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Structural correlations and dependent scattering mechanism on the radiative properties of random media

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ABSTRACT

The dependent scattering mechanism is known to have a significant impact on the radiative properties of random media containing discrete scatterers. Here we theoretically demonstrate the role of dependent scattering on the radiative properties of disordered media composed of nonabsorbing, dipolar particles. Based on our theoretical formulas for the radiative properties for such media, we investigate the dependent scattering effects, including the effect of modification of the electric and magnetic dipole excitations and the far-field interference effect, both induced and influenced by the structural correlations. We study in detail how the structural correlations play a role in the dependent scattering mechanism by using two types of particle system, i.e., the hard-sphere system and the sticky-hard-sphere system. We show that the inverse stickiness parameter, which controls the interparticle adhesive force and thus the particle correlations, can tune the radiative properties significantly. Particularly, increasing the surface stickiness can result in a higher scattering coefficient and a larger asymmetry factor. The results also imply that in the present system, the far-field interference effect plays a dominant role in the radiative properties while the effect of modification of the electric and magnetic dipole excitations is subtler. Our study is promising in understanding and manipulating the radiative properties of dipolar random media.

1. Introduction

Studying the radiative properties of micro/nanoscale disordered media is not only of great fundamental importance in understanding the light-matter interaction physics, like Anderson localization [1–4] and anomalous transport behaviors of radiation (or light) [5–8], but also has profound implications in applications such as random lasers [9,10], solar energy harvesting and conversion [11–14], radiative cooling [15,16] and structural color generation [17], etc. In such media, radiation is scattered and absorbed in a very complicated way, which is usually described by the radiative transfer equation (RTE) in the mesoscopic scale. The radiative properties entering into RTE, including the scattering coefficient κ_s, absorption coefficient κ_a and phase function $P(\mathbf{\Omega}', \mathbf{\Omega})$ (where $\mathbf{\Omega}'$ and $\mathbf{\Omega}$ denote incident and scattered directions, respectively), depend on the microstructures as well as the permittivity and permeability of the composing materials. Conventionally, for disordered media containing discrete scatterers, the radiative properties are theoretically predicted under the independent scattering approximation (ISA), i.e., in which the discrete inclusions scatter electromagnetic waves independently without any interference effects taken into account [18–22].

ISA is valid only when the scatterers are far-apart from each other (i.e., the far-field assumption) and no inter-particle correlations exist (i.e., independent scatterers) [19–23]. When the two conditions are violated, the scattered waves from different scatterers interfere substantially and thus ISA fails [24–27]. This fact leads to many authors into the considerations on the effect of "dependent scattering" in the last a few decades [24–36], in order to correctly predict the radiative properties. Generally, the mechanisms of dependent scattering can be classified into two categories. The first category is the recurrent scattering mechanism [37,38], which denotes the multiple scattering trajectories visiting the same particle more than once and resulting in a closed or half-closed loop. This includes the well-known phenomena such as Anderson localization [1] and the coherent backscattering cone [23]. The other category is the interference induced by the structural correlations. Taking the random media composed of hard particles as an example, the finite size of a particle would create a rigid exclusion volume that forbids other particles to penetrate into, which leads to structural correlations in terms of the particle position distribution probability functions [39–41]. The structural correlations will lead to definite phase differences among the scattered waves, which can well preserve over the statistical average procedure. Therefore

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constructive or destructive interferences among the scattered waves occur and thus affect the transport properties of radiation. This is also called "partial coherence" by Lax [42,43]. Moreover, when the correlation length of particle positions is comparable with the wavelength, the structural correlations then play a central role in determining the radiative properties [39,41,44].

Generally speaking, the structural correlations are not only affected by the size and packing density (or volume fraction) of the scatterers, but also by the interaction potential between them. Several typical kinds of interaction potential among particles, for example, the pure hard-sphere potential [39,45], the surface adhesive potential [46,47] and the interparticle Coulombic electrostatic potential [41,48], can be realized experimentally. By controlling the interaction potential and thus the structural correlations, a flexible manipulation of the radiative properties of random media can be achieved [49–51].

In this paper, we consider random media consisting of nonabsorbing, dipolar spherical nanoparticles, in which high-order Mie multipolar modes in the particles are negligible and only electric and magnetic dipoles are excited [52–56]. We aim to comprehensively reveal the dependent scattering effects on the radiative properties, which are induced and influenced by the structural correlations, based on our recently developed rigorous theory [35]. The theory provides analytical expressions for the effective propagation constant, scattering coefficient and phase function for the random media, by means of the multipole expansion method and quasicrystalline approximation (QCA) for the Foldy-Lax equations (FLEs). By investigating two types of particle systems, i.e., the hard-sphere system and the sticky-hard-sphere system, we demonstrate in detail how the structural correlations play a role in the dependent scattering mechanism, including the effect of modification of the electric and magnetic dipole excitations and the far-field interference effect. We show that the inverse stickiness parameter, which controls the interparticle adhesive force and thus the structural correlations, can tune the radiative properties significantly. We also demonstrate the complicated interplay among the random packing effect, the surface adhesiveness and the single particle scattering property, which determines the manifested radiative properties. The results imply that in the present system composed of moderate-refractive-index dipolar particles, the far-field interference effect plays a dominant role in the radiative properties while the effect of modification of the electric and magnetic dipole excitations is subtler. Our study is promising in understanding and manipulating the radiative properties of dipolar random media.

2. Theory

In this paper, we will consider the radiative properties of a random medium consisting of N identical dipolar particles. In the random media, all the particles are assumed to be isotropic, homogeneous and hard spheres with a radius of a. Their positions are regarded as fixed if they are static or move sufficiently slower than the electromagnetic waves [23,57]. Furthermore, the random medium is also supposed to be statistically homogeneous and isotropic. We will not take any quantum or nonlinear effects into account. Under these assumptions, we will briefly summarize the main formulas of this theory to determine radiative properties of such media considering the dependent scattering effects [35].

2.1. Effective propagation constant and scattering phase function

Following from our assumptions on the random medium, the electromagnetic interaction of the incident light with it is then described by the well-known Foldy-Lax equations (FLEs), which are equivalent to Maxwell equations in terms of multiple scattering of light. The FLEs for N particles are given by Tsang et al. [40,42,58,59]

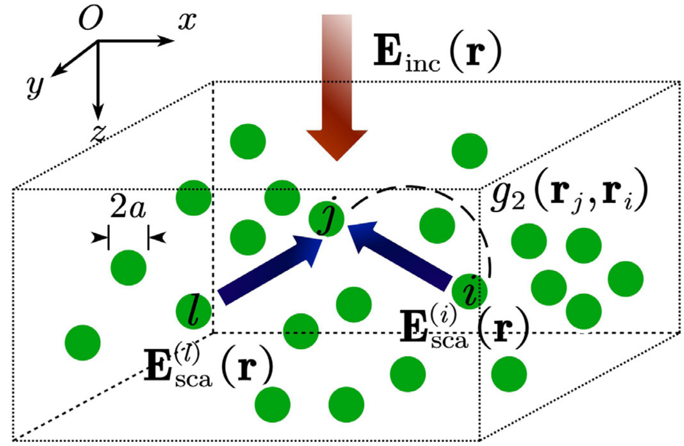

Fig. 1. A schematic of Foldy-Lax equations for multiple scattering of electromagnetic waves in randomly distributed spherical particles. The dotted lines stand for an imaginary boundary of the random medium slab. The particles are denoted as *i, j, l*, etc. The dashed line stands for the pair distribution function (PDF) $g_2(\mathbf{r}_j, \mathbf{r}_i)$ between the jth and ith particle. The thick arrow indicates the propagation direction of the incident wave (which is along the z-axis), while the thin arrows stand for the propagation directions of the partial scattered waves from particle i to j and from l to j.

$$\mathbf{E}^{(j)}_{\rm exc}(\mathbf{r}) = \mathbf{E}_{\rm inc}(\mathbf{r}) + \sum_{\substack{i=1\\ i\neq j}}^{N} \mathbf{E}^{(i)}_{\rm sca}(\mathbf{r}), \tag{1}$$

where $\mathbf{E}_{\rm inc}(\mathbf{r})$ is the electric field of the incident radiation, $\mathbf{E}^{(j)}_{\rm exc}(\mathbf{r})$ is the electric component of the so-called exciting field impinging on the vicinity of the jth particle, and $\mathbf{E}^{(i)}_{\rm sca}(\mathbf{r})$ is electric component of partial scattered waves from the ith particle. The schematic of FLEs is shown in Fig. 1.

For spherical particles, it is convenient to expand the electric fields in vector spherical wave functions (VSWFs), i.e., the eigenfunctions of the vectorial Helmholtz equation under the spherical boundary condition, where the expansion coefficients correspond to multipoles excited by the particles [59,60]. The exciting field $\mathbf{E}^{(j)}_{\rm exc}(\mathbf{r})$ is then expanded as

$$\mathbf{E}^{(j)}_{\rm exc}(\mathbf{r}) = \sum_{mnp} c^{(j)}_{mnp}\mathbf{N}^{(1)}_{mnp}(\mathbf{r}-\mathbf{r}_j), \tag{2}$$

where $\mathbf{N}^{(1)}_{mnp}(\mathbf{r}-\mathbf{r}_j)$ is the regular VSWF, defined in Appendix A). The summation Σ_{mnp} is abbreviated for $\sum_{n=1}^{\infty}\sum_{m=-n}^{m=n}\sum_{p=1}^{2}$. Here n is set to be 1 because only electric and magnetic dipole modes are taken into account. This is valid when the dipolar particles are not so densely packed that the near-field coupling may excite higher order multipoles. The subscript $p = 1, 2$ denotes magnetic (TM) or electric (TE) modes, respectively. From the expansion coefficients of the exciting field, the scattering field from the ith particle can be expressed as [20,59]

$$\mathbf{E}^{(i)}_{\rm sca}(\mathbf{r}) = \sum_{n=1,mp} c^{(i)}_{mnp}T_{np}\mathbf{N}^{(3)}_{m1p}(\mathbf{r}-\mathbf{r}_i), \tag{3}$$

where $\mathbf{N}^{(3)}_{mnp}(\mathbf{r}-\mathbf{r}_j)$ is the outgoing VSWF, defined in Appendix A). For a homogeneous spherical particle the T-matrix elements T_{np} are Mie coefficients, i.e., $T_{12} = a_1$ for the electric dipole and $T_{11} = b_1$ for the magnetic dipole, which can be found from standard textbooks [61,62] and are not listed here. Inserting Eqs.(2) and (3) into Eq.(1) and using the translation addition theorem for VSWFs at different origins as well as the orthogonal relation of VSWFs with