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Observation of the temporal Bragg-diffraction-induced laser-pulse splitting in a linear photonic crystal

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Temporal Bragg-diffraction-induced laser-pulse splitting into two pulses propagating with different group velocities is observed in multilayered linear photonic crystals (PCs). This phenomenon originates from spatially inhomogeneous light localization within the PCs at the Laue scheme of the dynamical Bragg diffraction. In a homogeneous medium at the PC output each pulse is spatially separated into two pulses, propagating in the transmission and diffraction directions, respectively. The experiments are carried out for a one-dimensional porous silicon-based PC consisting of 375 spatial periods of 800 nm thickness using a femtosecond Ti:sapphire laser as a probe. A linear dependence of the time splitting of each pair of transmitted and diffractively reflected pulses on the crystal thickness is demonstrated and is supported by theoretical estimations.

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I. INTRODUCTION

Periodic multilayer structures referred to nowadays as photonic crystals (PCs) are known to reveal a number of exciting phenomena that originate from spatial periodicity of their optical properties and cannot exist in homogeneous media. These structures demonstrate the existence of the photonic band gap (PBG), which brings about the effects of a strong light localization within a PC and a manyfold amplification of the magneto-optical effects in magnetophotonic crystals, microcavities, etc. It has been shown that the dynamical Bragg diffraction of the electromagnetic radiation in a periodic medium can arise in a structure with a small number of defects, when all the diffracted waves are coherent and strongly coupled [1–7]. If the periodic structure layers are parallel and the reciprocal lattice vector is oriented normally to the PC surface, the incident waves can be reflected from the structure because of the total Bragg reflection (or PBG) [4]. This geometry is called the diffraction-Bragg-reflection scheme.

The Laue transmission geometry is a less popular optical scheme that can reveal the dynamical diffraction effects under the fulfillment of the Bragg-diffraction conditions. In this case PC layers are oriented close to the normal to the input surface and the light waves propagate along the structure without the total reflection [3,4]. Such a scheme of light propagation leads to the so called pendular effect, when the energy of the light traveling through a PC structure is periodically transferred from the transmitted to diffracted beams and vice versa. This effect was studied experimentally in x-ray optics for regular crystals [4] and for PC structures as well [6–9]. Another dynamical diffraction phenomenon is an anomalous weak x-ray absorption in a crystal at the Laue scheme of diffraction, or the Borrmann effect [4,10]. The mechanism of the effect is a different spatial localization of the so called *Borrmann* and *anti-Borrmann modes* within the bulk of a crystal.

While most of the effects mentioned above can exist in both the x-ray and the visible spectral ranges, still the radiation-matter interaction in these spectral areas reveals remarkable differences. The most substantial one is the magnitude of the refractive index modulation which can exceed 10% in the case of a PC that is four orders of magnitude larger than for regular crystals in x rays. Due to this, as was predicted by the dynamical Bragg-diffraction theory for the nonlinear resonant photonic crystals [11] and linear PCs [12], diffraction-induced pulse splitting (DIPS) effect can show up at the Laue-diffraction scheme. Spatially inhomogeneous field localization within the PC at the Laue geometry of the Bragg diffraction is at the heart of the DIPS phenomenon. It was shown that under the described conditions the incident pulse within a PC splits into two pulses that propagate with different group velocities. The first "fast" pulse is formed by two coupled diffracted waves which are localized in low-refractive-index PC layers, thus forming the so called Borrmann mode of the electromagnetic field, whereas the second "slow" pulse is mostly concentrated in high-refractive-index layers and forms the anti-Borrmann mode. Controlling either high- or low-index domains of a PC by an appropriate choice of the constituting material, one can determine separately the parameters for each type of the propagating pulses.

Diffraction-coupled waves in the Borrmann and anti-Borrmann pulses at the output of a PC in a homogeneous medium are spatially separated into transmitted (T) and diffractively reflected (R) waves [Fig. 1(a)], each of them being split into a pair of temporally shifted pulses. According to the theoretical description [12], the time interval between the paired pulses propagating in R and T directions is proportional to the optical path in the PC and to the refractive-index modulation of the neighboring PC layers.

Nevertheless, experimental observation of the DIPS effect has not been performed, mostly due to the difficulty of fabrication of a highly periodic multilayer structure with a large number of periods. In order to perform the optical measurements in the Laue-diffraction scheme, the required thickness of a PC is about several dozens of micrometers. Fabrication of such a thick structure using the convenient multilayer film deposition techniques is a complicated task.

In this paper the experimental observation of the dynamical Bragg diffraction of femtosecond laser pulses at the Laue-diffraction geometry in multilayered porous silicon-based one-dimensional (1D) linear photonic crystals is presented.

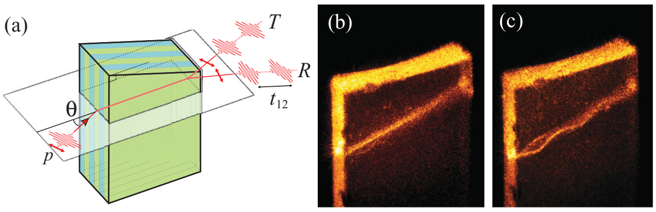

FIG. 1. (Color online) (a) Schematic diagram of doubling of the number of laser pulses caused by splitting of an incident pulse within PC at the Bragg diffraction in the Laue geometry; T and R are transmitted and diffractively reflected pairs of pulses at the PC output; photographs of the laser beam propagating at (b) the exact Bragg angle and (c) tilted to 4° from the Bragg angle.

We demonstrate that the laser pulses transmitted through a PC are propagating in two directions that correspond to the transmitted and diffractively reflected waves. In each of these directions the pulses are split into pairs of pulses with time interval between them of hundreds of femtoseconds. A linear increase of the time shift between the paired pulses with the PC thickness is demonstrated. The experimental results are shown to be in good agreement with the theoretical description based on the DIPS effect.

II. EXPERIMENT

Experimental samples of 1D porous silicon photonic crystals were fabricated using the electrochemical etching technique described in detail elsewhere [13]. A p-type 400-μm-thick Si (001) wafer with the resistivity 0.005 $\Omega\,$cm was used as the anode in a two-electrode electrochemical cell, the second electrode being a platinum wire. A 21% (weight-to-weight) HF:ethanol solution was used as an electrolyte. A one-dimensional porous silicon PC composed of alternating layers of high and low porosity was made by periodic-in-time modulation of the etching current density (40 and 200 $\mathrm{mA/cm^2}$). Consequently, layers with alternating refractive indexes were formed with the porosities $\rho_1 = 0.65$ and $\rho_2 = 0.55$, which corresponds to the refractive indexes of the layers $n_1 = 1.8$ and $n_2 = 2.2$. The PC spatial period is determined by the sum of the thicknesses of the neighboring layers; $d = d_1 + d_2 = 800 \pm 30$ nm, as $d_1 = d_2 = 400$ nm. The sample consisted of 375 spatial periods that correspond to the total thickness of about 300 μm, which was convenient for the optical studies of the DIPS effect.

In order to make a structure transparent, the porous silicon PC was annealed in an oxygen atmosphere at $T = 900\,^{\circ}\mathrm{C}$ for 2 h thus forming a porous silicon oxide PC with the refractive indexes of the alternating layers $n_1 = 1.32 \pm 0.03$ and $n_2 = 1.48 \pm 0.03$, respectively, while the PC thickness was nearly the same. Thus we avoided both the high Si absorption in the visible range, as well as the nonlinear effects such as nonlinear refraction and absorption. The dimensions of the annealed PC sample were $0.3 \times (0.6\text{–}2) \times 5$ mm. The sides of the structure were mechanically poled to form a plane-parallel slab with an accuracy of 0.5°, which is shown schematically in Fig. 1(a). The second sample under study was a wedgelike PC slab with vertex angle of ~10°, so that the PC length along the light path can be varied from 0.9 to 2 mm.

Optical experiments were performed using a Ti:sapphire laser with wavelength of 800 nm, pulse duration of 110 fs, at 80 MHz repetition rate and an average power of 100 mW. The p-polarized laser beam was focused on a cleaved edge of the PC onto a spot of Ø ≈ 30 μm in the Laue geometry as shown in Fig. 1(a). The sample was maintained on a goniometer; the angle of incidence of the probe beam θ was varied in the range from 0° to 45°. The signal at the fundamental wavelength passed through the PC was sent to a correlometer aligned for the transmitted (T) or diffractively reflected (R) light pulses, and the second-order intensity autocorrelation function $I_{AC}(\tau) \propto \int_{-\infty}^{\infty} I(t)I(t+\tau)dt$ was measured, where τ is the time delay between the pulses and I is the intensity of the registered signal.

III. RESULTS AND DISCUSSION

Prior to the temporal measurements, the spatial splitting of the laser pulses in the PC at the Laue-diffraction scheme was studied in order to reveal the structural quality of the PC as well as of the internal interfaces between the porous layers. It was checked that both T and R beams are registered under the exact fulfillment of the Bragg angular condition at $\theta = \theta_B = 31^{\circ}$ for the laser wavelength $\lambda = 800$ nm, in accordance with the theoretical estimations for the structures under study. Figures 1(b) and 1(c) show the photographs of the beam propagation within the PC at the angles of incidence $\theta = 31^{\circ}$ and $\theta = 35^{\circ}$, respectively. It can be seen that in the first case as the Bragg angular condition is satisfied, the light is propagating within the PC structure as a single beam parallel to the constituting PC layers [Fig. 1(b)], while a multiple reflection of the beam within the PC is achieved when the angle of incidence is detuned to 4° from the Bragg angle [Fig. 1(c)]. The Rayleigh scattering in the nanopores of the PC allows visualization of the light beam track within the photonic crystal. For the experiments described below we always affirmed that the laser beam propagation in the sample is similar to that shown in Fig. 1(b).

Temporal splitting of the transmitted and reflected pulses passed through the structure was studied by measuring the autocorrelation function $I_{AC}(\tau)$ under the fulfillment of the Bragg condition. Figure 2 shows the $I_{AC}(\tau)$ function measured for the T and R pulses in a PC of 2.00 ± 0.03 mm thickness normalized to its value at $\tau = 0$. It can be seen that in both cases three maxima are observed, the first one centered at $\tau = 0$ and the two shifted for $\tau = \pm 0.57$ ps, which corresponds to the detection of the *two* pulses shifted in time for $t_{12} = 0.57 \pm 0.01$ ps.

It is worth noting that the temporal shape of all three maxima of $I_{AC}(\tau)$ is identical and correlates with the autocorrelation function of a single laser pulse with FWHM of approximately 110 fs, which is clear evidence that the observed $I_{AC}(\tau)$ dependence corresponds to a true temporal splitting of a femtosecond laser pulse within the PC.

A wedgelike shape of the PC sample allowed studying of the dependence of the time delay t_{12} between the split pulses on the PC length being sure that the structure of the sample is unchanged. The correlation functions and the time delay between the temporally split pulses were measured when displacing the PC wedge with respect to the focused laser

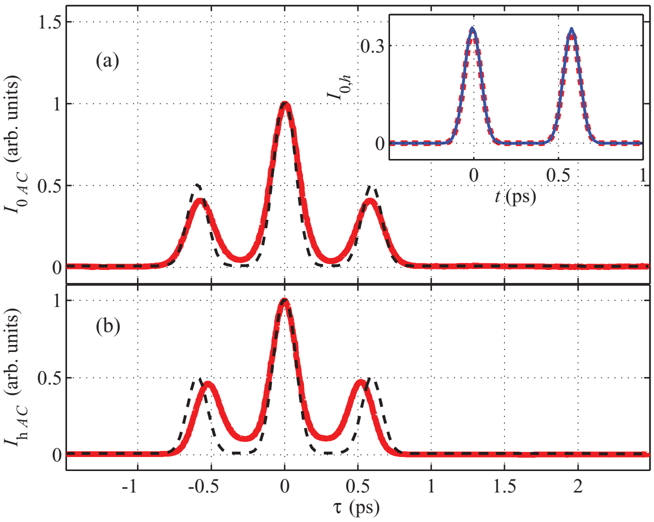

FIG. 2. (Color online) Measured (solid line) and calculated (dashed lines) autocorrelation functions $I_{0,h_{AC}}(\tau)$ of laser pulses passed through the PC in the direction (a) of the transmitted wave at 31°and (b) diffracted wave at -31° in the Laue geometry under the fulfillment of the Bragg-diffraction conditions. (Inset) Output intensities of transmitted $I_0(t) = |E_0(t)|^2$ (solid lines) and diffracted $I_h(t) = |E_h(t)|^2$ (dashed lines) pulses calculated by Eq. (2). The refractive indexes of the layers are $n_1 = 1.350$ and $n_2 = 1.455$, $d = 775$ nm, $d_1/d = 0.45$, $z = 2$ mm, $\lambda = 800$ nm, $\theta = \theta_B = 31^\circ$, pulse duration is 110 fs, and input pulse size along the x axis is 30 μm.

beam. The obtained data are shown in Fig. 3 and demonstrate that t_{12} is a linear function of the PC thickness in agreement with the theoretical predictions for the DIPS effect.

The experimental results were compared with the theoretical calculations of the correlation function $I_{AC}(\tau)$ for the transmitted and reflected pulses (Fig. 2). The pulse propagation within a PC was described when solving the boundary problem of the dynamical Bragg diffraction at the Laue-transmission scheme for spatially confined laser pulses. The field of a femtosecond pulse incident on a PC at $z = 0$ is represented in the form of a two-dimensional Fourier expansion, i.e., as a set of monochromatic plane waves with the central frequency ω_0, amplitudes $A_{\text{in}}(K,\Omega)$, frequencies $\omega = \omega_0 + \Omega$, and the wave vector $k = (\omega/c)$ with the components $k_x = k_{0x} + K$, $k_z = (k^2 - k_x^2)^{1/2}$, $k_{0x} = k_0 \sin\theta$, where θ is the angle of incidence and $k_0 = 2\pi/\lambda_0 = \omega_0/c$. In a two-wave approximation under the Bragg condition $\sin\theta_B = \lambda_0/2d$ [12] the following expression for a complete p-polarized field at a depth z at a time t within the PC bulk were obtained:

$$\mathbf{E}(x,z,t) = [\mathbf{E}_0(x,z,t) + \mathbf{E}_h(x,z,t)\exp(-ihx)] \times \exp(ik_{0x}x - i\omega_0 t), \tag{1}$$

where the amplitudes of the transmitted and diffracted waves are given by

$$E_g(x,z,t) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} B_g A_{\text{in}}(K,\Omega)\exp(iKx - i\Omega t)\,dK\,d\Omega. \tag{2}$$

Here the index $g = 0$, h corresponds to the transmitted and diffracted waves; $B_g = \sum_{j=1,2} F_{gj}\exp(iq_{0z}^{(j)}z)$ are the amplitude coefficients for T and R plane waves in a PC of the thickness z, respectively; $F_{01} = 2/(a_1 + a_2)$, $F_{02} = -bF_{01}$, $b = s_{h1}R_1/s_{h2}R_2$, $a_1 = (s_{01} - bs_{02})/s_{\text{in}}$, $a_2 = (q_{01} - bq_{02})/k$, $s_{\text{in}} = k_z/k$, $s_{gj} = q_{0z}{}^{(j)}/q_{gj}$, $q_{gj} = [(k_x - g)^2 + q_{0z}{}^{(j)2}]^{1/2}$, $R_j = 2\gamma_0\beta_j/C\chi_{-h}$, $q_{0z}{}^{(j)} = k(\gamma_0 + \beta_j)$ are the projections of the wave vectors on the z axis within a PC. The index $j = 1$ corresponds to the Borrmann mode, $j = 2$ to the anti-Borrmann mode of the field, $\beta_j = (1/4\gamma_0)[\alpha \mp (\alpha^2 + 4C^2\chi_h\chi_{-h})^{1/2}]$, $\gamma_0 = (n_e{}^2 - \sin^2\theta)^{1/2}$, $\alpha = h(2q_{0x} - h)/k^2$, $n_e = (n_1d_1 + n_2d_2)/d$ is the average PC refractive index, $C = \cos\theta'$ is the polarization factor for the p-polarized radiation, θ' is the angle between the wave vectors of the T and R waves within a PC, $h = 2\pi/d$, and $\chi_{h,-h}$ are the Fourier components of the permittivity [12].

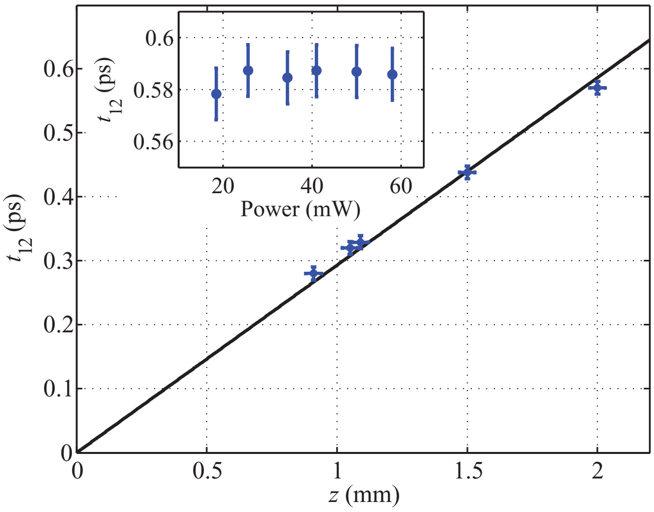

FIG. 3. (Color online) Experimental measurements of the time delay $t_{12}(z)$ of the transmitted pulses vs PC thickness and $t_{12}(z)$ calculated by Eq. (3) (solid line). Refractive indexes $n_1 = 1.347$ and $n_2 = 1.460$; the other parameters are the same as in Fig. 2. (Inset) Time delay t_{12} vs input power in a PC with the thickness of $z = 2.00 \pm 0.03$ mm.

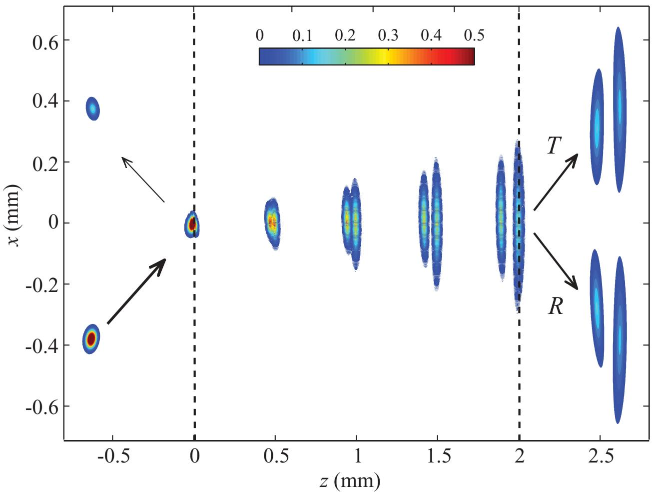

FIG. 4. (Color online) Spatial-temporal dynamics of the total pulse field $|E_0(x,z,t)| + |E_h(x,z,t)|$ in the PC calculated by Eqs. (1) and (2) under exact Bragg condition at different instants t. The parameters are the same as in Fig. 2.

Figure 4 shows the simulation of the spatial-temporal incident pulse dynamics in the PC for the above-described experimental conditions. The T and R fields are calculated using Eqs. (1) and (2) under the Bragg condition. The pulses are represented for different t values that correspond to different pulse positions within the PC. At a depth of $z \approx 1$ mm the propagating pulse is split into two pulses that correspond to the Borrmann and anti-Borrmann modes characterized by different group velocities $v_z^{(j)} = \partial\omega/\partial q_{0z}^{(j)}$. At the PC output ($z = 2$ mm) each of the temporally split pulses is spatially separated into transmitted and diffractively reflected pulses which are observed in the experiment ($z > 2$ mm). Their intensities $I_{0,h}(t)$ are shown in the inset of Fig. 2. The corresponding intensity correlation functions calculated using Eq. (2) for the T and R pulses are in good agreement with the experimental curves as can be seen in Fig. 2.

In the case of a small refractive index modulation $(n_2 - n_1)/n_e \ll 1$ and for the p-polarized radiation the time delay $t_{12} = z/v_z^{(2)} - z/v_z^{(1)}$ can be described by

$$t_{12} = (C|\chi_h|/c\gamma_0^3)\left[2\gamma_0^2(1+p) - n_e^2\right] \cdot z, \qquad (3)$$

where $p = 2\sin^2\theta_B/Cn_e^2$, $C = 1 - 2(\sin^2\theta_B/n_e^2)$, which stays in good agreement with the experimental results (Fig. 3).

IV. CONCLUSION

In conclusion, diffraction-induced pulse splitting of the femtosecond laser pulses at the Laue geometrical scheme of the dynamical Bragg diffraction in one-dimensional periodic multilayer porous silicon-based structures is observed. We have demonstrated that a temporal delay up to 570 fs for the laser pulse of 110 fs duration is achieved for a 2-mm-thick PC. It is proven experimentally that DIPS is a linear effect as it does not depend on the pulse intensity for peak powers up to 2.3 GW/cm^2. The DIPS effect can arise also in two- and three-dimensional PCs where the dynamical diffraction at the Laue scheme can be realized, and could be used, for instance, for the controllable time delay in pump-probe spectroscopy and in laser physics.

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