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Ultrafast switching of photonic density of states in photonic crystals

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It is shown theoretically that the photonic density of states (DOS) of three-dimensional semiconductor photonic crystals can be dramatically changed on ultrafast time scales through two photon excitation of free carriers. Calculations for GaAs inverse opals show that the photonic band gap exhibits a large shift in frequency and a change in width with an appropriate excitation pulse. At certain frequencies, the DOS can be switched from a high value to zero, from zero to a high value, and from a high to zero to a high on 100-fs time scales, independent of the relaxation time of the semiconductor. This technique allows for ultrafast control of spontaneous emission and trapping of photons.

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There is a fast growing interest in photonic crystals; periodic composite materials with typical length scales that match the wavelength λ of light.[1] The optical properties of photonic materials are determined by a spatially varying refractive index, analogous to the periodic potential for an electron in a crystal. Large spatial variations of the refractive index cause a strong interaction between light and the composite structure. A major goal of the field is the realization of three-dimensional (3D) structures that possess a photonic band gap.[2,3] At frequencies inside the band gap, the density of states (DOS) vanishes. This should completely inhibit spontaneous emission of sources inside the photonic crystal.[2] In the presence of weak controlled disorder, Anderson localization of light is also predicted.[3] In this case, a photon may be trapped at a point defect that serves as a cavity with a high quality factor.[4]

Here we investigate switching the DOS in photonic crystals. In contrast with a number of recent proposals[5–7] we show that extremely large changes in the DOS and ultrafast (100 fs) time scales can be simultaneously achieved. Such control opens the door to a number of fundamental studies and applications. An excited atom with a transition frequency in the band gap could suddenly experience a finite DOS, allowing the emission of a photon. The ability to quickly open and close a gap may also allow photons to be encaged near defect cavities for a specified period of time. An ultrafast switchable photonic DOS is of fundamental interest and may be compared and contrasted with the switchable DOS in electronic systems. The ability to control emission and store light is also useful for applications involving lasers and optical information processing.

Experimentally, fast control over the optical properties in the photonic crystal can be achieved by changing the complex index of refraction ($n'+in''$) of one or both of the constituent materials. Such a time-resolved control technique can be judged by at least four factors: (1) the magnitude of the induced relative change in n', which determines the degree to which a band gap is modified, (2) the degree to which induced absorption is minimized, gauged by n'', (3) the time scale over which switching occurs, and (4) the degree of uniformity of the change in a 3D material. In this paper, optical pumping of free carriers in semiconductor photonic crystals via two photon absorption is presented as an ideal means of controlling the properties of a 3D photonic crystal in time. It is shown that with a careful choice of material, pump frequency, pump intensity, and pulse width, this switching technique is optimal with respect to all four of the criteria given above.

It is important to note that photonic band-gap switching is distinct from the switching of Bragg diffraction and concomitant stop gaps.[8–12] At a given frequency in a stop gap, propagation is inhibited for a limited range of wave vectors, but the corresponding change in DOS is usually small.[13] While switchable Bragg diffracting materials may produce effects such as optical limiting and bistability,[14–17] only switching of 3D structures allows complete control over the DOS.

To change the refractive index contrast, the solid backbone of the photonic crystal is excited with a short optical pulse to create a free-carrier plasma via two photon absorption. For the purposes of calculation, the backbone is assumed here to be crystalline GaAs in an fcc inverse opal structure, though free-carrier excitation is easily achieved in any semiconductor material.[18–20] Recent results suggest that the complex index of refraction of excited free carriers in a semiconductor agrees well with the Drude model.[18,20] Thus the optical properties of the excited electrons are determined by the plasma frequency ω_p, and the electron and hole momentum relaxation times $\tau_{e,h}$. The optimum excitation maximizes the change in n' in order to obtain the largest change in the DOS, and minimizes the increase in n'' to prevent photons from being absorbed. This is achieved at moderate free-carrier densities with probe frequencies on the order of ω_p. The effects of such an excitation are shown for GaAs in Fig. 1.[21,22] An optically excited carrier density of $N = 4.8\times10^{19}\ \text{cm}^{-3}$, i.e., $\omega_p = 1.6\times10^{15}$ Hz, gives a large change in n' from 3.36 to 3.08 in the telecom band at $\lambda = 1500$ nm (Fig. 1). Dramatic optically induced changes in n' on short-time scales were empirically demonstrated in recent work on bulk GaAs.[19]

Consider two possible experiments: (1) Light sources such as excited atoms are embedded near the center of a photonic structure with a width of $\sim$20 lattice spacings, sufficiently thick to show a band gap near its center.[23] Efficient sources with emission lines in the band gap will be frozen in their excited state over time scales long compared with the

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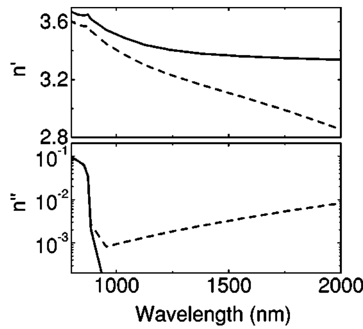

FIG. 1. Drude model predictions for n' and n'' for crystalline GaAs before (solid) and after (dashed) the free-carrier plasma is excited. The excitation described here requires a $\sim 0.1\ \mu J$ pulse to excite a $(20\ \mu m)^3$ region.

lifetime in vacuum. After excitation of free carriers, the DOS is increased from zero to large values, allowing emission of photons to occur (Fig. 2). Because the absorption length is sufficiently long, light can escape from the photonic crystal to be detected. (2) An air-filled defect is made inside a photonic crystal. Light is sent into this cavity at frequencies for which the DOS is finite. Then the DOS is switched to a low value [Fig. 3(c)]. Photons localized in the cavity cannot propagate through the bulk of the surrounding crystal anymore, and are trapped for a duration of the order of the absorption time of the semiconductor, or longer, considering that the largest energy density will be inside the cavity rather than the semiconductor. At this point, the DOS is switched up and the photons are allowed to escape.

For a quantitative prediction of the effects of excitation,

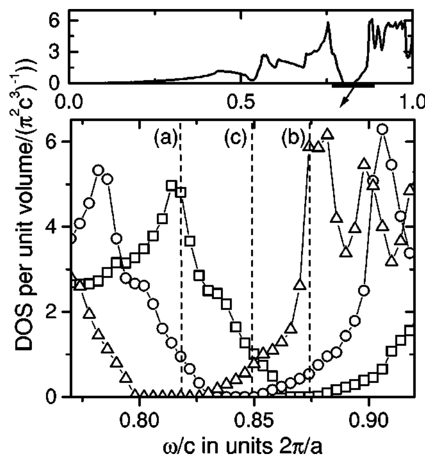

FIG. 2. Top: DOS per unit volume vs frequency for an fcc inverted opal with a backbone refractive index n' of 3.36. The frequency region of interest in the vicinity of the band gap is marked with a solid underscore. The frequency is given in units of $2\pi c/a$ where a is the lattice parameter of the photonic crystal and c the vacuum speed of light. Bottom: DOS in the region of the band gap at $n'=3.36$ (triangles), 3.22 (circles), and 3.08 (squares). Upon excitation of the crystal, the refractive index contrast decreases, causing the band gap to shift to higher frequencies and to narrow slightly. Three frequencies of interest are highlighted by dashed lines, $\omega/c=0.818$ (a), 0.873 (b), and 0.850 (c). The DOS vs time at these frequencies are shown in Fig. 3.

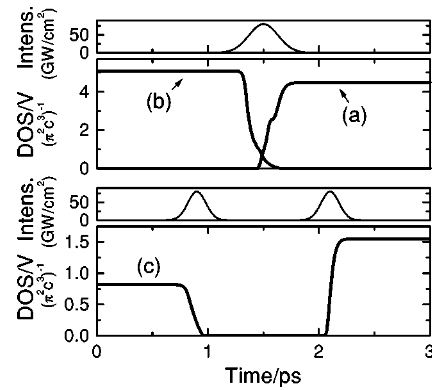

FIG. 3. DOS per unit volume vs time (larger plots) with incident optical excitation pulses (narrower plots) for the three frequencies of interest marked by dashed lines in Fig. 2: $\omega/c=0.818$ (a), 0.873 (b), and 0.850 (c). The pulse parameters are described in the text.

the DOS has been calculated for a nonabsorbing fcc close-packed inverted opal with relevant refractive indices and is shown in Fig. 2.[24,25] The DOS has been calculated for each value of the index of refraction assuming an infinite photonic crystal. We find that changing the refractive index of the GaAs backbone by excitation (cf. Fig. 1) has two effects on the photonic DOS. The reduced index contrast between GaAs and air results in a slight narrowing of the band gap. The change in the effective index of refraction of the photonic crystal causes in essence a rescaling of the frequency axis of the DOS. Indeed, rescaling the frequency axis by the square root of the volume averaged dielectric constant roughly aligns the gross features of the curves. At three key frequencies, marked with dotted lines in Fig. 2, the DOS as a function of the index of refraction $\rho(n')$ is linearly interpolated for $3.08\leqslant n'\leqslant 3.36$, the relevant range at $\lambda=1500$ nm. The index of refraction as a function of time $n'(t)$ is calculated assuming two photon absorption in the GaAs backbone due to the pump pulse. In this way, the DOS as a function of time $\rho(n'(t))$ is obtained under quasistatic conditions for 200–300 fs excitation pulses (Fig. 3).

Several switching effects can be expected at ultrafast time scales. Figure 3 curve (a) shows that at frequencies within the band gap of the unexcited state, the DOS is dramatically increased for experimentally reasonable pulse energies of $\sim 0.1\ \mu J$ injected into a region of $(20\ \mu m)^3$. The peak intensities, fluences, and energies are below the damage thresholds for bulk GaAs.[19] The DOS is increased from nearly zero to a value beyond the DOS of a corresponding homogeneous dielectric, thereby maximizing the relative change. For excited atoms in the photonic crystal, such an event implies that the environment is suddenly switched from inhibiting spontaneous emission to enhancing it. The DOS returns to nearly zero at times longer than the relaxation time of the semiconductor. Figure 3 curve (b) shows that at frequencies above the band gap in the unexcited state, the DOS can be switched from a high value to (nearly) zero. Consequently, emission by excited atoms inside the crystal becomes suddenly inhibited. Figure 3 curve (c) shows the situation for a frequency just above the unexcited-state band edge when two excitation pulses separated by 1 ps impinge on the pho-

tonic crystal. The first excitation pulse switches the DOS from a high to near zero value. The second excitation pulse switches the DOS back up to a high value. In this way, a photonic band gap can be opened and closed within a picosecond, i.e., at a rate that is *independent* of the relaxation time of the backbone.

The effects of absorption, extinction, and finite size can be treated as perturbations to our calculations.[26] The excited free carriers induce absorption as gauged by the increase in n'' in Fig. 1. For a mode with complex frequency $\omega=\omega'+i\omega'', \omega''^{-1}=(n''/n')\omega'^{-1}$ gives the damping time. Here $n'/n''=1340$ since $n''=0.0023$ at $\lambda=1500$ nm.[22] In bulk GaAs, $\omega''^{-1}=1.1$ ps corresponding to a distance of 0.10 mm or ~ 80 lattice spacings for the photonic crystals described here. The damping time in *bulk* GaAs will be the *minimum* possible damping time for a mode in the photonic crystal since any given mode will be distributed over both the GaAs and air in the crystal. The absorption length of a given mode in a photonic crystal is given by $L_a=v_g/\omega''$ where v_g is the group velocity.[26] Extinction due to random scattering can also be described through n'' and experimentally is found to be ~ 3 orders of magnitude smaller than n' in inverse opals.[27] The increase of the DOS in the gap due to absorption and extinction can be approximated from the relation $\rho(\omega)=A(\omega''/\omega_c)/\sqrt{\omega'/\omega_c}$ where ω_c is the frequency at the edge of the gap and A is a geometrical factor of order unity.[26] Inserting the bulk GaAs value for ω'' gives the upper bound $\rho\sim 0.01\rho_{vacuum}$ inside the band gap. Recent work on the effects of finite size suggest that even 5 unit-cell spacings into the photonic crystal $\rho(\omega)<10^{-3}\rho_{vacuum}$ (Ref. 23) and cavities can have a Q of better than 10^4 (Ref. 28) inside the band gap. Thus absorption, extinction, and finite size produce small effects compared to those induced by the large change in n'.

The rise time of the switching will be ultrafast since electrons are excited on a femtosecond time scale.[18–20] The duration of the low index state after excitation is determined by the relaxation time of the carriers, which depends on the degree of disorder in the semiconductor and the nature of the semiconductor band gap. Relaxation times range from 10 ps in amorphous Si (Ref. 10) to ns in GaAs to microseconds in crystalline Si and Ge.[18,20] The free-carrier relaxation time here is assumed to be much longer than the 3-ps time scale in Fig. 3. We expect that relaxation times are relatively short in structured materials such as photonic crystals. An advantage of short relaxation times is that the whole switching process can be repeated quickly.

To uniformly excite free carriers, light must penetrate a substantial distance into the photonic crystal before being absorbed. Such long penetration depths can be achieved by using two photon absorption.[10] In the absence of other processes,[32] the penetration depth for a bulk material is given by $d_{bulk}=1/(\beta I_{peak})$ where β is the nonlinear absorption coefficient and I_{peak} is the peak intensity of the pump beam. At a pump wavelength of $\lambda=1.06$ μm, $\beta=26$ cm/GW for GaAs.[33,34] The penetration depth for the structured material d_{pc} can be estimated at ~ 4 times larger than d_{bulk} since GaAs makes up at most 26% of the volume fraction. With these parameters, I_{peak} may be chosen such that $d_{pc}=20$ μm while the energy per pulse, set by the pulse width δt, determines the maximum excited free-carrier density. Assuming a Gaussian pulse profile and a maximum carrier density of $N=4.8\times 10^{19}$ cm^{-3}, the pulse parameters are $I_{peak}=80$ GW/cm^2 and $\delta t=330$ fs. For the two pulse case, δt can be reduced by lowering the pulse energy since each pulse excites half of the total free carriers (Fig. 3). The parameters here have been chosen with the goal of minimizing δt for the given penetration depth. A larger penetration depth and thus more spatially uniform excitation can be achieved by increasing δt and decreasing I_{peak} at constant pulse energy. The pump beam must also be chosen such that its center frequency is sufficiently far from photonic band gaps to allow for penetration into the material. The uniformity and size of the excitation can be further enhanced by pumping the crystal from several sides simultaneously, see, e.g., Ref. 35.

The induced change $\delta n'/n'$ compares favorably with other techniques such as liquid-crystal reorientation and nonlinear effects. The ordering and alignment of birefringent materials such as liquid crystals, ferroelectric, or ferromagnetic materials can be controlled by temperature or electric and magnetic fields to tune band gaps, as proposed theoretically.[5,6] Several groups have recently tuned photonic Bragg conditions thermally[8,9] and with electric fields.[11] With liquid crystals, large refractive index variations up to $\delta n'/n'\approx 0.1$ are feasible, but typical time scales are limited to the millisecond to microsecond range. Nonlinear optical techniques incorporating the optical Kerr effect can produce changes in the index of refraction at ultrafast time scales. However, the achievable change in n' is at least an order of magnitude less than that achieved via free-carrier absorption. Nevertheless, optical Kerr switching may prove to be complementary to free-carrier excitation for ultrafast switching studies.

The quasistatic approach followed here gives a first description of the dramatic changes expected in the DOS or local DOS. The actual experimental situation is expected to be richer. For instance, an important time scale is given by the inverse of the frequency width Δf of the band gap. Here, $1/(\Delta f)=160$ fs, which is less than the pump pulse duration, hence the quasistatic approximation is expected to capture the essential physics of switched local DOS, while much shorter pump pulses would give rise to new effects. Another interesting issue is the relevant time scales for the frequency of a photon in a defect cavity to adjust to the shifting band gap and to what extent similar considerations apply to excited atomic resonances near a gap. These time scales may depend on the size of the defect and the spacing of defect mode frequencies. Future finite difference time-domain calculations,[28] perhaps including the carrier dynamics, could help to resolve such questions in detail.

It is interesting to briefly compare the physics of modifying an electronic DOS to switching the photonic DOS. In electronic systems, the DOS is usually tuned by an external potential. This potential breaks the translational symmetry of the electron cloud and raises the energy by a constant value. In order to calculate the physical effects, the Fermi level of

the electronic system must also be considered. In contrast, the switching of the photonic DOS described here involves a uniform change in the index of refraction with no change in the lattice symmetry. To first order this rescales the photon energy axis (Fig. 2). Furthermore, since photons are bosons, occupation of levels need not be considered. Since the degree of uniformity of the excitation may be controlled by tuning the pump intensity, other effects such as photonic Bloch oscillations might be observable. Efforts are currently underway in our group to fabricate 3D semiconductor photonic crystals, hence a switchable photonic DOS may be possible in the near future.

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