Mechanically flexible polymeric compound one-dimensional photonic crystals for terahertz frequencies

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We describe the fabrication and characterization of fully flexible one-dimensional photonic crystals for terahertz frequencies based on alternating layers of a highly refracting polymeric compound and a pure polymer. Due to the high permittivity contrast and low intrinsic material absorption, even a low number of layers yields structures with a pronounced artificial band gap, with a center frequency reflectivity close to unity. Besides the high filter performance, the polymeric compound one-dimensional photonic crystals can be employed for beam shaping by applying a curvature to the mechanically flexible structures, since the spectral characteristics remain nearly unchanged when the structure is flexed. © *2010 American Institute of Physics*. [doi:10.1063/1.3341309]

Despite the considerable interest that the terahertz (THz) frequency range has inspired in the scientific community,[1] the persistent lack of THz system components hampers the broad scale use of THz technology outside the research environment. Considerable effort continues to be directed toward the development of useful THz components.[2–4] In particular, THz interference filters and one-dimensional (1D) photonic crystals have been the subject of intense research. The first implementations were based on alternating polymer layers.[5] However, the performance of these inexpensive and flexible structures was limited by the low refractive index contrast between adjacent layers. Replacing the polymers by thinned silicon wafers[6] and air[7,8] leads to much better filter characteristics but the fabrication costs increase and the structures become fragile. An alternative approach based on ceramics exhibits similar problems.[9,10] Very recently, THz filters based on nanoporous silicon multilayers have been proposed with a differing degree of porosity in the high and the low refracting layers.[11] Especially for devices operating above 1 THz, this fabrication technique seems to be very promising, as very thin layers can be realized. But, the demonstrated filter performance is below that of the hybrid structures discussed in Refs. 6–8.

In this paper, we describe the fabrication and characterization of flexible 1D THz photonic crystals, which can be used as high quality filters or reflectors and additionally may serve for beam shaping purposes. The proposed structures consist of alternating layers of a polymer containing highly refractive rutile titania (TiO_2) and a base polymer. Combining the high dielectric index contrast and the low absorption loss of the hybrid structures with the flexibility, robustness, and inexpensive fabrication of purely polymeric designs, polymeric compound 1D photonic crystals could become key components in THz systems.[12]

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In order to achieve a pronounced artificial band gap, the materials of 1D photonic crystals should exhibit low intrinsic absorption and scattering losses. Furthermore, a high permittivity contrast leads to strong reflections at each individual interface so that a good performance can be obtained even with a small number of layers. This is favorable because the resulting photonic crystal remains thin and flexible. In addition, temporal THz pulse broadening is minimized for high permittivity contrasts between the layers and omnidirectional reflection inside the artificial band gap is achieved. Polymeric compounds, which allow custom tailored dielectric properties by embedding dielectric fillers inside a polymeric host matrix, are ideally suited for this purpose. As base polymers, nonpolar plastics, for example poly(propylene) (PP) or polyethylene, which are nearly transparent for THz frequencies, can be employed. Furthermore, ultrafine filler materials such as rutile titania powder offer relatively low loss and very high permittivity at THz frequencies.[13] Using fillers of a fine grade with particle sizes in the submicron range, scattering effects can be neglected for the relatively long THz wavelengths.

We chose two different pure polymers for the low refracting layers: the polyolefin PP (Moplen HP548R, Basell) and the fluoro-copolymer poly(vinylidene fluoride cohexafluoropropylene) (PVDF). The crystalline domains of the PVDF consist of α phase resulting in a broad absorption band in the vicinity of 1.6 THz.[14,15] The highly refracting layers were obtained by compounding the respective base material with passivated, submicron sized rutile TiO_2 particles. In case of PP, a small fraction of Maleic anhydride grafted PP (Licomont AR 504 fine grain, Clariant) was added by blending with the neat PP to compatibilize the mixture of the non-polar polymer matrix and the polar filler.[16] To fabricate the single layers of the photonic crystal, two following techniques were employed: drop casting[17] in the case of the PVDF-based and film extrusion[18] in case of the PP-based structures. In the drop casting process, a small volume of the PVDF or the PVDF-based compound dissolved in the or-

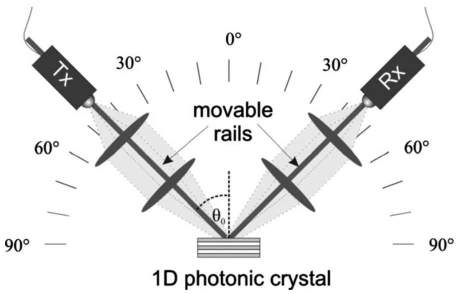

FIG. 1. Reflection setup for angle dependent THz time-domain spectroscopy measurements. The THz radiation is generated and detected by photoconductive antennas gated by femtosecond laser pulses and guided by high-density polyethylene lenses.

ganic solvent cyclohexanone $[(CH_2)_5CO]$ is deposited on a silicon wafer substrate. A drying step follows, so that the solvent evaporates and only the polymer or polymeric compound sheet remains. Afterwards, the single films are stacked with a drop of cyclohexanone in between and the layers are combined under mechanical pressure in a high-temperature sandwiching process. The extrusion process employed in case of the neat PP and the PP-based compound layers utilizes a multi-section extrusion screw to first melt the polymer blend, add the titania, homogeneously disperse the components and degas the melt. Layers with the desired thickness are fabricated by releasing the viscous melt through a variable slot die. An ultrathin layer of a THz transparent epoxy (3M Scotch-Weld Spray 90) is employed to join the single layers to the 1D photonic crystal structure.

In the following section, two photonic crystal designs will be further discussed, both consisting of five layers. In case of the PP-based structure, a center frequency of 200 GHz is chosen. The thickness Δ_i of the layer i in the structure is given by the condition of constructive interference of all internal reflections as

$$\Delta_i = Nc_0/4n_i f_c, \quad N = 1,3,5, \ldots, \tag{1}$$

where c_0 is the speed of light in vacuum, n_i is the refractive index of the ith layer, and f_c is the center frequency of the first band gap. At 200 GHz, the TiO_2-filled PP compound films have a refractive index of 3.42 and the pure PP of 1.52 while the absorption of both the polymer and the compound remain negligible, i.e., typically below 3 cm^{-1}.[19] The thicknesses of the single layers were measured under a light microscope and determined to be 111 μm in case of the highly refracting layers and 269 μm in case of the low refracting layers.

In case of the PVDF-based structures, a center frequency of 300 GHz is chosen. The refractive index of the TiO_2-filled PVDF compound at 300 GHz is 3.18 and 1.48 for the pure PVDF. In contrast to the non-polar PP, PVDF exhibits a non-negligible absorption of 9.8 cm^{-1} at 300 GHz due to its polarity.[20] The single layer thicknesses were determined to be 78 μm in case of the highly refracting and 111 μm in case of the low refracting layers.

All measurements are performed using a commercial fiber-coupled THz time-domain spectrometer. The THz beam is collimated and focused by four 120 mm focal length lenses with the sample placed in the intermediate focus. The THz setup is mounted on movable rails, to enable both reflection and transmission measurements (see Fig. 1). To analytically verify the experimental results of the reflection measurements, conventional transfer matrix simulations are performed.[21] These simulations include losses and dispersion via the frequency-dependent complex refractive index of the component materials.

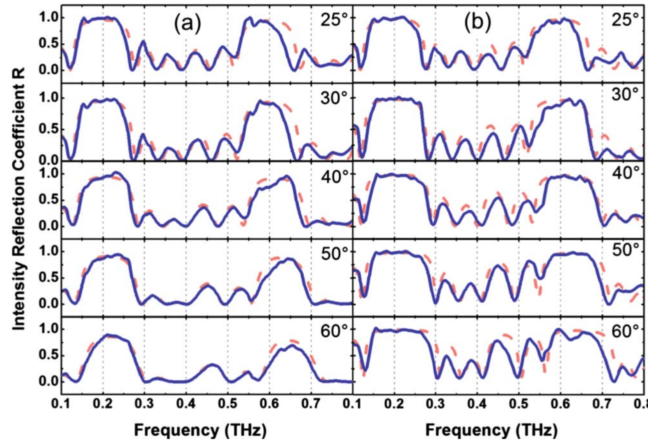

FIG. 2. (Color online) Intensity reflection spectrum of a five layer PP-based 1D photonic crystal for (a) p-polarized and (b) s-polarized incident plane waves. The solid lines represent the measured data and the dashed lines the corresponding transfer matrix simulations.

Figure 2 shows the intensity reflection spectrum for p- and s-polarized incoming waves, respectively, for the PP-based sample. Transfer matrix simulations (dashed curves) agree well with the measured data (solid lines) for all angles and polarizations. Two artificial band gaps are observed within the investigated spectral region, near 200 GHz $[N=1$ in Eq. (1)$]$ and 600 GHz $(N=3)$. In the case of p-polarized incoming waves, the reflectivity decreases toward higher incoming angles and the formerly sharp reflection band edges are softened. This phenomenon results from the presence of Brewster's angle. Yet, unlike photonic crystals with low dielectric contrast, only a small reduction in the reflectivity is observed with increasing angle.

We now explore another interesting property inherent to these structures, namely, their mechanical flexibility. Figure 3 shows the measured and the simulated intensity transmission coefficient of a five-layer PVDF-based 1D photonic crystal. Three different geometries are investigated as fol-

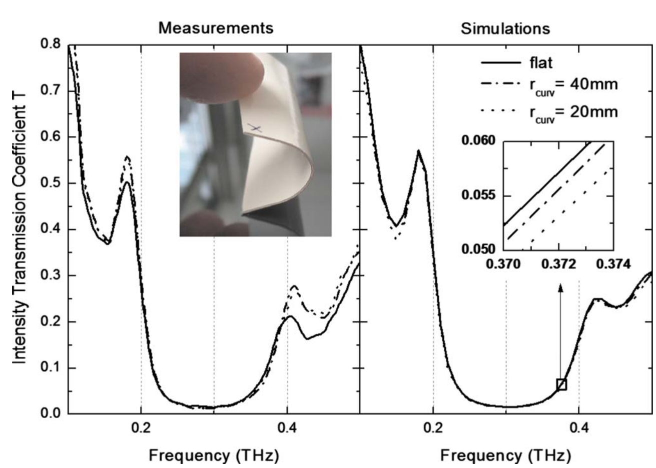

FIG. 3. (Color online) Measured (left) and simulated (right) transmission spectrum for different degrees of curvature applied to a five layer PVDF-based structure. In the left inset, a photo of the investigated curved structure is shown. The right inset presents a zoom-in to visualize the small frequency shift due to curvature.

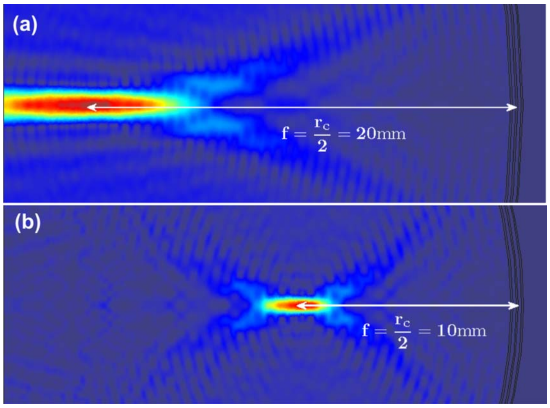

FIG. 4. (Color online) Simulated spatial distribution of the normalized, time-averaged power flow for a curved 1D photonic THz crystal with a curvature radius of (a) r_c=40 mm and (b) r_c=20 mm. For a 300 GHz plane wave excitation originating from the left boundary, a focusing of the reflected waves is observed with the focal length determined by the curvature radius.

lows: the flat structure, and the structure with two different curvature radii (r_c=40 and 20 mm). The measurements are obtained with a collimated THz beam to avoid inaccuracies which might arise from spatial fluctuations in the layer thicknesses. Since transfer matrix simulations cannot account for curvature, we use commercial finite element method software to compute the transmission. All simulations use the experimentally obtained complex permittivity of the single layer materials. We observe that the transmission behavior remains nearly unaffected by the curvature. Only a closeup of the simulated data reveals a slight frequency shift on the order of 1 GHz, which is below the frequency resolution of the measurements. We conclude that compared to the frequency shift induced by varying the angle, the shift due to curvature is negligible.

In contrast to the spectral transmission characteristics, the spatial power flux distribution is strongly affected by the curvature. Figure 4 shows numerical simulations of the normalized, time-averaged power flow for curvature radii of r_c =40 mm and 20 mm, respectively. The curved surfaces focus the wave with a focal length $f=r_c/2$. These 1D photonic crystals can therefore provide integrated beam shaping and filtering functionality.

In summary, we present two different high-contrast 1D photonic crystals for THz frequencies, which employ polymer compounding technology to tailor the dielectric properties of single layers to the design requirements. The structures combine excellent optical performance with mechanical flexibility. In the future, coextrusion technology could be employed to fabricate such devices at low costs in roll-to-roll mass production processes.

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