Coherent control of Floquet-mode dressed plasmon polaritons

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(Received 28 March 2012; published 30 May 2012)

We study the coherent properties of plasmon polaritons optically excited on periodic nanostructures. The gold grains are coupled to a single-mode photonic waveguide which exhibits a dramatically reduced transmission originating from the derived quantum interference. With a nonequilibrium description of Floquet-dressed polaritons we demonstrate the switching of light transmission through the waveguide due to sheer existence of intraband transitions in gold from right above the Fermi level driven by the external laser light.

DOI: 10.1103/PhysRevB.85.195463 PACS number(s): 42.50.Ct, 05.70.Ln, 42.65.Re, 42.79.Ta

I. INTRODUCTION

One of the ultimate challenges of optical technologies is the realization of efficient optical switching and computing devices.[1–3] Therefore, quantum-optical functional elements have received increasing interest over the past years. Among the promising candidates, quantum-well structures exhibiting Kerr nonlinearities, metamaterials, or plasmonic systems[4–6] have shown the potential to be used for ultrafast optical switches,[7–10] and expeditiously progressing ultrashort laser sciences open an avenue to their exploration.[11,12] Besides its immense technological importance, the theoretical description of externally driven nonequilibrium quantum systems is a challenge itself.

II. SETUP

We study a photonic silicon-on-insulator hollow core waveguide (SOI) in contact with gold nanograins exposed to an external field. The external time-periodic field modifies the electronic band structure within the nanograins by generating photo-induced Floquet bands which can be attributed to the Franz-Keldysh effect.[13] All metals exhibit intraband transitions occurring within the conduction band featuring a more or less small absorption rate, whereas the governing processes are the interband transitions. In gold (Au) a significantly different behavior has been found and attributed to the specific electronic structure of closed packed Au, namely, to the high polarizability of the $5d^{10}$ cores.[14] The collective resonance exhibits a large red shift to approximately 2.4 eV which leads to the fact that the corresponding intensity is governed by the interband but its sheer existence results from the occurrence of intraband transitions which can be recognized as steplike structures in the spectrum.[15] These transitions, which result from a non-Fermi-like distribution of states, have been observed in pump probe experiments with Au nanospheres of diameters below 30 nm. Their occurrence has been interpreted as the border from bulklike characteristics to the quantum regime of nanoparticles. We show that the pump laser modifies the single plasmon band which results in the development of Floquet bands. Both frequency and amplitude of the external field allow easy and fast-switching control over the position in energy of these bands and sensitively control the generation of a Fano resonance with the photonic SOI mode. We show that this switching may significantly alter the transmission properties within the SOI by the formation of polaritons, electron photon bound states, at the surface of the nanograins. Consequently, the potential of this quantum-optical functional element for all optical switching based on polaritonics is proven.

III. MODEL AND METHODS

Our setup (Fig. 1) is described by a Fröhlich Hamiltonian for fermion-boson interaction which has to be solved by applying the Keldysh formalism with respect to the nonequilibrium character of the considered processes on the femtosecond time scale. We consider the SOI in contact with nanograins, the nanograin in the external field and finally discuss the nonequilibrium solution of the complete system in terms of electron Keldysh-Green's function and SOI transmission. As our starting point we choose a single-band electronic tight binding model with nearest-neighbor hopping,[16] characterized by the hopping amplitude t, with the dispersion for a cubic lattice $\epsilon_k = 2t\sum_i \cos(k_i a)$, a is the lattice constant and k_i are the components of the wave vector. We assume a SOI supporting a single mode $\hbar\omega_0 = 2.34$ eV. The SOI itself shall be coated and therefore not be exposed to laser radiation. The electrons may couple with strength g weakly to SOI photons with frequency ω_0. Hence the full Hamiltonian reads

$$\begin{aligned} H = &\sum_{k,\sigma} \epsilon_k c^\dagger_{k,\sigma} c_{k,\sigma} c^\dagger_{i,\sigma} c_{j,\sigma} + \hbar\omega_o a^\dagger a \\ &+ g\sum_{k,\sigma} c^\dagger_{k,\sigma} c_{k,\sigma}(a^\dagger + a) - t\sum_{\langle ij\rangle,\sigma} c^\dagger_{i,\sigma} c_{j,\sigma} \\ &+ i\vec{d}\cdot\vec{E}_0\cos(\Omega_L\tau)\sum_{\langle ij\rangle}(c^\dagger_{i,\sigma} c_{j,\sigma} - c^\dagger_{j,\sigma} c_{i,\sigma}). \end{aligned}$$

For a clear description we treat first the setup without the external laser field, than the interaction of laser and band electrons, and finally the full compound of external laser radiation interacting with band electrons and the coupling of the latter by a Fano resonance with the waveguide mode. The Hamiltonian without the external laser field reads

$$H = \sum_{k,\sigma} \epsilon_k c^\dagger_{k,\sigma} c_{k,\sigma} + \hbar\omega_o a^\dagger a \;+ g\sum_{k,\sigma} c^\dagger_{k,\sigma} c_{k,\sigma}(a^\dagger + a). \tag{1}$$

Here, we assume the spatial extension of the Au nanograins to be small (<30 nm) compared to the wavelength of the photonic mode inside the SOI. Therefore, the momentum of

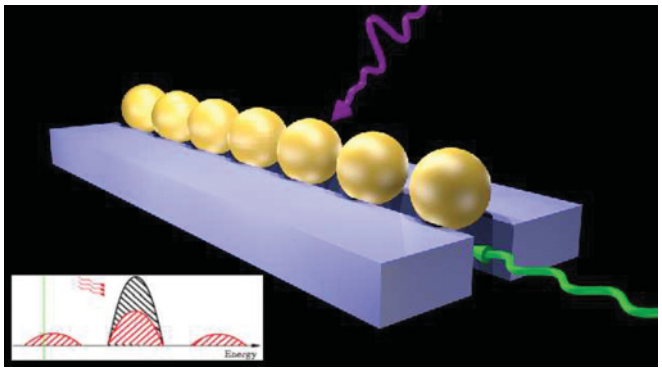

FIG. 1. (Color online) Gold nanograins in contact with a hollow core SOI. Waveguide photons (green) and electrons in the metal form a coupled light-matter state, a polariton controlled by an external laser (pink). Inset: Sketched development of Floquet sidebands.

the photons is much less than the electron's momentum and we can set $q_{\text{photon}} \simeq 0$ whenever we consider the electronic subsystem. Thus, $a^\dagger$ (a) does not carry an index. In Eq. (1), ϵ_k is the electronic band energy, $c^\dagger_{k,\sigma}$ $(c_{k,\sigma})$ creates (annihilates) an electron with momentum k and spin σ. $\hbar\omega_0 a^\dagger a$ is the photon energy eigenstate, where $a^\dagger$ (a) creates (annihilates) a photon inside the SOI with energy $\hbar\omega_0$. The last (coupling) term on the r.h.s. is the standard term resulting from coupling the electronic and the photonic subspaces. Due to the weak interaction between the SOI photons and the electrons in the nanograins we treat this interaction perturbatively. In second order a self-energy contribution as shown in Fig. 2 is obtained from Eq. (1). The coupling of the electronic system with a continuous energy spectrum to the photons with a discrete one leads to a Fano resonance, which is observed in the electronic density of states, as demonstrated in Fig. 3. Here we show the electron's spectral function for various frequencies of the SOI mode for coupling strength $(g/t)^2 = 0.09$ at zero temperature for a spectral width $\tau = 0.005$ of the waveguide mode (measured in units of the hopping t) at half filling, yielding a suppression of the spectral function around the Fermi level (half width τ) where electrons are transferred to the high (low) energy tails of the spectral function. We note that if the energy of the SOI mode $\hbar\omega_0$ is distinctly different from the energy $\hbar\omega$ of the band electrons, the electronic density of states remains unchanged.

The subsystem of a nanograin exposed to a semiclassical electromagnetic laser field is described by the Hamiltonian

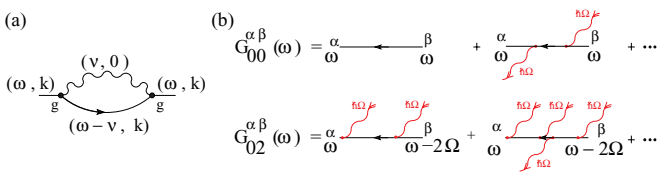

FIG. 2. (Color online) (a): Contribution to the electronic self-energy $\Sigma(\omega,k)$ in second-order perturbation theory. The photon propagator does not carry a momentum as explained in the text. (b): Floquet Green's function in terms of absorption/emission of external energy quanta $\hbar\Omega_L$. $G^{\alpha\beta}_{00}(\omega)$, α,β being Keldysh indices, e.g., represents the sum of all balanced processes. $G^{\alpha\beta}_{02}(\omega)$ describes a net absorption of two photons.

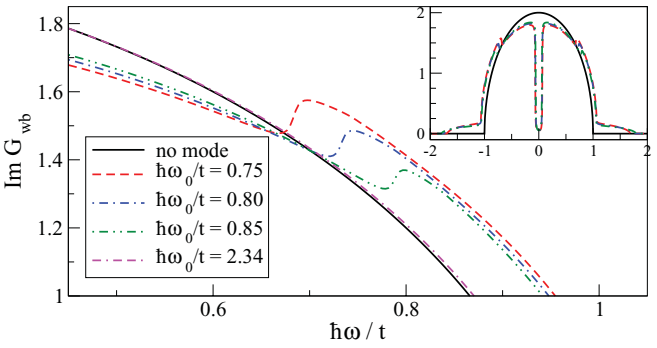

FIG. 3. (Color online) The electron's spectral function for various frequencies of the SOI mode for coupling strength $(g/t)^2 = 0.09$ (about 30% of the coupling A_0/t) at zero temperature for a spectral width $\tau = 0.005$ of the waveguide mode. Shape and position changing of the resonance is clearly visible. If the photonic mode is energetically far off the electronic band, as for $\hbar\omega_0 = 2.34$ eV, the electronic band structure remains practically unchanged. The inset shows the overall behavior of spectral function.

(Lb = laser + band electrons)

$$H_{\text{Lb}} = -t \sum_{\langle ij\rangle,\sigma} c^\dagger_{i,\sigma} c_{j,\sigma} + H_C(\tau), \tag{2}$$

where $\langle ij\rangle$ implies summation over nearest neighbors. $H_C(\tau)$ represents the coupling to the external, time-dependent laser field, described by the electric field $\vec{E} = \vec{E}_0 \cos(\Omega_L \tau)$, via the electronic dipole operator $\hat{d}(\vec{x})$ with strength $\vec{d}$. It is given by

$$H_C(\tau) = i\vec{d}\cdot\vec{E}_0 \cos(\Omega_L\tau) \sum_{\langle ij\rangle} (c^\dagger_{i,\sigma} c_{j,\sigma} - c^\dagger_{j,\sigma} c_{i,\sigma}). \tag{3}$$

The Hamiltonian specifically describes the excitation of a plasmon polariton, which corresponds to spatially delocalized intraband electronic motion caused by an external electromagnetic wave. The accelerating energy is immediately transferred into the motion of electrons by means of single-band nearest-neighbor hopping without interaction between the electrons. Due to the time dependence of the external field, Green's functions truly depend on two separate time arguments. Therefore, we use a double Fourier transform from time- to frequency space introducing relative and center-of-mass frequency

$$\begin{aligned} G^{\alpha\beta}_{mn}(\omega) &= \int \mathrm{d}\tau_1^\alpha \mathrm{d}\tau_2^\beta e^{-i\Omega_L(m\tau_1^\alpha - n\tau_2^\beta)} e^{i\omega(\tau_1^\alpha - \tau_2^\beta)} G\left(\tau_1^\alpha, \tau_2^\beta\right) \\ &\equiv G^{\alpha\beta}(\omega - m\Omega_L, \omega - n\Omega_L), \end{aligned} \tag{4}$$

where (m,n) label the Floquet modes and (α,β) specify on which branch of the Keldysh contour $(\pm)$ the respective time argument resides. In general, a Floquet state is the analog to a Bloch state: The first one results from a time-periodic potential whereas the latter is the result of a space-periodic potential and both induce a band structure. A physical interpretation of such a Keldysh-Floquet Green's function is given in Fig. 2. The special case of noninteracting electrons allows an analytical solution for $G_{mn}(k,\omega)$ by solving the equation of motion. Including photo-induced hopping, the exact retarded Green's

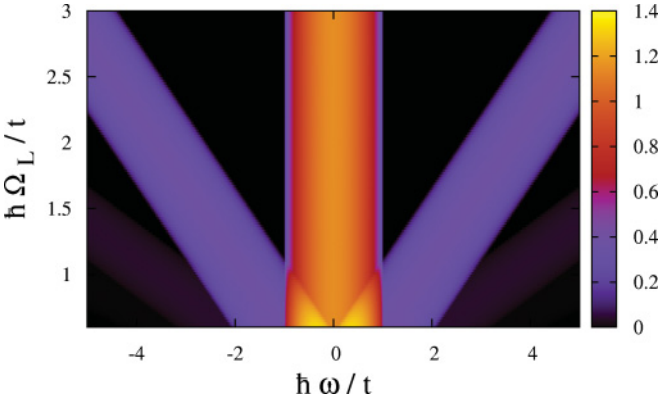

FIG. 4. (Color online) The imaginary part of the local Green's function, Eq. (6), is displayed as a function of quasiparticle energy $\hbar\omega/t$ and external frequency $\hbar\Omega_L/t$ at zero temperature for external amplitude $A_0/t = 2.5$. The original semicircular DOS evolves sidebands as the laser frequency increases. Sidebands of first (bright fuchsia) and second order (faded violet) can be identified.

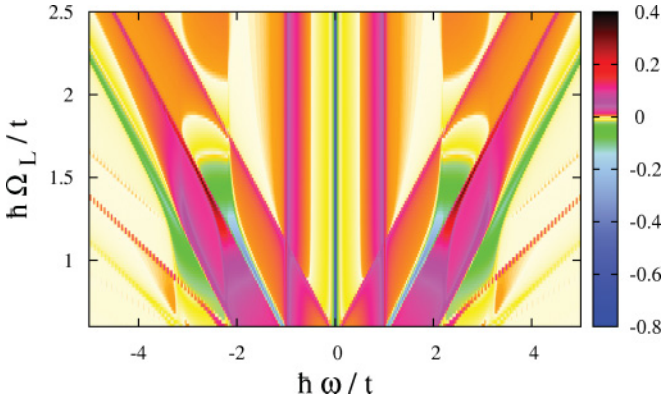

FIG. 5. (Color online) Laser-induced change of the electronic density of states $\delta G(\omega,\Omega_L)$. A Fano resonance around quasiparticle energies $\hbar\omega_0 = 2.34$ eV is found as soon as the external laser field redistributes the electronic spectral weight such that the SOI mode finds electrons with about the same energy to efficiently interact with each other, namely, to get absorbed.

function for this subsystem reads

$$G^R_{mn}(k,\omega) = \sum_\rho \frac{J_{\rho-m}(A_0\tilde{\epsilon}_k)\, J_{\rho-n}(A_0\tilde{\epsilon}_k)}{\omega - \rho\Omega_L - \epsilon_k + i0^+}, \tag{5}$$

where $\tilde{\epsilon}_k$ represents the dispersion relation induced by the external field [Eq. (3)] and is different from ϵ [Eq. (1)]. The J_n are the cylindrical Bessel functions of integer order, $A_0 = \vec{d}\cdot\vec{E}_0$, and Ω_L is the laser frequency. The physical Green's function is given according to

$$G^R_{\mathrm{Lb}}(k,\omega) = \sum_{m,n} G^R_{mn}(k,\omega). \tag{6}$$

IV. RESULTS AND DISCUSSION

We present a numerical evaluation of Eq. (6) in Fig. 4, where $\mathrm{Im}\, G^R_{\mathrm{Lb}}(k,\omega)$ is displayed as a function of quasiparticle energy $\hbar\omega$ and external frequency Ω_L at zero temperature for $A_0/t = 2.5$. As a typical value for the hopping we chose $t = 1$ eV. The SOI is operated at the frequency $\hbar\omega_0 = 2.34$ eV which corresponds to a frequency doubled Nd-YAG laser ($\hbar\omega_0 = 1.17$ eV). The external laser shall be characterized by 10 fs pulses, and shall be $E_0 = 6.299 \times 10^9$ V/m ($I \sim 5.266 \times 10^{12}$ W/cm^2) in the surface region of the Au grains, including Mie-type field enhancement effects due to the small particle sizes. For the nanograins we choose a damage threshold of 0.5 J/cm^2 and $|d| = 6.528 \times 10^{-29}$ A·s·m (in SI units) (with the lattice constant for Au $a_{\mathrm{Au}} = 4.08 \times 10^{-10}$ m) resulting in $A_0 \equiv \vec{d}\cdot\vec{E}_0 = 2.5$ eV and the peak of the electronic response characteristics is developed after 0.3 ps after pulse injection, whereas the damping sets in right afterward and so does the bleaching which is reduced with decreasing particle size.[17] Temperature effects are significantly lowered for nanosize particles and the observed transmission change can be totally attributed to the electronic response.[18] For later use, we also assume a density of 50% nanograins per 10 μm of the SOI. The original semicircular density of states develops photonic sidebands, the band structure, as the external laser frequency Ω_L increases. Because of the point-inversion symmetry of the underlying lattice, the first sideband represents the two-photon processes, the less pronounced second sideband the four-photon processes. Their occupation is described by the nonequilibrium distribution function as calculated from the Keldysh component of Green's function. In a last step, we combine the relaxation processes due to the interaction between the band electrons and the SOI as described by Eq. (1), with the impinging external laser as introduced in Eq. (2). The resulting Green's function consequentially describes the SOI with Au nanograins that themselves are now exposed to the external laser radiation. We treat the weak coupling between the electrons and the SOI photons by second-order perturbation theory and the interaction between the electrons and external laser in terms of the Floquet theory, as demonstrated above. Since we are interested in possible switching effects, we choose as the initial situation the case where the photonic mode, $\hbar\omega_0 = 2.34$ eV, is far off the electronic band edge. Electrons have band energies in the range $-1 \leqslant \hbar\omega/t \leqslant +1$. A Fano resonance is only observable in the presence of external radiation of appropriate frequency, i.e., only if the induced sidebands meet the energy of the SOI mode. This resonance will also affect the transmission properties in the SOI, since now SOI photons can be absorbed in the formation of a mixed state of light and matter with the laser-induced charge excitations in the Au nanograins. Thus a waveguide polariton is created yielding to a significant reduction of the SOI's transmission. In Fig. 5, we display the laser-induced change of the density of states δG as a function of quasiparticle energy $\hbar\omega$ and external laser frequency Ω_L.The quantity δG measures

$$\begin{aligned}\delta G = &[\mathrm{Im}\, G(\omega,\Omega_L) - \mathrm{Im}\, G_{\mathrm{Lb}}(\omega,\Omega_L)] \\ &- [\mathrm{Im}\, G_{\mathrm{wb}}(\omega) - \mathrm{Im}\, G_{\mathrm{b}}(\omega)]\,,\end{aligned} \tag{7}$$

the effect of the impinging laser field on the electronic density of states, and vanishes as the external laser and the coupling to the SOI are turned off. In Eq. (7), G represents the Green's function including all processes, G_{Lb} is the interaction between the laser field and the band electrons as given in Eq. (6), G_{wb} describes the SOI in presence of the band electrons

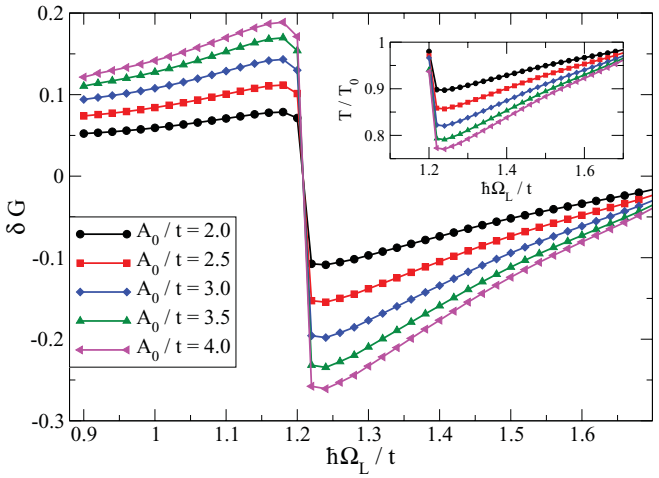

FIG. 6. (Color online) Laser-induced change of the electronic DOS, δG, at fixed quasiparticle energy which meets the discrete value of the SOI mode $\hbar\omega = 2.34$ eV $= \hbar\omega_0$. Inset: Relative photon transmission in a SOI of unit length $l = l_o$ as a function of laser frequency Ω_L.

and is the solution to Eq. (1), and finally G_b is the Green's function of just the noninteracting band electrons. In Fig. 5 the laser-induced change of the electronic density of states (DOS) $\delta G(\omega,\Omega_L)$ experiences a Fano resonance when the external laser redistributes electronic spectral weight leading to the absorption of the SOI mode at $\hbar\omega_0 = 2.34$ eV. That behavior is derived when the first photonic sideband meets the energy of the SOI mode yielding a sign change in δG at this energy. In Fig. 6, the laser-induced change of the electronic DOS δG is shown at fixed quasiparticle energy $\hbar\omega = \hbar\omega_0$, where ω_0 is the frequency of the SOI mode. Asymptotically, i.e., for large Ω_L, δG vanishes, as already indicated in Fig. 4, because in this limit there is no electronic spectral weight at the energy of the SOI mode. In the opposite limit, $\Omega_L \to 0$, the influence of the laser field is nonzero, because here higher-order laser-induced sidebands exist, yielding spectral weight at the resonance position already for smaller laser frequencies. That result can also be concluded from the second-order sideband in Fig. 4. In a SOI of length l, the ratio between the initial and the transmitted intensity is given by $T \sim \exp(-\alpha\, l/l_o)$. Here α/l_o is the absorption coefficient divided by the unit length l_o, where α includes an average over one period of the external periodic driving field with frequency Ω_L. We recognize that $\omega\delta G$ can be understood as the leading contribution to the relative absorption coefficient as discussed in detail in Ref. 19. The relative transmission of photons T/T_0 within the SOI of unit length $l = l_o$ is shown in Fig. 6 as a function of the external laser frequency Ω_L. System parameters are given in the caption of Fig. 5. Depending on the frequency of the driving field, an intensity drop of up to 25% is observed (Fig. 6), and by varying the length of the SOI the transmission inside the SOI can in fact be turned on and off.

V. CONCLUSION

We have presented a quantum-field theoretical model for a SOI in contact with gold nanograins which themselves are exposed to external laser irradiation. The strong and coherent external laser is described in terms of the Floquet theory, assuming classical behavior of this oscillatory-in-time field, whereas the interaction with the SOI mode reflects a quantum interference. The obtained results demonstrate the high potential of SOI polaritons for all-optical switching. Both the frequency and amplitude of the external laser control transmission through the SOI, and each of these features ensures ultrafast switching processes.

ACKNOWLEDGMENTS

The author thanks A. Lubatsch, K.-P. Bohnen, K. Busch, F. Hasselbach, W. Nisch, A. v. Raaij, G. Schön, and M. Wegener for various fruitful discussions. P. Hommelhoff and his group are gratefully acknowledged for highly efficient correspondence. The author is a fellow of the Karlsruhe School of Optics & Photonics (KSOP) and thanks KSOP for support and funding.

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