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Supplementary Materials

Extinction and Independent Scattering Criterion for Clusters Of Spherical Particles Embedded in Absorbing Host Media

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Abstract: In practical applications, the independent scattering approximation (ISA) is widely used to analyze light transfer in nanoparticle systems. However, the traditional independent scattering criterion is obtained under the assumption that the host medium surrounding particles is nonabsorbing, and thus may be invalid in certain circumstances. In this work, to explore the applicability of the ISA for small particles in absorbing host media, we calculate the extinction efficiency of particle clusters by direct solutions of macroscopic Maxwell equations. Using the far-field and distance-independent definitions of extinction, the computational efficiency multi-sphere method is applied for particle clusters in absorbing host, and its accuracy is verified with the discrete dipole approximation method. It is well known that for small particles, the dependent scattering in transparent host always enhances the extinction of the cluster and the criterion for the ISA is nearly independent of the particle refractive index and particle size. We show, however, that when the host medium is absorbing, the dependent scattering between particles can lead to a decreased or even negative extinction, and thus the ISA criterion depends on the particle refractive index, size, and host medium absorption index. In this result, the generalized criteria for absorbing host media may differ significantly from the conventional ones for transparent host media. The results can provide guidance in solving problems related to light transfer in nanoparticle systems, particularly in the presence of absorption in the host medium.

Keywords: absorbing host medium; radiative properties; multi-sphere method; dependent scattering; independent scattering approximation

Figure S1 illustrates the nondimensional extinction cross section η_{ext} as a function of the ratio of average interparticle clearance to wavelength c^* for a_i = 500, 750 and 1000 nm, $m'_{p,i}$ = 1.38, 2.1216 and 3.0308 ($m'_{p,i}/m'_h$ = 0.911, 1.4 and 2.0), and m''_h = 0.0 and 0.001, respectively. The incident wavelength is λ = 4.0 μm. The blue region indicates that the ISA is satisfied for all the three $m'_{p,i}/m'_h$ considered. Two dashed lines corresponding to η_{ext} = 0.95 and 1.05, respectively, are also plotted for comparison. As shown, when a_i = 500, 750 and 1000 nm, even in the nonabsorbing host medium (m''_h = 0.0), the values of η_{ext} are affected by the particle refractive index. Moreover, it is found in Figures S1c and Figures S1f that the dependent scattering can lead to decreased extinction. Furthermore, the host medium absorption has a limited impact on the dependent scattering between particles for a_i = 750 and 1000 nm. On the other hand, compared to the small particles, it needs larger c^* to ensure the accuracy of the ISA for large particles.

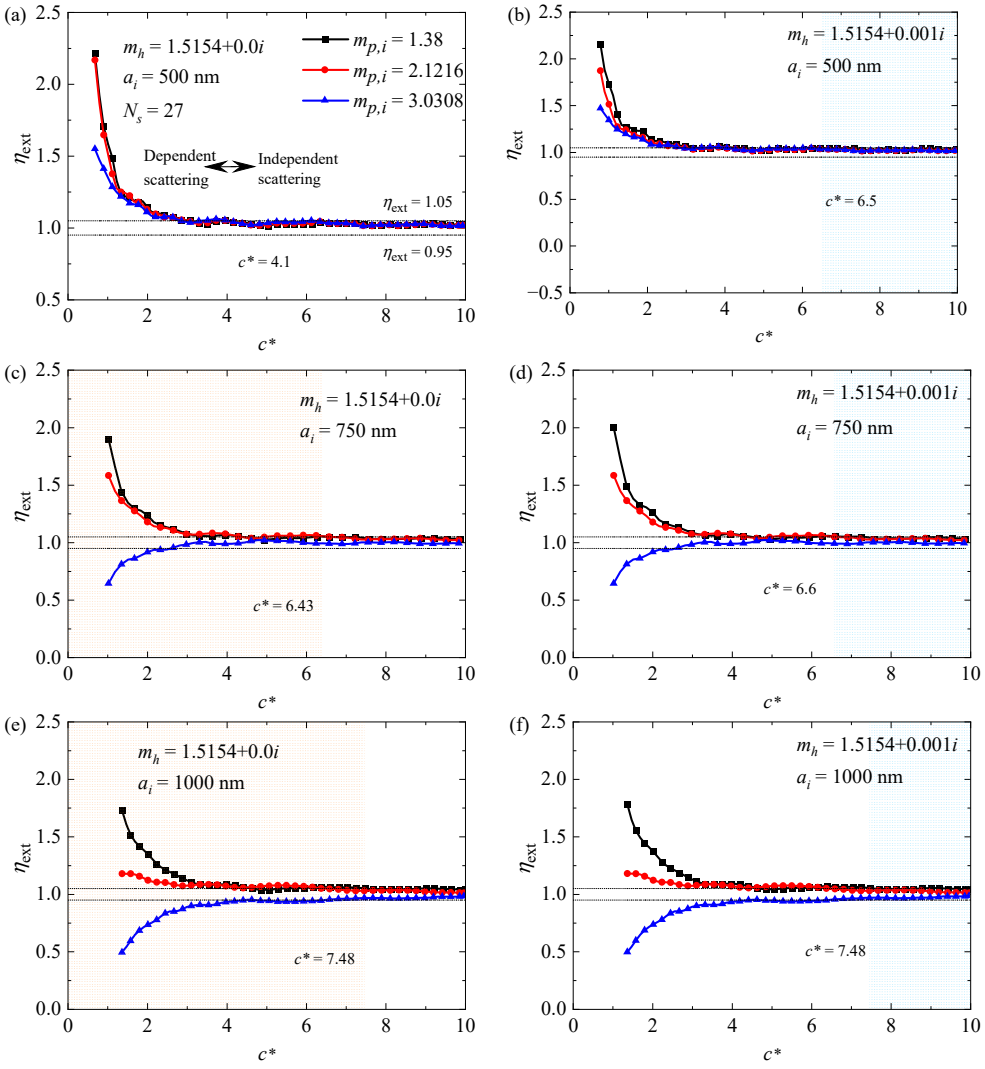

Figure S1. The nondimensional extinction cross section η_{ext} of 27-sphere cluster versus the ratio of average interparticle clearance to wavelength c^* with incident wavelength of λ = 4.0 μm, particle radius of a_i = 500, 750 and 1000 nm, and particle refractive index of $m'_{p,i}$ = 1.38, 2.1216 and 3.0308 ($m'_{p,i}/m'_h$ = 0.911, 1.4, and 2.0). The complex refractive index of the host medium is $m_h = 1.5154 + im''_h$ with m''_h = 0.0, and 0.001. (**a**) a_i = 500 nm, m''_h = 0.0, (**b**) a_i = 500 nm, m''_h = 0.001, (**c**) a_i = 750 nm, m''_h = 0.0, (**d**) a_i = 750 nm, m''_h = 0.001, (**e**) a_i = 1000 nm, m''_h = 0.0, and (**f**) a_i = 1000 nm, m''_h = 0.001. The blue region indicates that the ISA is satisfied for all the three $m'_{p,i}/m'_h$ considered. The results are averaged over 132 different orientations and 10 different configurations.