

Suppression of the local density of states in a medium made of randomly arranged dielectric spheres

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Light propagation in a medium made of densely packed dielectric spheres is investigated by using a rigorous diffraction theory. It is shown that a substantial suppression of the local density of states occurs in spectral domains where the single constituents exhibit Mie resonances. The local density of states decreases exponentially at the pertinent frequencies with a linearly increasing spatial extension of the aggregated spheres. It is shown that a self-sustaining random arrangement of core-shell spheres shows the same fundamental characteristics. Such approach offers a path toward easy to fabricate photonic materials with omnidirectional gaps that may find use in various applications.

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In the past several years light propagation in random media composed of discrete scattering objects has attracted a considerable deal of interest. It derives its fascination not only from numerous potential applications such as, e.g., the random laser,^{1,2} but also because it challenges our fundamental understanding of light propagation. Most notably, the effect of light localization attracted considerable attention.³ Light localization can be understood as a synonym for the occurrence of a complete photonic band gap where spectral components of light within the photonic band gap must not propagate within the medium because Maxwell's equations provide only evanescent wave solutions. By placing a source that emits light at a frequency within the gap inside such material it cannot couple to a radiating field. It remains localized. Light localization in one-dimensional structures may be easily achieved by, e.g., introducing a defect into a sequence of dielectric layers arranged to form a Bragg stack. In two-dimensional media made of infinitely extended cylinders of high permittivity this localization was feasible by exploiting scattering resonances of sufficient strength.^{4,5} As a signature of localization one may use the exponential decay of the local density of states (LDOS) at a point inside the medium with increasing size of a finite system. A significant suppression of the LDOS in a finite system is interpreted as a sign for the appearance of at least a pseudogap. Because scattering resonances also affect the formation of a gap for a periodic arrangement of the cylinders, a complementary computation of the photonic band structure proved that the band gaps occur indeed in the same spectral domain as the reduced LDOS.⁵

In three-dimensional (3D) systems, made of spheres,⁶ this possibility of light localization is controversially discussed as in experiments, absorption may potentially shade localization effects.⁷ To investigate theoretically the possibility of a suppression of the LDOS, one may take advantage of an analytical approach.⁸ But as the structure involves a high index contrast and a size of the scatterers that is comparable to the wavelength, usually the problem requires a rigorous solution of Maxwell's equations.^{9,10} Using a supercell approach rigorous computations of the photonic density of states can be performed. For an amorphous photonic material consisting of a continuous-random network a complete photonic band

gap has been identified recently.¹¹ This work has shown that appropriate short-range order might also permit for light localization, in addition to the well-known suppression of light propagation in periodic media due to Bragg resonances.¹²

However, most experimental investigations of light propagation in 3D random media¹³ employed sufficiently dense packed high permittivity spheres. This is mainly due to their easy fabrication procedure that relies on colloidal chemistry. The excitation of Mie resonances in these spheres is usually regarded as the primary reason for suppressing propagation.^{14,15} To theoretically verify the effects associated with light localization, various efforts were undertaken in the past. In Ref. 16 the density of states was computed for small clusters of spheres using a generalized Mie theory. In Ref. 17 scattering effects depending on the cluster size were studied for low index contrast using the superposition T -matrix method. A large scale finite-difference time-domain (FDTD) method was also used to trace light propagation in ensembles of spheres with a strongly varying size.¹⁸ All these works gave indication that it is possible to significantly suppress the LDOS in a random media, though a proof seems to be missing. Such finding, however, would permit obtaining an optical material with an omnidirectional band gap that could find manifold applications. This report aims at proving that this is indeed feasible and derives criteria that have to be met by the structure in order to observe such an omnidirectional gap.

In this work we focus on investigating the LDOS of large clusters made of randomly arranged identical high permittivity spheres. For this purpose we use the FDTD method.¹⁹ It is shown that the LDOS decays exponentially with increasing size of the cluster in certain frequency intervals. The spectral positions of these domains are correlated with Mie resonances. They occur independently of the arrangement and are tunable by modifying the permittivity of the spheres. However, below a certain contrast the frequency intervals of the suppressed LDOS are hard to recognize. We also investigate the LDOS of core-shell spheres which were arranged by using the sphere dropping method. Consequently, they form a self-sustaining ensemble of spheres. The approach allows maintaining the main optical characteristics if the permittivity of the shell is sufficiently low. This work opens a path toward an easy to fabricate photonic material that al-

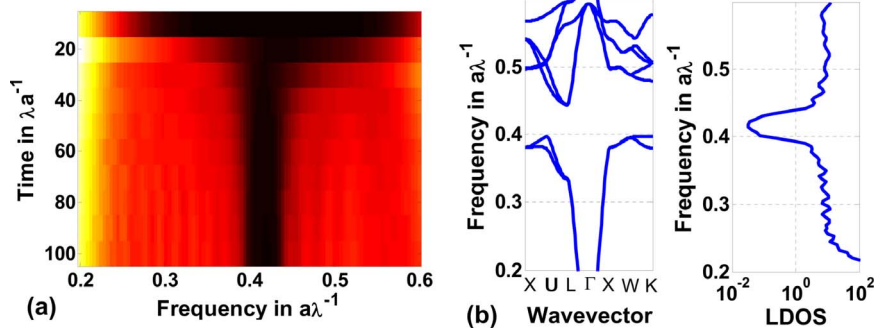


FIG. 1. (Color online) (a) Frequency dependent LDOS on a logarithmic scale for a finite system of dielectric spheres arranged on a diamond lattice as a function of the computational time. (b) Comparison of the band structure for the respective infinite periodic lattice with the LDOS of the system. The significant reduction of the LDOS clearly corresponds to the spectral region of the complete band gap.

lows for a suppression of light propagation regardless of its direction and polarization. Such a material can be applied as, e.g., isotropic filters for solar cells.

In a first investigation we applied our procedure to a system where the spheres are arranged on a diamond lattice.²⁰ The overall purpose is to verify our computational approach. In general, it is well known how the appearance of the LDOS is modified in the presence of a complete photonic band gap.²¹ The results are compared to photonic band-structure computations of the infinite lattice, proving the presence of a complete band gap. We have chosen the spheres to have a permittivity of $\epsilon = 11.56$ and a radius of $R=0.25a$, with a being the lattice constant. Spheres of such size are already slightly overlapping in the diamond lattice. The finite diamond lattice was defined by a $7a \times 7a \times 7a$ cube. The spatial permittivity distribution was discretized in the FDTD method with a resolution of $0.025a$. To compute the LDOS, we located a point source at the center of the structure but outside any sphere and computed the time dependent transmitted power through a sphere that includes the entire structure. The point source was excited by a temporal Gaussian pulse. Fourier transformation of the time dependent signal yields the frequency dependent transmission. Upon a normalization with the power spectrum of the source, we obtain the transmission. In a lossless medium this transmission corresponds to the LDOS associated with the spatial point where the source was placed. On a logarithmic scale the spectrum of the LDOS as a function the normalized computational time is shown in Fig. 1(a).

After a sufficient computational time of $\approx 100\lambda a^{-1}$ all

transient effects faded away. The LDOS converges to a stationary distribution. The logarithmic scale clearly reveals a strong suppression of the LDOS in a frequency interval between $0.4a\lambda^{-1}$ and $0.44a\lambda^{-1}$. The main features of the LDOS are independent on the exact position of the point source within a unit cell as long as it is placed outside any sphere. Comparison of the LDOS to the band structure [Fig. 1(b)] clearly reveals that the spectral region of a suppressed LDOS coincides with the band gap. For a spatial extension of the structure of $7a$ in all dimensions, the LDOS is decreased by approximately 2 orders of magnitude when compared to spectral regions for which propagation of light is allowed.

After having verified the accuracy of the computational approach and having showed how band gaps and reduced LDOS are related, we proceed in analyzing random arrangements of the same spheres where we keep the filling fraction of the spheres constant at 41%. Although a ceases to have a meaning of being the period we use it further as the length scale. Exemplarily, Fig. 2(a) shows a particular arrangement of the spheres. The arrangement was achieved by generating at first randomly a coordinate for a new sphere. Subsequently it was verified if a sphere is already present in the volume to be occupied by the new sphere. If this is excluded, the position is accepted, otherwise it is rejected. The procedure is repeated until the predefined particle density is met. The exclusion of penetrating spheres was necessary to avoid that resonances associated with the spherical shape of the particles (Mie resonances) were appreciably altered. Moreover, as a result of preliminary numerical experiments, nearly touching spheres had to be excluded. This was ensured by

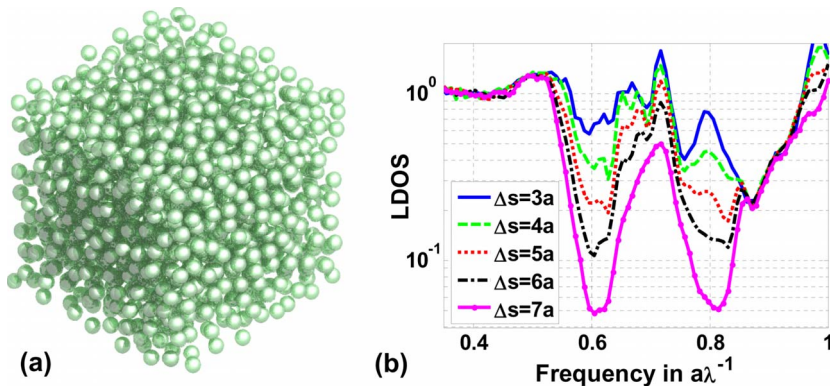


FIG. 2. (Color online) (a) Example of a random arrangement of spheres for which the LDOS was computed. (b) LDOS as a function of the spatial extension of the domain where spheres were randomly arranged.

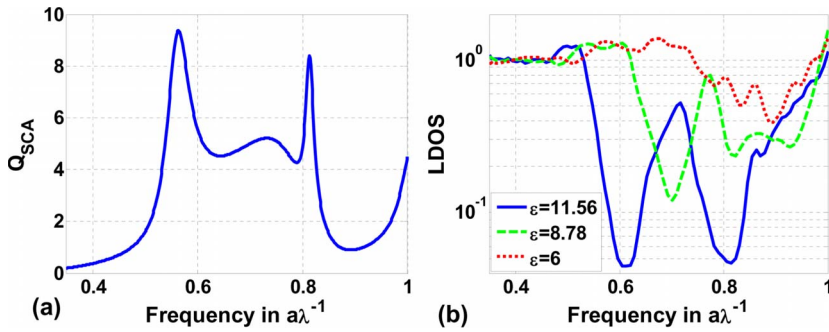


FIG. 3. (Color online) (a) Scattering cross section calculated using Mie theory for a single sphere of radius $R=0.25a$ and permittivity $\epsilon=11.56$. The sphere is embedded in air and illuminated by a plane wave. (b) LDOS for randomly arranged spheres as a function of the sphere's permittivity.

defining the forbidden volume slightly larger. Nearly touching spheres are sufficiently strong coupled to lift the degeneracy of the spectral position of a Mie resonance, causing an inhomogeneous line broadening and a damping of the resonance of the ensemble. As a consequence, the LDOS would not have been sufficiently suppressed. Hence placing a new sphere is inhibited within a radius of $0.3a$ of an existing sphere. This seemingly limitation will be subsequently lifted by covering the spheres in the simulation with a core material having a low dielectric constant. Such arrangement permits for a dense packing of the spheres but maintaining the characteristics of the LDOS.

The LDOS of a medium composed of randomly arranged spheres as a function of the spatial extension of the domain that hosts the spheres is shown in Fig. 2(b). At low frequencies the LDOS remains constant. The source is always located in the center of the spatial domain of interest but outside any sphere. On the contrary, at slightly elevated frequencies two bands exist where the LDOS shows the exponentially decaying strength with increasing linear size of the system. It suggests the appearance of at least a pseudogap. The two bands are centered at $\approx 0.59a\lambda^{-1}$ and at $\approx 0.81a\lambda^{-1}$, respectively. By using different random arrangements slight modifications in the LDOS were encountered, though the principal features of a suppressed LDOS in the same spectral domains were always observed. The gaps therefore seem to be completely independent on the relative orientation of the source with respect to the structure.

Both peculiar frequencies can be correlated with Mie resonances of the isolated dielectric sphere.²² For this purpose the scattering cross section is shown in Fig. 3(a). Two sharp resonances can be seen at $0.56a\lambda^{-1}$ and at $0.81a\lambda^{-1}$. From the Mie coefficients it can be deduced that they correspond to resonances of the lowest eigenmodes of the electric (c_0) and the magnetic (d_0) potentials inside the sphere, respectively. The reason why the suppression of the LDOS occurs at slightly larger frequencies when compared to the Mie resonances lies potentially in the requirement that the field scattered by the sphere oscillates π out of phase with respect to the incident field. This will effectively suppress the propagation of light in the forward direction since scattered and illuminating fields annihilate due to destructive interference provided that the scattering strength of either sphere is sufficiently strong. This required phase difference is encountered, as in all resonance phenomena, slightly above the resonance frequencies.

Reducing the permittivity of the sphere will have various implications. Foremost, the spectral position of the reso-

nances shifts toward larger frequencies, the resonance gets weaker and broader, and overall, the scattering strength is reduced. These modifications of the LDOS for randomly arranged spheres are shown in Fig. 3(b). The spatial extension was chosen to be $(7a)^3$. The spectral regions of the reduced LDOS shifts to higher frequencies being in perfect agreement with the prediction of Mie's theory. Furthermore, the anticipated smaller reduction of the LDOS for a smaller permittivity is evident. If the permittivity of the spheres is smaller than $\epsilon \approx 6$, defining a domain of suppressed LDOS is pointless.

Because the structures investigated up to now cannot be fabricated, we now investigate core-shell particles. The shell is made of a material with a sufficiently low permittivity. It should not affect the strength of the scattering resonance of the core. Furthermore, the shell should have an appropriate thickness to allow for a sufficient separation of the core spheres. As mentioned we have chosen a core-shell radius of $0.3a$ with a shell permittivity of $\epsilon=2.25$. The core features were kept invariant ($R=0.25a$, $\epsilon=11.56$). The self-sustaining ensemble, reasonable for fabrication, was generated with the sphere dropping method. In this method a sphere is dropped into a fictitious box. Gravity forces the sphere to sink down (along the z axis) to the bottom of the box. If the dropped sphere touches another sphere, a translation in the x - y plane of the dropped sphere is performed. The translation is chosen such that the distance between the dropped sphere and the other spheres is maximized. It allows to sink the dropped sphere further down until it touches again another sphere. The procedure is repeated until the dropped sphere cannot sink further, although it is incrementally replaced in the x - y plane in an arbitrary direction. Once this stage in the procedure is reached, the sphere is stably trapped by its surrounding.

The procedure allows to generate self-sustaining ensembles of randomly arranged spheres with structural integrity. To force the randomness we assumed hard boundaries. The final spatial extension of the structure is the same as before and amounts to $(7a)^3$. On passing we note that in small spatial domains a certain periodicity is encountered as the spheres tend to form an opal structure. But as no energy minimum for the global arrangement was forced, the structure will retain its random character in general. A distribution of spheres generated with this method is shown in Fig. 4(a). The filling fraction amounts to 54%.

The corresponding LDOS for such a core-shell structure is shown in Fig. 4(b). For the sake of comparison we show also the LDOS for the structure without shells. In this case

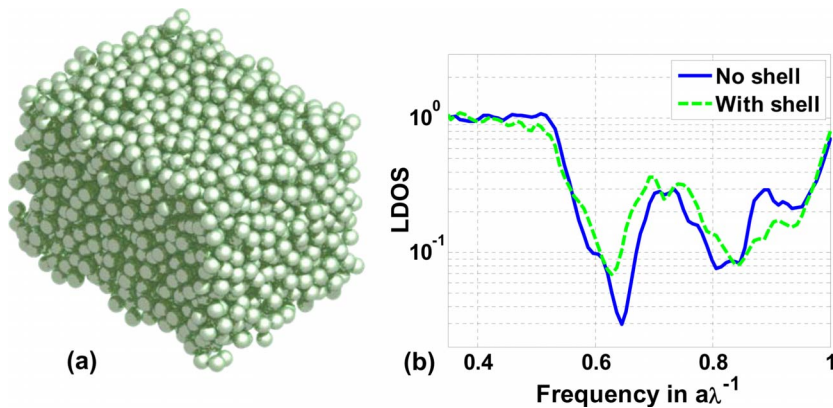


FIG. 4. (Color online) (a) Spatial arrangement of a self-sustaining ensemble of randomly arranged spheres as generated by the sphere dropping method. (b) LDOS for cores with or without shells. The arrangement is only self-sustaining if the spheres consist of cores and shells.

the larger density of spheres results in a stronger suppression of the LDOS when compared to the scenario analyzed before. However, the spectral positions of the suppression dips coincide. The optically weak shell causes primarily a slightly increased LDOS in the spectral domain of interest, though it remains some orders of magnitude smaller compared to spectral domains where light propagation is allowed. A slight shift of the spectral domains of reduced LDOS appears for the second resonance frequency of $0.805a\lambda^{-1}$. Thus we conclude that the shell does not significantly affect the properties of the photonic material, though a minor degradation can be observed. On the other hand, the shell is sufficiently thick to guarantee the structural integrity and to make the fabrication of the photonic material feasible.

Such compactness is extremely important if this photonic material shall be used in applications. The omnidirectional gap can be exploited, e.g., by using this material as intermediate reflector in tandem solar cells. The problem to be solved in such tandem solar cells is to define an intermediate reflector that reflects all light in a certain frequency interval and independent of the angle of incidence back into the top cell, leaving the other spectral components unaffected. Maximizing the light absorbance in the top cell allows to increase

the overall efficiency of the solar cell. The proposed photonic material is very promising for such an application, as it acts sufficiently efficient and can be fabricated at low costs.

In conclusion, we have investigated the local density of states for a photonic material made of randomly arranged spheres. It was shown that a significant suppression of the LDOS is encountered in frequency domains slightly above the Mie resonances of the single sphere. The observation of the effect requires a minimum distance to adjacent spheres. The magnitude of the LDOS decays exponentially with a linearly increasing size of the photonic material indicating the presence of a photonic band gap of the infinite structure. Covering the spheres with a shell of a permittivity as small as feasible allows to observe the same phenomena for self-sustaining ensembles. The theoretical concept might be a way to fabricate a photonic material at low costs that suppresses the light propagation along all directions, representing an omnidirectional band-gap material.

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