Measurement of group velocity dispersion for finite size three-dimensional photonic crystals in the near-infrared spectral region

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We measure group delay and group velocity dispersion of silica colloidal photonic crystals in the near-infrared with white light interferometry. While group delay and group velocity dispersion increase with sample thickness, scattering on sample imperfections is found to counteract this development, resulting in group delays lower than theoretically expected. For thin photonic crystals, group velocity dispersion imposed by Fabry–Pérot oscillations is as strong as the one resulting from the periodicity. Group velocity dispersions as high as 30 $ps^2\ m^{-1}$ are observed. All measurements are in good quantitative agreement with numerical calculations. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1857076]

One of the fascinating aspects of photonic crystals is their ability to control group velocity and group velocity dispersion. Sample size and sample quality influence and change these properties. Experimental verification of theoretical predictions is therefore sought after to understand this influence in detail. Most of the experiments have concentrated on one-dimensional (1D)[1–3] and two-dimensional photonic crystals[4,5] and have been designed for microwave frequencies, for which high sample quality is easily achievable and phase shifts due to the macroscopic samples can be measured with network analyzers. Few experiments in the visible were carried out for thick three-dimensional (3D) colloidal photonic crystals using a Mach–Zehnder interferometer[6,7] or time resolved measurements.[8,9] Both experiment types require a tunable laser to cover the spectral region of interest. Strong attenuation at midgap frequencies hindered a detailed analysis close to the band edges and especially across the stop gap for these interferometric experiments. On the other hand, time resolved experiments suffer from pulse reshaping, which complicates the task of assigning a well-defined delay time close to the stop band. Recent theoretical work predicted finite size modification on the propagation of light through 1D photonic crystals.[10–12]

This letter presents white light interferometry measurements of the field transmission coefficient $t(\omega)$ of 3D colloidal photonic crystals (artificial opals) along the ΓL direction for the near-infrared spectral region. Fast Fourier transformation (FFT) of the interferogram allows one to retrieve amplitude and phase spectra over a spectral width of more than 1400 nm and yields therefore a direct measurement of the field transmission coefficient. From the phase information, group delay and group velocity dispersion (GVD) for opals of different thickness are extracted, allowing us to systematically study the influence of finite size on the optical properties. High quality opals of different thickness are prepared for our study from silica spheres of 630 nm diameter using isothermal heating evaporation induced self assembly.[13]

In the experimental setup light from a halogen lamp is coupled into an electronically stabilized, balanced Michelson interferometer via an optical fiber. The light is focused down to a spot diameter of about 200 μm in both arms, one of which contains the sample. The transmitted light of both arms is carefully superimposed and the interferogram is measured with a liquid nitrogen cooled InSb detector (sensitivity range 800–2200 nm). Amplitude $|t|$ and phase $\Delta\phi$ of the field transmission coefficient $t=|t|\exp[iL(k(\omega)-k_0)]$ are calculated via FFT. Measurement and processing times are below 1 s per spectrum. All measurements presented here are averaged over 200 spectra. The first derivative of the phase with respect to the angular frequency results in the so-called group delay[1,9] (or phase time) $\partial\Delta\phi/\partial\omega=L[(\partial k(\omega)/\partial\omega)-(1/c_0)]$, and is proportional to the density of modes.[10] From the group delay, group velocity v_g and GVD $\beta_2=\partial^2 k(\omega)/\partial\omega^2$ can be calculated. c_0 is the vacuum speed of light and L the sample thickness. L can be directly measured in scanning electron micrographs, thus the only free parameter is determined. Although L can be measured quite precisely, care has to be taken during experiments, as the opals are grown on a substrate. To account for the substrate thickness, an identical piece of bare substrate is introduced into the reference arm of the interferometer. Spectra of photonic crystals are normalized to the ones taken from the empty interferometer.

Transmittance spectra of three opal films with different thickness L are depicted in Fig. 1. As in all following figures, the experimental raw data are shown as solid grey lines. The solid black lines represent the experimental data after smoothing over five neighboring points for clarity. The high quality of the opal films is documented by the Fabry–Pérot fringes on the red and blue side of the fundamental stop gap. With increasing thickness [(a) 7 layers, (b) 13 layers, (c) 19 layers], the transmission dip becomes more pronounced, a behavior well known from numerous studies on this system (see, e.g., Ref. 14).

The corresponding group delays, calculated from the phase,[15] are presented in Fig. 2. If parts of the incoming light

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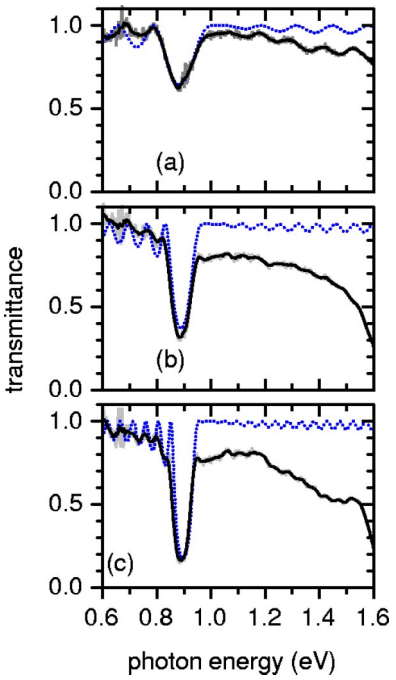

FIG. 1. (Color online) (a) Transmittance spectrum for a 7-layer 630 nm silica opal. Raw experimental data (solid grey lines) are smoothed for clarity (solid black lines). The dotted lines are SWA calculations. (b) and (c) Same as (a) but for opals with 13 and 19 layers, respectively. Note the increasing loss of light due to scattering in the thicker samples.

are absorbed, reflected, or removed from the detection channel by other means, e.g., rotating the polarization,[5] group delay loses its strict meaning as time delay between both interferometer arms. As expected from theory,[10] increasing negative group delay with increasing sample thickness is observed for the fundamental stop band (photon energy of approximately 0.9 eV). Calculated group velocities v_g (not shown) remain well below the limit of $c_0/|t|^2$, preserving causality.[11] Positive group delays observed away from the stop band can directly be assigned with the photonic band structure and interference effects arising from the interfaces (the finite size). With increasing sample thickness, the group delay at the edges of the fundamental stop band increases, indicating the evolution of the density of modes from a slab of effective material to a photonic crystal. Due to the finite size, the density of modes cannot have a singularity at the band edge.[10] Thus the ideal case of zero group velocity (infinite group delay) as derived from band structure calculations cannot be reached. Even come close seems impracticable, as samples with more than 100 sphere layers are required to reduce the group velocity significantly below 10% of the vacuum speed in our low index contrast system. Furthermore, the Fabry–Pérot fringes additionally influence the group delay. Our measurements allow us to quantify upper limits for amplifications factors[16] for devices based on slow photons:[17] for the 19-layer sample, c_0/v_g just reaches a factor of 1.67 at the band edge. The direct applicability of theoretical predictions of amplification factors of several orders of magnitude based on band structure calculations therefore seems questionable for low index contrast structures.

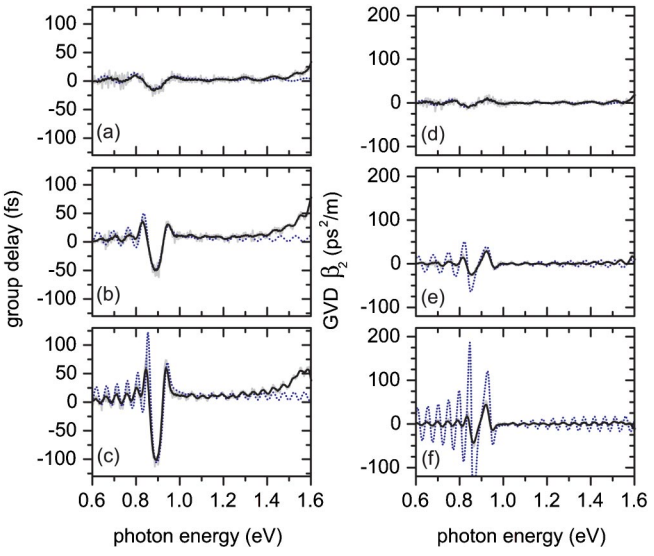

FIG. 2. (Color online) (a) Group delay for the same sample position as in Fig. 1(a). (b) and (c) Same as (a) but for opals with 13 and 19 layers, respectively. Note the almost perfect quantitative agreement between experiment and calculation. (d) Group velocity dispersion derived from the data in 2(a). (e) and (f) Same as (d) but for opals with 13 and 19 layers, respectively. Note that the influence of the finite size for thin opals is almost as strong as the band structure influence. Again, raw experimental data are shown as solid grey lines, smoothed data as solid black lines for clarity. The dotted lines are SWA calculations.

To support our experimental results, we compare them with numerical calculations using scalar wave approximation (SWA) (see, e.g., Ref. 18). By neglecting polarization and taking into account scattering only for the reciprocal lattice vector along the [111] direction, the complex 3D problem is reduced to an analytical solvable one-dimensional problem. Despite these strong limitations SWA is proven to be in good agreement with experiments along high symmetry directions of *finite size* colloidal photonic crystals.[18] As SWA calculates the field transmission coefficient, results can be directly compared with the experiments. For the calculations we assume an index of refraction of $n=1.425$ for the silica spheres[19] and $n=1.5$ for the substrate. Sphere size and sample thickness are obtained from SEM micrographs. There are no free parameters. The results of our calculations are shown as dotted lines in Figs. 1 and 2. The position as well as the strength of the transmission peak are well reproduced in Fig. 1, while we achieve good *quantitative* agreement for the group delay in Fig. 2. Even the small modification of the group delay due to Fabry-Pérot fringes is reproduced. Their experimental detection in the group delay is surprising, as these finite size fringes almost vanish in the transmittance spectra due to increased scattering on lattice imperfections or surface roughness [see Fig. 1(b) and 1(c)]. Measurements of the group delay seem to be much more robust against these imperfections. The increasing discrepancies for higher photon energies (>1.2 eV) between experiment and calculation are explained with the onset of higher bands,[20] which are not accounted for in SWA. We would like to note the qualitative similarity between our group delay measurements and experiments, in which the diffuse intensity is studied.[14] High diffuse intensity is observed, where we find slow group velocity: due to longer interaction times between slow photons and crystal imperfections, the probability of scattering is enhanced or, in other words, the effective mean free path is reduced if the photons are slowed down. Both techniques in combination might prove to be a valuable tool for sample quality characterization.

As slightly different substrate thicknesses for reference and sample arm will introduce an erroneous constant offset

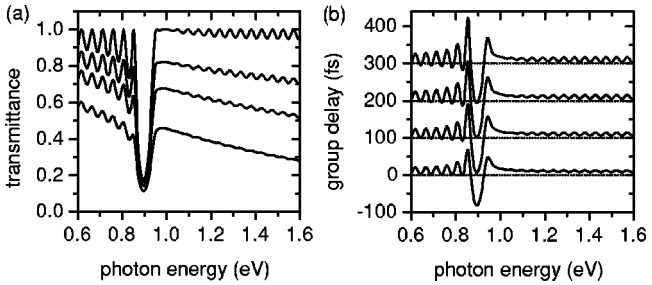

FIG. 3. Theory: (a) Influence of absorption in the spheres on transmittance spectra ($n''=0.000, 0.0025, 0.005, 0.01$ from top to bottom). (b) Group delay for the spectra in (a). Note that increasing absorption in the material mainly reduces the group delay for the materials mode as seen for the data in Fig. 2. Spectra are vertically displaced for clarity.

in the group delay measurements, GVD poses an unambiguous quantity,[8] displayed in Figs. 2(d)–2(f). Although a clear transmission dip around 0.9 eV can be observed in Fig. 1(a), the change in the GVD due to the photonic crystal is only slightly stronger than the one imposed by the Fabry–Pérot fringes. Upon increasing the number of layers [Fig. 2(e) and 2(f)], the dispersion at the band edges increases, but stays below the values expected from SWA calculations, especially seen at the red side of the fundamental stop band [(b) and (c) or (e) and (f) in Fig. 2]. The same observation holds for the group delay. This discrepancy grows with the number of sphere layers, as does the loss of transmitted intensity due to scattering. This behavior can be qualitatively understood: with increasing number of sphere layers the amount of defects increases, leading to enhanced scattering of photons and therefore reducing the effective number of sphere layers sampled by the photons, counteracting the positive effect of increased sample thickness. As scattering mainly occurs for the materials mode, we introduce an effective absorption $n_{\mathrm{silica}}=1.425+n''$ for the spheres into the calculations to model the influence of scattering on group delay and GVD. The results are depicted in Fig. 3. With increasing effective absorption ($n''=0.000, 0.0025, 0.005, 0.01$, from top to bottom) the transmittance spectra become close to the experimental data [Fig. 3(a)], while the maximal group delay for the air mode is reduced from 118 to 68 fs comparable to the experimental values [Fig. 3(b)]. Absorption in the simulation hinders the photons to sample the whole structure, as scattering does in the experiment, resulting in less group delay. This might explain why earlier experiments[6,7] only observed tiny changes in the group delay: the sample quality was too low.

In conclusion, we have presented white light interferometric measurements of group delay and group velocity dispersion in silica colloidal photonic crystals. Due to the low index contrast, group delay and group velocity dispersion for finite size samples stay well below theoretical expectations from band structure calculations. To reduce this discrepancy the sample thickness has to be increased, increasing the amount of intrinsic defects. Enhanced scattering of slow photons on imperfections, on the other hand, is shown to reduce the achievable group delays. The application of finite size, low index contrast photonic crystals for slow photon enhancement of optical properties therefore requires extremely high material quality not yet achievable.

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