Direct demonstration of the concept of unrestricted effective-medium approximation

Michael I. Mishchenko,[1,*] Zhanna M. Dlugach,[2] and Nadezhda T. Zakharova[3]

[1]*NASA Goddard Institute for Space Studies, 2880 Broadway, New York, NY 10025, USA*

[2]*Main Astronomical Observatory of the National Academy of Sciences of Ukraine, 27 Zabolotny Str., 03680, Kyiv, Ukraine*

[3]*Trinnovim LLC, 2880 Broadway, New York, NY 10025, USA*

**Corresponding author: michael.i.mishchenko@nasa.gov*

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The modified unrestricted effective-medium refractive index is defined as one that yields accurate values of a representative set of far-field scattering characteristics (including the scattering matrix) for an object made of randomly heterogeneous materials. We validate the concept of the modified unrestricted effective-medium refractive index by comparing numerically exact superposition T-matrix results for a spherical host randomly filled with a large number of identical small inclusions and Lorenz–Mie results for a homogeneous spherical counterpart. A remarkable quantitative agreement between the superposition T-matrix and Lorenz–Mie scattering matrices over the entire range of scattering angles demonstrates unequivocally that the modified unrestricted effective-medium refractive index is a sound (albeit still phenomenological) concept provided that the size parameter of the inclusions is sufficiently small and their number is sufficiently large. Furthermore, it appears that in cases when the concept of the modified unrestricted effective-medium refractive index works, its actual value is close to that predicted by the Maxwell-Garnett mixing rule. © 2014 Optical Society of America

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For more than a century [1,2], the notion of an effective refractive index of a heterogeneous material has remained highly attractive and widely used [3]. Indeed, if it proves to be sufficiently accurate then it allows one to drastically simplify the calculation of scattering and absorption properties of small particles made of heterogeneous materials and thereby bypass the potentially daunting task of running direct computer solvers of the macroscopic Maxwell equations (MMEs) [4–6]. Especially significant is the notion of the unrestricted effective-medium approximation (UEMA) defined by Bohren [7] "as one that yields effective dielectric functions with the same range of validity as those of media that are usually taken to be homogeneous (e.g., pure water)." Despite the potentially great importance and utility of the UEMA, it has remained an unproven hypothesis: neither has it been derived directly from the MMEs in the form of a specific recipe nor has its very feasibility been established numerically. The direct analytical derivation of the UEMA by spatially averaging the solution of the time-domain MMEs appears to be highly problematic. Therefore, as explained below, the main objective of this Letter is to take advantage of the recent generalization of the superposition T-matrix method (STMM) and give the first, to our knowledge, quantitative validation of the UEMA hypothesis by invoking numerically exact computer solutions of the frequency-domain MMEs.

It is well known that both the time-domain MMEs and the derivative concept of the refractive index are an outcome of averaging the microscopic electromagnetic field over volume elements that are infinitesimally small compared to the wavelength and yet contain vast numbers of elementary charges [8–12]. The concept of an unrestricted effective refractive index (UERI) is an intuitively obvious extension wherein the electromagnetic field is averaged over volume elements that are significantly (but not infinitesimally) smaller than the wavelength and contain large (but not vast) numbers of elementary inhomogeneities. While the inhomogeneities are assumed to be sufficiently small, they are considered macroscopic, i.e., can be characterized by a refractive index that is different from that of the host material. Let us now assume that the resulting volume-averaged electromagnetic field satisfies the time-domain MMEs written for a homogeneous material characterized by a certain refractive index. Obviously, this artificial refractive index serves as the UERI of the heterogeneous material.

It is clear that for the UEMA to work in application to a heterogeneous target with a fixed distribution of inclusions, each volume element used for spatial averaging must contain a very large number of inclusions with sufficiently random (even though fixed) positions [8–12]. This requirement can be expected to be unnecessarily restrictive in many practical applications. To give the UEMA a better chance of success, it is appropriate to let the inclusions move and perform averaging over time t as well as over the elementary volumes.

It should be kept in mind, however, that averaging a time-harmonic electromagnetic field over t yields a zero net result owing to the rapidly oscillating complex exponential $\exp(-\mathrm{i}\omega t)$, where ω is the angular frequency and $\mathrm{i} = (-1)^{1/2}$ [13,14]:

$$\frac{1}{T}\int_t^{t+T} \mathrm{d}t' \exp(-\mathrm{i}\omega t') \underset{T\gg 2\pi/\omega}{=} 0. \tag{1}$$

Therefore, one must instead perform temporal averaging of a representative set of *optical observables* defined such that the complex exponential $\exp(-\mathrm{i}\omega t)$ becomes canceled out upon multiplication by its complex-conjugate counterpart [13,14]. Doing that can be further simplified by assuming statistical ergodicity of the

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random heterogeneous object and replacing temporal averaging with ensemble averaging, i.e., by averaging over a statistically random and uniform distribution of the positions of the inclusions [13,14]. This so-called modified UEMA (MUEMA) is declared to work if all the resulting ensemble-averaged optical observables coincide with those calculated by solving the MMEs for a homogeneous object with a certain value of the refractive index. The latter quantity is called the modified UERI (MUERI). A conceptually similar quantity was called the "equivalent refractive index" in Ref. [15].

In this Letter, we test the feasibility of the MUEMA by using the set of six far-field optical observables that form the real-valued normalized Stokes scattering matrix according to

$$\tilde{\mathbf{F}}(\Theta) = \begin{bmatrix} a_1(\Theta) & b_1(\Theta) & 0 & 0 \\ b_1(\Theta) & a_2(\Theta) & 0 & 0 \\ 0 & 0 & a_3(\Theta) & b_2(\Theta) \\ 0 & 0 & -b_2(\Theta) & a_4(\Theta) \end{bmatrix} \tag{2}$$

and serve to characterize the angular distribution and polarization state of the scattered light in the far zone of a random heterogeneous object [13,14]. In Eq. (2), $\Theta \in [0°, 180°]$ is the angle between the incidence and scattering directions, while the (1, 1) element is the conventional phase function normalized according to

$$\frac{1}{2}\int_0^{\pi} \mathrm{d}\Theta \sin\Theta a_1(\Theta) = 1. \tag{3}$$

The specific block-diagonal structure of the matrix (2) implies that the random scattering object is assumed to be statistically isotropic and mirror-symmetric.

For the purposes of our analysis, we model the heterogeneous object as a spherical particle randomly filled with N identical small spherical inclusions, as shown by the inset in Fig. 1(a). The size parameter of the spherical host $X = 2\pi R/\lambda$ is fixed at 10, where R is the particle radius and λ is the wavelength. The radius r of the inclusions is varied such that the inclusion size parameter

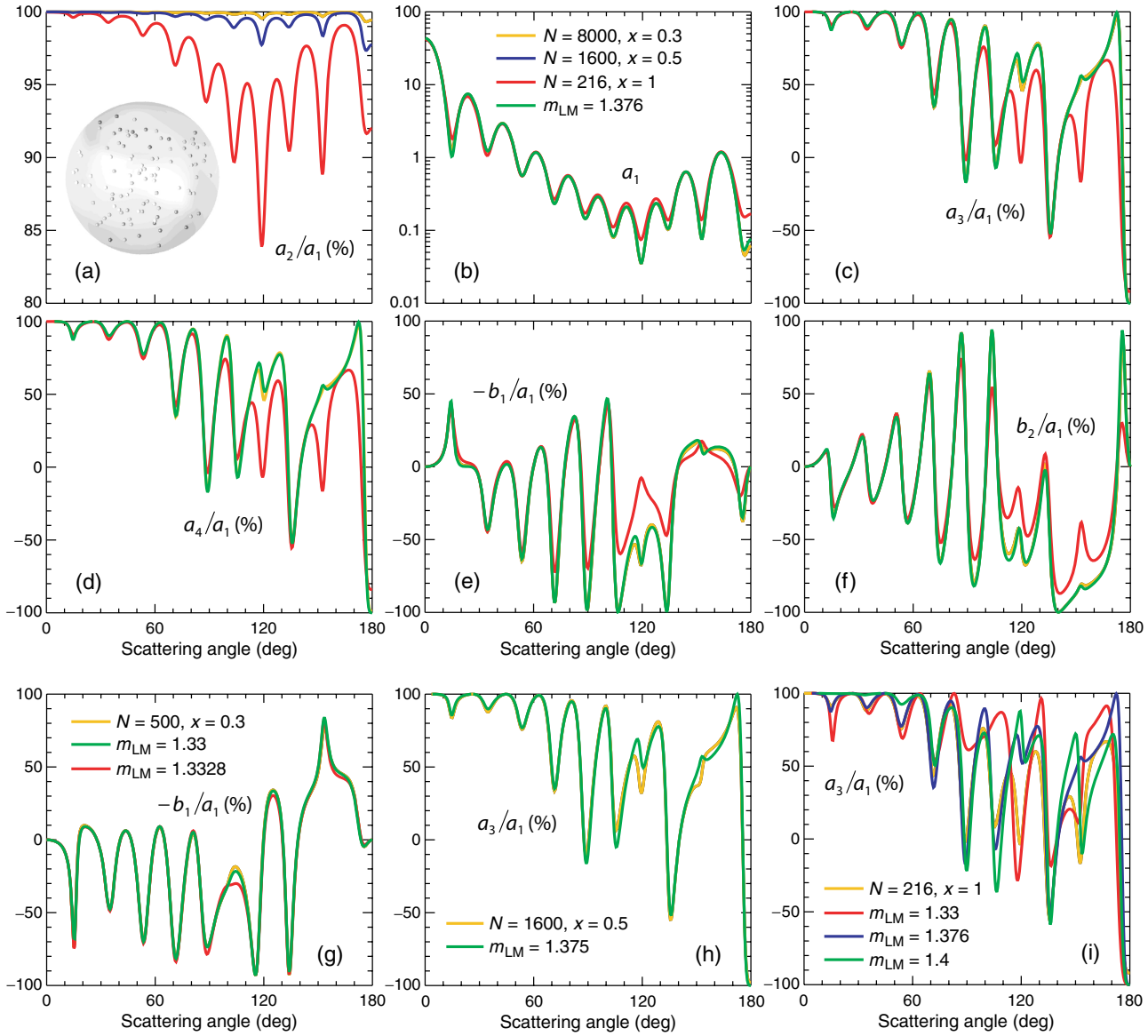

Fig. 1. Elements of the normalized scattering matrix (2) for randomly heterogeneous as well as homogeneous spherical particles with a fixed size parameter $X = 10$. The color legend in panel (b) applies to panels (a)–(f).

$x = 2\pi r/\lambda$ takes on values 0.3, 0.5, and 1. The refractive indices of the host and the inclusions are fixed at 1.33 and 1.55, respectively.

Densely packed inclusions separated by distances much shorter than the wavelength cannot be considered "independent" scatterers characterized by "individual" optical cross-sections and scattering matrices. This factor obviously disqualifies approximate approaches such as the ray tracing Monte Carlo technique [16]. Therefore, in this Letter the elements of the scattering matrix (2) for the heterogeneous object are calculated using a direct numerically exact computer solver of the MMEs, viz., the recently extended STMM, which is now applicable to arbitrarily clustered and nested spherical domains [17]. Unlike the numerical approach used in Ref. [15], the extended STMM is not based on a recursive scheme known to generate questionable results [18,19].

Following the methodology introduced in Refs. [20,21], the statistical randomness and uniformity of the object's interior is modeled in two steps. First, a random-number generator is used to create a fixed quasi-random and quasi-uniform configuration of the N inclusions while making sure that the volumes of the inclusions do not overlap and do not cross the host's boundary. Second, all far-zone optical observables are averaged over the uniform orientation distribution of the resulting heterogeneous object.

It is obvious that if the boundary of the host particle is perfectly spherical then any effective-medium approximation must satisfy the well-known Lorenz–Mie identity [22]

$$a_2(\Theta)/a_1(\Theta) \equiv 1. \tag{3}$$

Therefore, a deviation of the ratio $a_2(\Theta)/a_1(\Theta)$ from 100% is the most direct and unequivocal indicator of the numerical inaccuracy of the MUEMA. Figure 1(a) shows that the inclusion size parameter $x = 0.3$ yields $a_2(\Theta)/a_1(\Theta)$ values hardly distinguishable from 100%, whereas the inclusion size parameter $x = 1$ results in an obvious failure of the MUEMA. The size parameter $x = 0.5$ causes deviations of the ratio $a_2(\Theta)/a_1(\Theta)$ from 100% that are noticeable, but do not necessarily invalidate the MUEMA. Note that the volume density of the inclusions is approximately the same in all three cases: it is 21.6% for $x = 0.3$ and $x = 1$ and 20% for $x = 0.5$.

The remaining nonzero elements of the scattering matrix are depicted in panels (b)–(f) of Fig. 1. The green curves show the results of Lorenz–Mie computations for a homogeneous spherical particle with $X = 10$ that provide the best fit to the STMM results for a heterogeneous particle with $x = 0.3$ and $N = 8000$ shown by the yellow curves. The nearly perfect agreement provides a convincing validation of the MUEMA hypothesis. The corresponding best-fit Lorenz–Mie refractive index value is $m_{\mathrm{LM}} = 1.376$, which is almost identical to the value predicted by the well-known Maxwell-Garnett effective-medium rule [1]. However, the red curves in these panels demonstrate that this rule definitely fails in application to larger inclusions with $x = 1$. The latter conclusion is consistent with the results reported in Ref. [20].

In Fig. 1(g), the yellow curve shows the STMM results for $x = 0.3$ and $N = 500$. The best-fit Lorenz–Mie results are shown by the green curve and correspond to the refractive index $m_{\mathrm{LM}} = 1.33$. This best-fit value is different from the value $m_{\mathrm{LM}} = 1.3328$ predicted by the Maxwell-Garnett rule. The red curve computed for $m_{\mathrm{LM}} = 1.3328$ reveals a noticeably worse agreement with the STMM curve. This result may indicate that the number $N = 500$ of inclusions may not be sufficiently large to ensure the requisite quasi-homogeneity of the interior of the heterogeneous spherical particle at spatial scales significantly smaller than the wavelength.

Figure 1(h) compares the STMM results for $x = 0.5$ and $N = 1600$ with the best-fit Lorenz–Mie results obtained for $m_{\mathrm{LM}} = 1.375$. Although this best-fit refractive index is close to that predicted by the Maxwell-Garnett rule, the deviations of the green curve from the yellow one are noticeable and suggest that the very concept of the MUEMA becomes questionable at inclusion size parameters comparable to and exceeding 0.5. This conclusion is reinforced by Fig. 1(i) which demonstrates that the STMM results for $x = 1$ and $N = 216$ cannot be reproduced by Lorenz–Mie computations for any m_{LM} value.

In summary, the recently improved STMM [17] has finally provided a reliable direct means of testing the general idea of the modified UEMA. Instead of calculating integral radiometric observables such as the optical cross-sections, we based our analysis on the elements of the Stokes scattering matrix as far-field observables much more sensitive to microphysical properties of the scattering object. Our comparisons of numerically exact results computed for randomly heterogeneous and homogeneous spherical particles have shown unequivocally that the MUEMA is a sound approach provided that the size parameter of the inclusions is sufficiently small and their number is sufficiently large. The threshold values of x and N can be expected to depend on the refractive indices of the host and the inclusions as well as on the size parameter of the host and should be further analyzed and quantified. It is important, however, that the remarkable quantitative agreement between the yellow and green curves in Figs. 1(b)–1(f) over the entire range of scattering angles validates the general concept of the modified unrestricted effective refractive index. This Letter did not aim at deriving the MUEMA from the MMEs and formulating a specific prescription for the calculation of the MUERI based on the physical characteristics of the host object and the inclusions. Nevertheless, it appears from our limited numerical data that if the MUEMA is valid then the MUERI is likely to be close to that predicted by the Maxwell-Garnett rule.

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References

1. J. C. Maxwell-Garnett, Phil. Trans. R. Soc. A **203**, 385 (1904).

2. D. A. G. Bruggeman, Ann. Phys. **416**, 636 (1935).
3. P. Chýlek, G. Videen, D. J. W. Geldart, J. S. Dobbie, and H. C. W. Tso, in *Light Scattering by Nonspherical Particles: Theory, Measurements, and Applications*, M. I. Mishchenko, J. W. Hovenier, and L. D. Travis, eds. (Academic, 2000), p. 273.
4. M. I. Mishchenko, J. W. Hovenier, W. J. Wiscombe, and L. D. Travis, in *Light Scattering by Nonspherical Particles: Theory, Measurements, and Applications*, M. I. Mishchenko, J. W. Hovenier, and L. D. Travis, eds. (Academic, 2000), p. 29.
5. F. M. Kahnert, J. Quant. Spectrosc. Radiat. Transfer **79–80**, 775 (2003).
6. A. Doicu, T. Wriedt, and Y. A. Eremin, *Light Scattering by Systems of Particles* (Springer, 2006).
7. C. F. Bohren, J. Atmos. Sci. **43**, 468 (1986).
8. H. A. Lorentz, *The Theory of Electrons* (B. G. Teubner, Leipzig, 1916).
9. L. Rosenfeld, *Theory of Electrons* (North-Holland, 1951).
10. G. Russakoff, Am. J. Phys. **38**, 1188 (1970).
11. J. Vlieger, Can. J. Phys. **49**, 1384 (1971).
12. J. D. Jackson, *Classical Electrodynamics* (Wiley, 1999).
13. M. I. Mishchenko, L. D. Travis, and A. A. Lacis, *Multiple Scattering of Light by Particles: Radiative Transfer and Coherent Backscattering* (Cambridge University, 2006).
14. M. I. Mishchenko, *Electromagnetic Scattering by Particles and Particle groups: An Introduction* (Cambridge University, 2014).
15. A. Doicu and T. Wriedt, J. Opt. A Pure Appl. Opt. **3**, 204 (2001).
16. M. I. Mishchenko and A. Macke, J. Quant. Spectrosc. Radiat. Transfer **57**, 767 (1997).
17. D. W. Mackowski, J. Quant. Spectrosc. Radiat. Transfer **133**, 264 (2014).
18. P. R. Siqueira and K. Sarabandi, IEEE Trans. Antennas Propag. **48**, 317 (2000).
19. C. Meiners, J. Psilopoulos, and A. F. Jacob, Microwave Opt. Technol. Lett. **40**, 523 (2004).
20. M. I. Mishchenko, L. Liu, and D. W. Mackowski, Opt. Lett. **39**, 1701 (2014).
21. M. I. Mishchenko, L. Liu, B. Cairns, and D. W. Mackowski, Opt. Lett. **39**, 2607 (2014).
22. H. C. van de Hulst, *Light Scattering by Small Particles* (Wiley, 1957).