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Non-local constitutive relations and the quasi-homogeneous approximation

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Abstract

The scattering from bodies exhibiting a non-local dielectric response is investigated within the weak scattering limit. A quasi-homogeneous approximation is introduced and investigated. A qualitative change in the scattering pattern of scattered radiation is seen to emerge with the onset of a non-local response.

1. Introduction

Theoretical investigations of the scattering of electromagnetic fields from condensed matter media often are carried out on the basis of macroscopic electrodynamics. In the macroscopic approach, local-field effects are neglected, and for non-magnetic media the linear electromagnetic response may be characterized by the local dielectric tensor $\bar{\epsilon}(\mathbf{r}, \omega)$, a complex quantity which, as indicated, can depend on position and angular frequency. In certain cases, (e.g., optical activity, exciton resonances, anomalous skin effects, superconductivity) it is necessary to go beyond the macroscopic framework and include local-field phenomena. The dielectric tensor now becomes a two-point quantity $\bar{\epsilon}(\mathbf{r}, \mathbf{r}'; \omega)$ relating the microscopic dielectric displacement field $\mathbf{D}$ to the prevailing electric field $\mathbf{E}$ in a spatially non-local manner, i.e.,

$$\mathbf{D}(\mathbf{r}, \omega) = \epsilon_0 \int \mathrm{d}^3 r \, \bar{\epsilon}(\mathbf{r}, \mathbf{r}'; \omega) \mathbf{E}(\mathbf{r}', \omega). \tag{1}$$

To determine $\bar{\epsilon}(\mathbf{r}, \mathbf{r}'; \omega)$, quantum mechanical calculations, based, for example, on the density matrix formalism, usually have to be carried out. If the medium under study can be assumed to possess translational invariance in space, then the dielectric tensor can only

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depend on the difference between $\mathbf{r}$ and $\mathbf{r}'$; that is $\bar{\epsilon}(\mathbf{r},\mathbf{r}';\omega)=\bar{\epsilon}(\mathbf{r}-\mathbf{r}';\omega)$. With this simplification, the constitutive relation in Eq. (1) takes the local form

$$\mathbf{D}(\mathbf{k},\omega)=\epsilon_0\bar{\epsilon}(\mathbf{k},\omega)\mathbf{E}(\mathbf{k},\omega) \tag{2}$$

in the wave-vector domain. Media exhibiting a non-local response of the simple form given in Eq. (2) are called spatially dispersive and these media can support the propagation of monochromatic plane waves.

It is clear that the translational invariance criterion is broken near surfaces and interfaces, and if surface/interface phenomena are important, the reduction from Eq. (1) to Eq. (2) most often cannot be employed. There are some situations where the reduction is meaningful even in the presence of surfaces, thus, in a free electron-like metallic object, Eq. (2) is an adequate starting point for the analysis provided (i) the potential barrier at the surface can be considered to be infinitely high (no photoemission), and (ii) the conduction electrons are scattered specularly on the surface.

In the context of electromagnetic wave scattering, the spatial dispersion phenomenon was investigated in a paper published by Pekar in 1958 [1]. Pekar noted that spatial dispersion might give rise to the existence of propagating longitudinal electromagnetic waves. The excitation of a longitudinal field mode complicates the solution of the electromagnetic boundary problem somewhat. Many investigated this problem, citing an apparent mismatch between the degrees of freedom in the problem and boundary conditions to constrain them [2–4]. Ostensibly, the problem with boundary conditions in spatially dispersive media arises as a result of the reduction from Eq. (1) to Eq. (2) also in the surface region. For objects of finite extent, the integro-differential equation for the electric field which comes out of the Maxwell equation with the constitutive relation in Eq. (1) does not transform in any simple way when going to momentum (wave-vector) space. For material with a strictly local response, the $\mathbf{E}$-field scattering is governed by a differential equation and relatively simple numerical methods are available for solving the scattering problem. When the *approximation* $\bar{\epsilon}(\mathbf{r},\mathbf{r}';\omega)=\bar{\epsilon}(\mathbf{r}-\mathbf{r}';\omega)$ is used right up to the boundary, it is not necessary to introduce *extra* boundary conditions as long as one carefully addresses certain mode-coupling relationships that are specific to the geometry of the problem [5]. In Ref. [5], a result for a semi-infinite slab geometry was derived that included reflection and transmission coefficients, as well the aforementioned mode-coupling conditions. The approach in Ref. [5] was applied to a spherical geometry, but a set of mode-coupling conditions that are specific to this geometry was not found [6].

The papers in Refs. [1–6] made use of an isotropic frequency and wave-number dependent dielectric function of the form

$$\epsilon(\omega)=\epsilon_0+\frac{4\pi\alpha\omega_0^2}{\omega_0^2-\omega^2-\mathrm{i}\Gamma\omega}. \tag{3}$$

By assuming that the non-local response emerges near a resonance in the exciton band at $\omega=\omega_e$ and that $k\simeq 0$, the effective mass approximation $\hbar\omega_0\approx\hbar\omega_e+\frac{(\hbar k)^2}{2m_e^*}$, may be made and yields the expression

$$\epsilon(\mathbf{k},\omega)=\epsilon_0+\frac{\chi}{k^2-\mu^2(\omega)}, \tag{4}$$

where $\chi=4\pi\alpha m_e^*\omega_e\hbar^{-1}$, and $\mu^2(\omega)=m_e^*(\omega^2-\omega_e^2+\imath\omega\Gamma)(\hbar\omega_e)^{-1}$. The constitutive relation is then

$$\mathbf{D}(\mathbf{k},\omega)=\epsilon_0\mathbf{E}(\mathbf{k},\omega)+\frac{\chi}{k^2-\mu^2(\omega)}\mathbf{E}(\mathbf{k},\omega). \tag{5}$$

By transformation to the spatial domain, it is seen that

$$\mathbf{D}(\mathbf{r},\omega)=\epsilon_0\mathbf{E}(\mathbf{r},\omega)+4\pi\epsilon_0\int \mathrm{d}^3r'\,\eta(\mathbf{r},\mathbf{r}',\omega)\mathbf{E}(\mathbf{r}',\omega), \tag{6}$$

where the integral is taken over all space and

$$\eta(\mathbf{r},\mathbf{r}')=\frac{\chi}{4\pi\epsilon_0}\frac{\mathrm{e}^{\mathrm{i}|\mathbf{r}-\mathbf{r}'|\mu}}{|\mathbf{r}-\mathbf{r}'|}. \tag{7}$$

It may be seen immediately that the susceptibility given in Eq. (7) describes an object infinite in extent. This basic problem may be addressed by simply demanding that a support constraint be imposed in the spatial domain. Of course this leads to a modification of the dispersion relation and calls into question the basic approach to deriving the particular form in Eq. (7). It may also be noted that this susceptibility implies that the material constituents exhibit very long range quantum correlations and so seems unreasonable on these grounds. One might expect that even near an exciton resonance, long range correlations would be limited by thermal decorrelation over length scales determined by the temperature.

Rather than considering the problem first in momentum space and then trying to fix the boundary conditions, one may consider the spatial domain to be of the fundamental importance. Thus, in this Letter, Eq. (6) will be taken as a starting point. A model for $\eta(\mathbf{r},\mathbf{r}')$ will be considered that is consistent with an object of finite extent and exhibits more rapid fall-off of the non-local response in the bulk material. The effects of an emergent non-local response on the radiation pattern of light scattered from such an object will be investigated. An approximation, valid as the length-scale of the non-local response becomes small compared to the object size, and that makes calculation of the scattered field for arbitrary objects more tractable, is introduced.

2. Scattering model

Maxwell's equations together with Eq. (6) yield an integro-differential wave equation,

$$\nabla\times\nabla\times\mathbf{E}-k_0^2\mathbf{E}=4\pi k_0^2\int\limits_V \mathrm{d}^3r'\,\eta(\mathbf{r},\mathbf{r}')\mathbf{E}(\mathbf{r}'), \tag{8}$$

where $k_0^2=\omega^2\mu_0\epsilon_0$. The material is chosen to possess a bulk non-local response that is Gaussian in form and homogeneously distributed throughout a sphere of radius b so that

$$\eta(\mathbf{r},\mathbf{r}')=\frac{\eta_0}{(2\pi)^{\frac{3}{2}}\sigma^3}\exp\left(-\frac{|\mathbf{r}-\mathbf{r}'|^2}{2\sigma^2}\right)B(\mathbf{r})B(\mathbf{r}'), \tag{9}$$

where

$$B(\mathbf{r})=\begin{cases}1, & \mathbf{r}\in V,\\ 0, & \mathbf{r}\notin V,\end{cases} \tag{10}$$

describes the distribution of material in the scatterer. The scattered field is determined through the normal method of Green's dyadics. By writing the total electric field as

$$\mathbf{E}(\mathbf{r})=\mathbf{E}^{(i)}(\mathbf{r})+\mathbf{E}^{(s)}(\mathbf{r}) \tag{11}$$

and inserting this into Eq. (8), the scattered field is seen to be given by the expression

$$\mathbf{E}^{(s)}(\mathbf{r})=k_0^2\int\limits_V \mathrm{d}^3r''\,\bar{G}_0(\mathbf{r},\mathbf{r}'')\int\limits_V \mathrm{d}^3r'\,\eta(\mathbf{r}'',\mathbf{r}')\mathbf{E}(\mathbf{r}'). \tag{12}$$

This field necessarily satisfies the radiation condition and the boundary conditions, continuity of the tangential components of the electric and magnetic fields. For objects of finite extent, the integral equation (12) has no known exact analytic solutions even for cases of high symmetry. A series solution for small values of the scattered field (the Born series) may be obtained by standard perturbative methods. To first order, the total field in Eq. (12) is replaced by the incident field. In the far-field the dyadic Green's function takes the asymptotic form [7],

$$\bar{G}_0(\mathbf{r},\mathbf{r}')\sim(\bar{\mathrm{I}}-\hat{\mathrm{r}}\hat{\mathrm{r}})\frac{\mathrm{e}^{\mathrm{i}k_0r}}{r}\mathrm{e}^{-\mathrm{i}k_0\hat{\mathrm{r}}\cdot\mathbf{r}'}, \tag{13}$$

where $\hat{\mathrm{r}}$ is the unit vector pointing in the $\mathbf{r}$ direction. The incident field may be decomposed into plane waves and so without loss of generality we consider the incident field to be an arbitrary plane wave,

$$\mathbf{E}^{(i)}(\mathbf{r})=\hat{\mathrm{a}}\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{r}}. \tag{14}$$

Thus Eq. (12) approximately reduces to the form

$$\mathbf{E}^{(s)}(\mathbf{r})=k_0^2(\bar{\mathrm{I}}-\hat{\mathrm{r}}\hat{\mathrm{r}})\hat{\mathrm{a}}\frac{\mathrm{e}^{\mathrm{i}k_0r}}{r}\int\limits_V \mathrm{d}^3r''\,\mathrm{e}^{-\mathrm{i}k_0\hat{\mathrm{r}}\cdot\mathbf{r}''} \times\int\limits_V \mathrm{d}^3r'\,\eta(\mathbf{r}'',\mathbf{r}')\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{r}'}, \tag{15}$$

which may be rewritten

$$\mathbf{E}^{(s)}(\mathbf{r})=k_0^2\big(\hat{\mathrm{a}}-\hat{\mathrm{r}}(\hat{\mathrm{r}}\cdot\hat{\mathrm{a}})\big)\frac{\mathrm{e}^{\mathrm{i}k_0r}}{r}\tilde{\eta}(-k_0\hat{\mathrm{r}},\mathbf{k}), \tag{16}$$

where $\tilde{\eta}(\mathbf{k}_1,\mathbf{k}_2)$ is the double Fourier transform of $\eta(\mathbf{r}_1,\mathbf{r}_2)$. For the choice of η in this Letter, the Fourier transform cannot be computed analytically due to the truncation of the Gaussian by the blocking functions. The response was simulated for a variety of values of σ and b.

As can be seen above, the change in the size parameter results in a marked change in the scattering profile. Beginning with the local response (a) and proceeding to cases that exhibit increasing non-locality (b)–(d), Fig. 1 shows an appreciable amount of back scattering as the size parameter increases. Also, as the size parameter becomes on the order of the radius of the sphere, oblique scattering emerges. This is a significant change from the forward scattering normally caused by spheres of this size with local response.

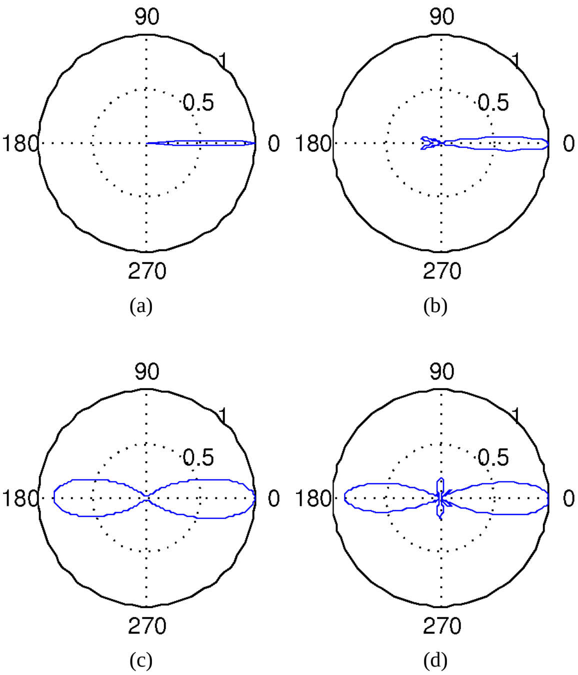

Fig. 1. Normalized scattering by a sphere of radius $b = 10\lambda$ and size parameter (a) $\sigma = 0$, (b) $\sigma = 1\lambda$, (c) $\sigma = 3\lambda$, (d) $\sigma = 6\lambda$. Note: forward scattering ($\theta = 0$) is normalized to 1.

3. Quasi-homogeneous approximation

The non-local response may be written

$$\eta(\mathbf{r}, \mathbf{r}') = \eta_\delta(\mathbf{r} - \mathbf{r}')S(\mathbf{r}, \mathbf{r}'), \tag{17}$$

where

$$\eta_\delta(\mathbf{r}, \mathbf{r}') = \frac{\eta_0}{(2\pi)^{\frac{3}{2}}\sigma^3} \exp\left(-\frac{|\mathbf{r} - \mathbf{r}'|^2}{2\sigma^2}\right) \tag{18}$$

and

$$S(\mathbf{r}, \mathbf{r}') = B(\mathbf{r})B(\mathbf{r}'). \tag{19}$$

It can be seen that the susceptibility is in part due to the form of the non-locality and in part due to the density of non-local material. If $B(\mathbf{r})$ is a broad function of its arguments compared to η_δ, then it can be replaced with

$$B(\mathbf{r}) \approx B(\mathbf{r}') \approx B\left(\frac{|\mathbf{r} + \mathbf{r}'|}{2}\right). \tag{20}$$

Then,

$$S(\mathbf{r}, \mathbf{r}') = B\left(\frac{|\mathbf{r} + \mathbf{r}'|}{2}\right) \equiv S_Q\left(\frac{|\mathbf{r} + \mathbf{r}'|}{2}\right). \tag{21}$$

This gives a quasi-homogeneous approximation

$$\eta(\mathbf{r}, \mathbf{r}') = \eta_\delta(\mathbf{r} - \mathbf{r}')S_Q\left(\frac{\mathbf{r} + \mathbf{r}'}{2}\right). \tag{22}$$

As shown in Eq. (16), the scattering amplitude for the non-local scatterer is proportional to the double Fourier transform of $\eta(\mathbf{r}, \mathbf{r}')$. Under the quasi-homogeneous approximation,

$$\tilde{\eta}(-k_0\hat{\mathrm{r}}, \mathbf{k}) = \tilde{\eta}_\delta\left(\frac{\mathbf{k} + k_0\hat{\mathrm{r}}}{2}\right)\tilde{S}_Q(\mathbf{k} - k_0\hat{\mathrm{r}}). \tag{23}$$

This result is general to any $\eta(\mathbf{r}, \mathbf{r}')$ that can be factorized or approximated by Eq. (22). Finding the scattering amplitude to lowest order reduces to the calculation of two Fourier transforms.

In Fig. 2, the results of three simulations are shown. In (a)–(c) the numeric result calculated with the exact form of the susceptibility, Eq. (9), is shown. In (d)–(f), the quasi-homogeneous approximation is made. The non-local susceptibility, simulated with either method, gives rise to a significant increase in scattering in the backward direction as the non-locality increases. The quasi-homogeneous result compares favorably with the numeric result in terms of predicting the distribution of back-scattered light. However, the quasi-homogeneous result predicts the amplitude of the back-scattering to be about three times larger than the numerical result in the final simulation (c), (f). The range of validity for the quasi-homogeneous approximation corresponds approximately to the local limit: the size parameter is very small and the sphere size is large in relation [8]. Thus, as the non-locality becomes more pronounced, the quasi-homogeneous approximation breaks down.

4. Concluding remarks

In cases where a local, macroscopic model of an object is insufficient to describe its interaction of the material with an applied electromagnetic field, a non-local model for the constitutive relations is necessary. This mesoscopic model takes into account quantum correlations within the material. When the object is of

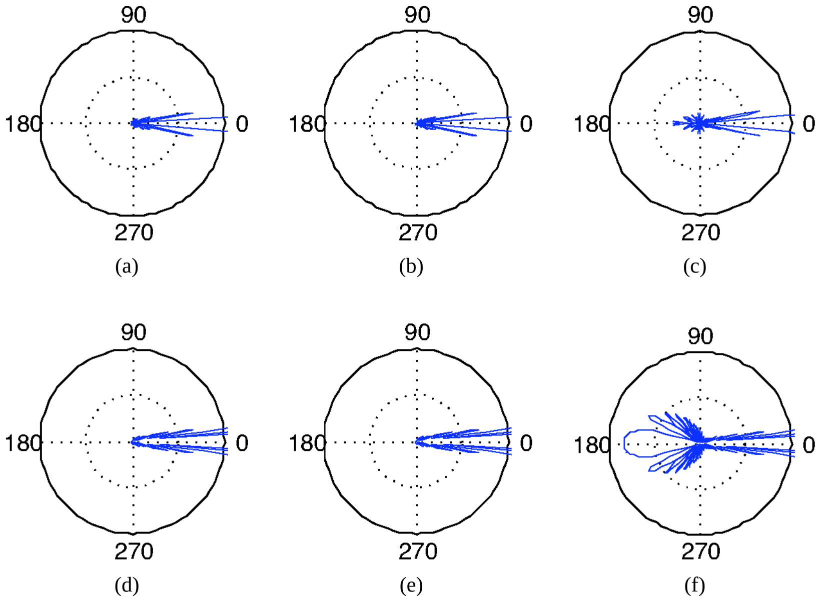

Fig. 2. Normalized scattering by a sphere of radius $b = 8\lambda$ for (a) the numeric result for $\sigma = 0.005\lambda$; (b) the numeric result for $\sigma = 0.05\lambda$; (c) the numeric result for $\sigma = 0.5\lambda$; (d) the quasi-homogeneous approximation for $\sigma = 0.005\lambda$; (e) the quasi-homogeneous approximation for $\sigma = 0.05\lambda$; (f) the quasi-homogeneous approximation for $\sigma = 0.5\lambda$. Note: forward scattering ($\theta = 0$) is normalized to 1. In this close-up view of the back-scattering, the outer ring represents $r = 0.05$ on the normalized scale.

finite extent, or boundary effects are important, a momentum space description of the interaction by a spatial dispersion relation is unsatisfactory. Rather, a coordinate space integral equation describes the interaction, including the boundaries. As has been shown, the scattering by an object with non-local response can be significantly different from scattering in the local response limit. In cases where the object exhibits a weak non-local response over a relatively large object, the quasi-homogeneous approximation is a valid substitute for determining the scattered field.

Both the quasi-homogeneous model and direct numerical integration predict emergent back-scattering as the size parameter of the non-locality becomes comparable to the resolvable features in the object. This increased back-scattering is then a possible indicator of non-local response. Either method can then be used in formulating the inverse scattering problem. For objects that are known to exhibit non-locality, the inverse problem is inherently underdetermined and is similar to the problem of determining the two point correlation function of random media [9]. If, a priori, $S(\mathbf{r}, \mathbf{r}')$ is known, then one can use inversion techniques to determine the form of the non-local response function $\eta_\delta(\mathbf{r} - \mathbf{r}')$.

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