Reduced density of states governs light scattering in photonic crystals

P.D. García, R. Sapienza, L.S. Froufe-Pérez, and C. López

Instituto de Ciencia de Materiales de Madrid (CSIC) and Unidad Asociada CSIC-UVigo, Cantoblanco 28049 Madrid España.

Contemporary photonic science is capable of addressing fundamental questions at the basis of light-matter interactions, of which the role of the photon density of states (DOS), in nanostructured media is one of the most intriguing.

Artificially engineered materials allow the control of light transport through interference in the internal nanostructure, rather than on the refraction in the body boundaries, engendering new materials properties. Photonic crystals, in which the dielectric constant is periodically modulated, manipulate electromagnetic states and the available phase space and control fundamental aspects of light-matter interaction like light emission [1] and light transport [2]. Unconventional light transport in partially-disordered photonic crystals has only been hinted at by pioneering experiments [3, 4].

Light scattering by weak topological disorder in a photonic crystal and the interplay between order and disorder has yet to be fully understood and explored. As an important step, the relation between scattering extinction and DOS has just been theoretically derived [6]. As pointed out by John [2] a dramatic change in light diffusion can occur for frequencies in or around the band-gap and eventually Anderson localisation of light can be reached, the photonic conductor becoming an insulator [7].

In this work we study the scattering mean free path, ℓ_s, the fundamental building block for any wave transport model, for the special case of photonic crystals with a controlled amount of disorder. We report experimental evidence of strong chromatic dispersion of ℓ_s from band-edge to band-gap, tightly linked to the projected density of states, and values of up to $\sim 100 - 500$ μm, i.e. ~ 300 times the lattice parameter (a), an order of magnitude higher than previously reported [8, 9, 10] and in a spectral region where the total density of states has just a hardly visible feature.

Single scattering events in a system with modified light modes and density of states, as in a photonic crystal, are expected to be very different from those occurring in vacuum due to: a) an increase of light-matter interaction and thus of scattering by defects, when LDOS is increased at the vicinity of band-edges and b) a suppression of the scattering channels, i.e. a increase of ℓ_s in the band-gap, where LDOS is strongly reduced.

The scattering strength can be studied via simultaneous reflection and transmission measurements, when absorption is negligible and for energies below the onset of diffraction. We assume that scattering losses follow Lambert-Beer's law, i.e. that after a thickness L, a ballistic beam attenuates as $I(L) = I_0 \exp(-L/\ell_s)$. The intensity balance can then be expressed as

$$T(L) + R(L) = \exp(-L/\ell_s), \tag{1}$$

where $T(L)$ and $R(L)$ are the ballistic transmission and reflection as a function of sample thickness in a given direction.

In our setup, the thickness of a photonic crystal with different vacancy concentration is accurately measured. This allows us to perform transmission and reflection measurements for different thicknesses and obtain ℓ_s through a fittting to the Lambert-Beer law (1).

Figure 1a shows the measured $\ln(T + R)$ for three different degrees of vacancy doping ?] i.e. for different degrees of extrinsic disorder, at a wavelength of 633 nm and for spheres of 237 nm in diameter $(a/\lambda = 0.52)$. In this type of representation, the slope yields directly $(-\ell_s)^{-1}$ according to Eq. 1. This wavelength is chosen to exemplify a spectral region where no photonic band features are present, as, at such a low energy, the photonic crystal band dispersion is the same as in a uniform homogeneous effective medium. In figure 1a three scattering regimes are clearly distinguishable. For thicknesses lower than *ca.* 10 layers, (regime I), up to $\sim 25 - 30\%$ of the incident light is scattered due to surface effects. When the second regime (II) sets in, the slope ℓ_s^{-1} reaches a stationary value that characterises the photonic crystal. Eq. 1 holds and scattering losses scale with sample thickness like $\sim \exp(L/\ell_s)$. Finally, for larger thicknesses, a third scattering regime (III) appears. Apparently, for thick samples > 50 layers, the self-assembling process loses its effectiveness, as it evident by the increase of intrinsic disorder and by the cracks that appear on thick sample as they start to lift from the substrate.

The physical picture we propose can be checked against consistency if additional disorder is added to the photonic crystals. This can be done by doping the original photonic crystal with a controlled concentration of vacancies. At a wavelength of 633 nm the values of ℓ_s, calculated from the fit to the Lambert-Beer law, are plotted as a function of vacancy density in Fig. 1b. The "perfect" crystal (that with 0% added vacancies) is highly ordered as it presents a scattering mean free path of 63 μm, hundreds of times the lattice constant (in this case $a = 0.33$ μm), and in particular much larger than the Bragg length . An addition of a very little amount of defects rapidly decreases the mean free path, hence the quality of the crystal. In this figure, the inverse scattering mean free path scales linearly
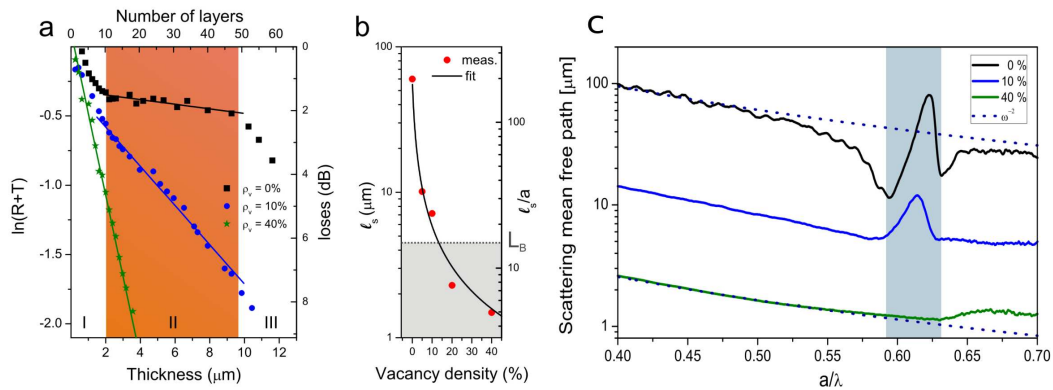
Figure 1: (a) Plot of $\ln(R + T)$ as a function of the sample thickness, at $\lambda = 633$ nm, for different vacancy density doped photonic crystals (from 0% to 40% vacancies doping), of 237 nm diameter. (b) ℓ_s obtained from linear fitting of the slope, for regime II. It also shows the Bragg length (L_B) in the case of $\rho_v = 0\%$ as shaded area. c) ℓ_s as a function of the light wavelength for 0%, 10% and 40% vacancy doped photonic crystals with $d = 237$ nm. The position of the pseudo-gap is shaded in cyan. The dotted line shows the ω^{-2} dependence of ℓ_s far from the band-gap.

with the vacancy concentration ρ_v, as shown by the black line, which is a fit for $\ell_s^{-1} = \rho_0\sigma_0 + \rho_v\sigma_v$ where ρ_0 and ρ_v are the density of intrinsic and intentionally added scatterers and σ_0 and σ_v their scattering cross-section respectively. From the fit of $\ell_s(\rho_v)$ as a function of the vacancy concentration we can estimate σ_v.

Fig. 1.c shows the strong chromatic dispersion of $\ell_s(\omega)$ in the visible range. This is the signature of the photonic crystal. In the low energy side of the pseudo-bandgap, ℓ_s takes on a value of the order of ~ 100 μm for sphere diameter d = 237 nm, the largest values reported so far. Far from the band-gap, $\ell_s(\omega)$ varies as $\sim \omega^{-2}$, dependence that has been confirmed also in previous experiments [8] and attributed to Rayleig-Gans type of scattering.

The large variation of ℓ_s cannot be attributed to the modified DOS only. It can be shown that, under some cirsumstances, the spatially averaged scattering cross section of a rayleigh scatterer is proportional to the DOS and to the reduced density of states (RDOS) which is the contribution to the DOS of modes propagating in the incident direction. Equivalently this magnitude is inversely proportional to the group velocity in this direction. This means that the main contributions to the scattering cross section are due to the coupling of the incident radiation to the scatterer (RDOS) and the total available modes to radiate from the excited scatterer (DOS).

In conclusion, we show that a controlled smooth transition from ballistic to diffuse transport in photonic crystals can be induced by the introduction of extrinsic disorder. We find that the strength of scattering is strongly determined by the projected density of states, which induces immense, up to 20-fold, variations in the scattering mean free path. We propose ℓ_s as a robust, easy to measure, figure of merit in assessing the quality of photonic crystals for technological applications. The possibility of controlling light scattering and diffusion in nano-structured optical media has important implications not only to test the quality of photonic devices, but also to properly address the proximity to the onset of Anderson localization in disordered lattices, or for the spectral control of lasing emission from disordered/ordered active media [5].

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