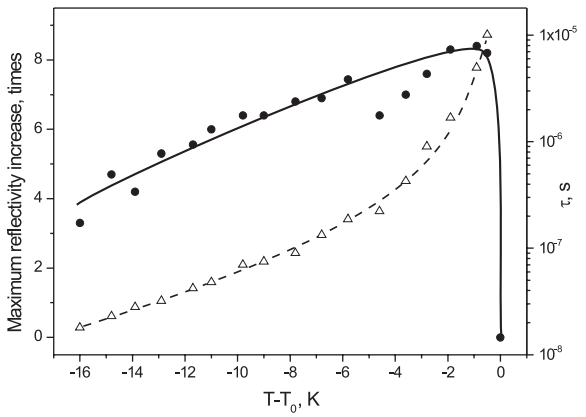

Figure 5. Temperature dependence of the maximum reflectivity increase (•) and corresponding relaxation time (△) at $Q = 15$ mJ cm^{-2}.

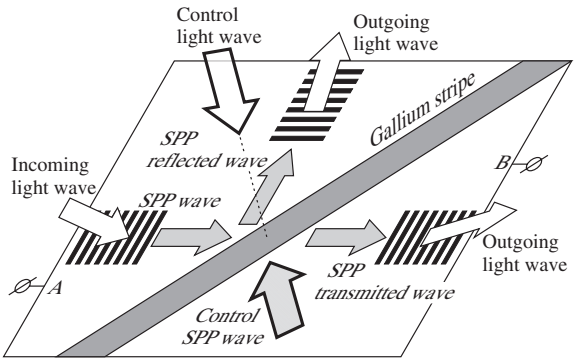

Figure 6. Generic arrangement for light wave, intense SPP wave and electric current control of SPP transmission through and reflection from a gallium stripe. The electrical arrangement can be used for detection of SPP waves.

$T_0 = 29.6\,^{\circ}\mathrm{C}$ is the melting temperature of gallium [24], so the further the system is from the melting temperature, the shorter the time required for the metastable metallized layer to re-crystalize back to the α-phase. The overall bandwidth of the registration system was 125 MHz, so the transient 'switch-on' time was not resolved in this experiment. It may have been as short as 4 ps, which is the intrinsic transition time for a transformation from the α-phase to the metallic phase [18]. It follows from previous studies of optically induced metallization of gallium [11] that the optical control mechanism is inherently optically broadband.

4. Concept applications

The proposed active plasmonic concept may be used far beyond the scheme outlined in figure 1. The control function of the intense light wave may be achieved by a beam of electrons instead [25]. One should also be able to manipulate the reflected SPP wave as well as the transmitted wave (see figure 6). The direction of the reflected and transmitted waves will have to obey a momentum conservation law similar to the light reflection law at the interface of two dielectrics. Here the wave intensities would be controlled by the dielectric properties of the interfacing metals, thus making the reflected wave intensity a function of the gallium structural phase.

Instead of an external optical stimulation, an electric current producing Joule heating in the gallium section of the waveguide could be used to induce a structural transformation in the film. For this purpose the gold sections of the structure adjacent to the gallium section can be used as electric terminals (A and B in figure 6), providing a configuration for an 'electro-plasmon–polariton' modulator for both transmitted and reflected SPP waves (this device is analogous to a conventional 'electro-optical modulator').

Recently it was observed that a photoconductive effect results from a light-induced structural phase transition in a surface layer of gallium: the light-induced structural phase change is accompanied by a change in the conductivity of the film, thus allowing for a new mechanism of light detection [26]. We believe that a similar principle can be used to detect SPP waves: energy dissipated by the SSP wave in the gallium layer will change its structural phase and therefore its conductivity,

which may be detected as a change in the resistance between terminals A and B.

Although nonlinear interactions between SPP waves have not yet been demonstrated, we believe that the structural phase transition in gallium may be induced by the energy dissipated by a strong SPP wave, making possible an all-plasmonic device. In this case two SPP waves interact on the gallium section of the waveguide: a 'control' SPP wave directed towards the Ga section of the waveguide switches the phase of the gallium film in the area where the 'signal' passes, thus modulating it (see figure 6). We also expect to see various self-action effects, such as self-focusing and de-focusing and self-phase modulations in a single intense SPP wave propagating through a gallium section. We also anticipate an SPP analogue of optical bleaching with increasing intensity of the wave propagating through the gallium section of the film, as a gallium layer of increasing thickness is converted into the metallic phase, making propagation less lossy.

5. Conclusions

In this work we have outlined a new concept for active plasmonics that exploits nanoscale structural transformations in the SPP waveguide materials. We show that SPP signals in a metal-on-dielectric waveguide containing a gallium section a few microns long can be effectively controlled by switching the structural phase of gallium. The switching may be achieved by changing the waveguide temperature or by external optical excitation.

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References

[1] Raether H 1988 *Surface Plasmons on Smooth and Rough Surfaces and Gratings* (Berlin: Springer)
[2] Barnes W L, Dereux A and Ebbesen T W 2003 *Nature* **424** 824
[3] Zayats A V and Smolyaninov I I 2003 *J. Opt. A: Pure Appl. Opt.* **5** S16
[4] Nikolajsen T, Leosson K, Salakhutdinov I and Bozhevolnyi S I 2003 *Appl. Phys. Lett.* **82** 668
[5] Krenn J R, Salerno M, Felidj N, Lamprecht B, Schider G, Leither A, Aussenegg F R, Weeber J C, Dereux A and Goudonett J P 2001 *J. Microsc.* **202** 122
[6] Maier S A, Kik P G and Atwater H A 2003 *Phys. Rev.* B **67** 205402
[7] Yatsui T, Kourogi M and Ohtsu M 2001 *Appl. Phys. Lett.* **79** 4583
[8] Volkov V S, Bozhevolnyi S I, Leosson K and Boltasseva A 2003 *J. Microsc.* **210** 324
[9] Krasavin A V and Zheludev N I 2004 *Appl. Phys. Lett.* **84** 1416
[10] Krasavin A V, MacDonald K F, Zayats A V and Zheludev N I 2004 *Preprint* cond-mat/0405448
[11] Zheludev N I 2002 *Contemp. Phys.* **43** 365
[12] MacDonald K F, Fedotov V A and Zheludev N I 2003 *Appl. Phys. Lett.* **82** 1087
[13] Soares B F, MacDonald K F and Zheludev N I 2004 *International Quantum Elecrtonics Conference* (*OSA Technical Digest Series*) (Washington, DC: Optical Society of America) paper IThM5
[14] Defrain A 1977 *J. Chem. Phys.* **7** 851
[15] Bosio L 1977 *J. Chem. Phys.* **68** 1221
[16] Gong X G, Chiarotti G L, Parrinello M and Tosatti E 1991 *Phys. Rev.* B **43** 14277
[17] Zuger O and Dürig U 1992 *Phys. Rev.* B **46** 7319
[18] Rode A, Samoc M, Luther-Davies B, Gamaly E G, MacDonald K F and Zheludev N I 2001 *Opt. Lett.* **36** 441
[19] Kofman R, Cheyssac P and Richard J 1977 *Phys. Rev.* B **16** 5216
[20] Teshev R S and Shebzukhov A A 1988 *Opt. Spectrosc.* **65** 693
[21] Palik E D (ed) 1984 *Handbook of Optical Constants of Solids* (New York: Academic)
[22] MacDonald K F, Fedotov V A, Zheludev N I, Rode A V, Luther-Davies B and Emel'yanov V I 2001 *J. Opt. Soc. Am.* B **18** 331
[23] Albanis V, Dhanjal S, Emelyanov V I, Fedotov V A, MacDonald K F, Petropoulos P, Richardson D J and Zheludev N I 2001 *Phys. Rev.* B **63** 165207
[24] Peteves S D and Abbaschian R 1991 *Metall. Trans.* A **22** 1259
[25] Pochon S, MacDonald K F, Knize R J and Zheludev N I 2004 *Phys. Rev. Lett.* **92** 145702
[26] Fedotov V A, Woodford M, Jean I and Zheludev N I 2002 *Appl. Phys. Lett.* **80** 1297