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Effect of dependent scattering on light absorption in highly scattering random media

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ABSTRACT

In the last a few decades, the approximate nature of radiative transfer equation (RTE) leads to a bunch of considerations on the effect of dependent scattering in random media, especially in particulate media composed of discrete scatterers. This effect usually indicates those deviations of RTE from experimental and exact numerical results due to electromagnetic wave interference. Here we theoretically and numerically demonstrate the effect of dependent scattering on absorption in disordered media consisting of highly scattering scatterers. By making comparisons between the independent scattering approximation-radiative transfer equation (ISA-RTE) approach and the full-wave coupled dipole method (CDM), we find that deviations between the two approaches increase as the scatterer density increases. The discrepancy also grows with the optical thickness of the whole random media. To quantitatively take dependent scattering effect into account, we develop a theoretical model of the dependent-scattering corrected radiative properties, based on the path-integral diagrammatic technique and the quasi-crystalline approximation (QCA) in the multiple scattering theory. The model results in a more reasonable agreement with numerical simulations. The present work is of practical importance in correctly modeling light absorptance in random media and interpreting the experimental observations in various applications for random media, such as solar energy concentration, micro/nanofluids, structural coloration, etc. It also has profound implications for the coherent scattering physics in random media with absorption.

1. Introduction

Light propagation in complex and random media is a very general problem in many disciplines of science and engineering, such as radiative heat transfer [1,2], optics and photonics [3,4], astrophysics and remote sensing [5,6], soft mater physics and chemistry [7], biomedical engineering [8] and so on. Generally, in such media, light is scattered and absorbed in a very complicated way, which not only depends on the features of incident light (frequency and polarization state, etc.), but also heavily depends on the properties of media. These properties mainly include the permittivity and permeability as well as their frequency and spatial dispersion (if any) of the composing materials, the morphology (size, shape and topology) of inclusions, and their time- and temperature-dependency (if any). Conventional parameters describing light propagation in random media, i.e., the scattering coefficient μ_s, absorption coefficient μ_a and scattering phase function $P(\Omega', \Omega)$, are defined in the framework of the radiative transfer equation (RTE) [9,10]. Although the RTE is derived phenomenologically in its initial stage, it is later found to be an approximate form of the Bethe-Salpter equation for classical photons [9–12]. The latter is an rigorous equation originally taken from quantum field theory and exactly equivalent to Maxwell equations for electromagnetic waves, accounting for all interference phenomena for the transport of electromagnetic field correlation function $\langle \mathbf{E}\mathbf{E}^* \rangle$ in random media (where $\mathbf{E}$ and $\mathbf{E}^*$ denotes the electric component of the field and its complex conjugate, and $\langle \cdot \rangle$ stands for the ensemble average).

Particularly, for discrete random media composed of a disordered distribution of scatterers, the RTE is valid when the following two conditions are simultaneously satisfied: (1) the scatterers are far apart from each other and each scatterer scatters light as if no other scatterers exist. This is called the independent scattering approximation (ISA); (2) the interference between each multiple scattering trajectory and its time-reversal counterpart is neglected [6,9,12]. The latter condition is called the ladder approximation because in this approximation, the Feynmann diagrammatic representation of the field correlation function resembles a series of ladders [6,9,12]. The approximate nature of RTE thus leads to a

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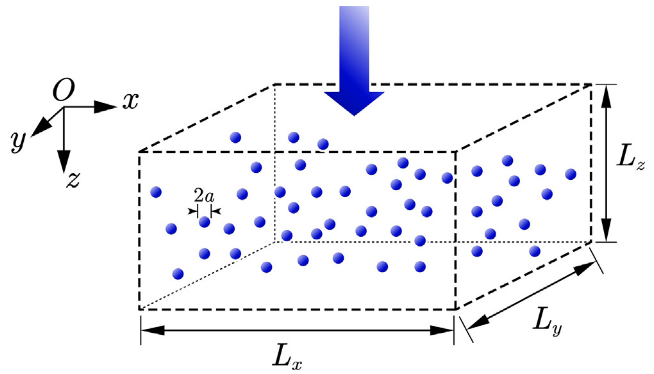

Fig. 1. A random medium with a slab geometry consisting of randomly distributed spherical particles. Incident radiation is denoted by the large, solid arrow.

bunch of considerations on the effect of "dependent scattering" in the last a few decades, which usually indicates those deviations of RTE (combined with ISA) from experimental and exact numerical results, due to the interference of electromagnetic waves [1,13–19]. Note herein "dependent scattering effect" is termed as a generalization for those interference effects that are not possible to explain under ISA [20,21].

There are already several dependent scattering mechanisms recognized both theoretically and experimentally, including the recurrent scattering effect [22,23], the coherent backscattering effect (also called weak localization) [6] and the well-known Anderson (strong) localization of light [24]. It should be noted that these mechanisms are not entirely independent concepts of each other. Typically, the dependent scattering mechanisms are treated in a renormalized way, i.e., by correcting conventional radiative parameters into effective parameters to include the dependent scattering effects and retaining the form of RTE or the diffusion equation (an approximate form of RTE in optically thick, highly scattering random media) to solve the transport problem [10].

Up to now, most researches focus on the role of dependent scattering mechanism in scattering properties of random media, because its impact on scattering properties is more prominent and can result in attractive phenomena as mentioned above. In terms of its role in absorption, very few studies, to the best of our knowledge, exist [13,25–27]. Actually, when the particle density is small (typically volume fraction $f_v < 0.05$) and absorption coefficient is much larger than the scattering coefficient, i.e., $\mu_a \gg \mu_s$, this ignorance gives rise to no substantial discrepancies because multiple and dependent scattering is weak. However, when particle density continues to increase or μ_s is comparable with or much larger than μ_a, a careful consideration of dependent scattering effect on total absorption is necessary because the inter-particle interference of scattering waves may lead to a redistribution in particle absorption. This issue is becoming important as the recent growing interests in nanofluids, as well as other nanoparticle-based solar absorbers, which usually utilize plasmonic resonances of metallic particles to enhance solar absorption [28–33] and find their applications in concentrating solar power and direct-steam generation. In this situation, the scattering coefficient might be comparable with the absorption coefficient for plasmonic particles with diameter approaching 100 nm. Actually this feature is exploited by some authors because multiple scattering of light can enhance the path length of light in the medium and thus improve the absorption efficiency [31]. Note the "multiple scattering" in this context is under the framework of RTE, meaning that light *intensity* is scattered many times, without any considerations on the wave aspect of the coherent scattering problem. Here throughout the rest of this paper, we term "multiple scattering" as the multiple scattering of *electromagnetic waves* implicitly. In fact in this circumstance, the strong scattering strength can also lead to a strong modification of the local electromagnetic field to be different with the external incident field, as a manifestation of dependent scattering mechanism [13,25]. Wei et al.[27] recently investigated the effect of dependent scattering on the absorption coefficient for very dense nanofluids (f_v up to 0.74) containing very small metallic particles (radius $a = 15$ nm) by using several different dependent-scattering models as well as developing a modified quasicrystalline approximation (QCA) model. However, their model, as well as the model proposed by Prasher et al. [26] can only treat the particle absorption is much larger than its scattering, which is only valid for very small particles.

Another active field is optofluidics, a combination of micro/-nano fluidic and photonic technologies, which aims to achieve desirable optical properties by controlling the flow rate, viscosity, etc., of nanofluids in a reconfigurable and compact manner. This technique is promising in integrated photonics applications, like lasers, sensors etc., as well as photocatalysis, solar thermochemistry and solar desalination applications [34,35]. One of the most popular optofluidic systems is the colloidal nanoparticle system controlled by microfluid chips, which shows the necessity of understanding the interplay between multiple scattering and absorption [34,35]. In some researches on the structural coloration based on disordered photonic structures, predictions based on ISA interpreted the experimental results poorly, partly due to the extremely-high absorption predicted by ISA for the short-range ordered, densely packed nanostructures. This may smear out the reflectance peak indeed observed by the experiments [7]. An in-depth physical interpretation for the scattering and absorption mechanism will also be helpful for these applications.

Here we aim to theoretically and numerically explore the role of dependent scattering mechanism on light absorption in disordered media consisting of highly scattering scatterers. By making comparisons between the absorptance data calculated by ISA combined with RTE and the full-wave coupled dipole method (CDM), we find that deviations between these two methods increase as scatterer density in the media increases. The discrepancy further grows with the optical thickness of the random media. To quantitatively take dependent scattering effect into account, we also develop a theoretical model of the dependent-scattering corrected radiative properties, based on the path-integral diagrammatic technique and the quasi-crystalline approximation (QCA) in the multiple scattering theory. The model results in a more reasonable agreement with numerical simulations. This study can provide physical insights on the applications of nanofluids in solar energy concentration, vapor generation using localized-light-induced-heat and heat transfer enhancement [36]. Our results also have implications in the dipole–dipole interaction effect on absorption in other highly scattering random media.

2. The coupled dipole method

We consider a slab geometry containing randomly distributed metallic particles in air as the investigated disordered medium with side lengths of L_x and L_y, and a thickness of L_z as shown in Fig. 1. The incident light propagates along the z-direction. Here the treatment of the background matrix as air doesn't affect the essential physics of multiple and dependent scattering, and can be easily extended to any kind of realistic background medium, such as gelatin, water, etc. We further assume that the scatterers are not so small that a is larger than a few nanometers, which means that the bulk permittivity can be used to describe the small particles and nonlocal (or quantum) effect in permittivity can also be neglected [37]. Moreover, we are working at the long