Comment II on "Resonant and antiresonant frequency dependence of the effective parameters of metamaterials"

A. L. Efros*
University of Utah, Salt Lake City, Utah 84112, USA
(Received 2 February 2004; published 29 October 2004)

In a recent paper, Koshny, Markos, Smith, and Soukoulis [Phys. Rev E **68**, 065602 (2003)] report that their computations show that the product of imaginary parts of electric permittivity and magnetic permeability in a passive media is negative. They also criticize a well-known theorem that both imaginary parts are positive as a result of the second law of thermodynamics. I argue that this criticism has no ground and that computational evidence may result from inadequate introduction of the very concept of $\epsilon(\omega)$ and $\mu(\omega)$ in a photonic crystal.

DOI: 10.1103/PhysRevE.70.048602 PACS number(s): 41.20.Jb, 42.70.Qs, 73.20.Mf

In a recent paper, Koschny, Markos, Smith, and Soukoulis (KMSS) presented their numerical study of frequency-dependent permittivity $\epsilon(\omega)$ and permeability $\mu(\omega)$ of two different metamaterial elements which they propose for construction of materials providing negative refraction [1]. I disagree with this work on two important points.

(i) The computations of KMSS show that for one metamaterial there is a region with $\mathrm{Im}\epsilon(\omega)\ \mathrm{Im}\mu(\omega)<0$. Moreover, they refer to other papers where similar results have been obtained.

I think that the statement

$$\mathrm{Im}\epsilon(\omega) > 0,\ \mathrm{Im}\mu(\omega) > 0 \tag{1}$$

for any system in thermodynamic equilibrium (passive system) follows directly from the second law of thermodynamics and that this is one of the most important theorems of macroscopic electrodynamics. I will skip an early history of the question. In 1951, Callen and Welton [2] proved the fluctuation-dissipation theorem (FDT). Using this theorem, one can show that the imaginary part of any linear response is positive (see modern presentation in the course of Landau and Lifshitz [3]). An elegant derivation of Eq. (1) was given in 1957 by Landau and Lifshitz in the first edition of *Electrodynamics of Continuous Media* and it has been included with minor changes in the second edition by Landau, Lifshitz, and Pitaevskii (LLP) [4]. This derivation shows that Eq. (1) follows directly from the second law of thermodynamics. The derivation is as follows. Using Maxwell's equations, LLP get for dissipation Q of a monochromatic electromagnetic field

$$Q = \omega[\mathrm{Im}\epsilon(\omega)\langle E^2\rangle + \mathrm{Im}\mu(\omega)\langle H^2\rangle]/4\pi, \tag{2}$$

where $\langle\rangle$ means time averaging. Then they write, "On account of the law of increase of entropy, the sign of these losses is determinate: the dissipation of energy is accompanied by the evolution of heat, i.e., $Q>0$. It therefore follows that imaginary parts of ϵ and μ are always positive [Eq. (1)] for all substances and at all frequencies."

*Electronic address: efros@physics.utah.edu

KMSS are definitely familiar with this derivation, but they criticize it. "There are indeed no doubts that the imaginary part of the response function … must be positive when only one external force (electric or magnetic field) acts on the body. In the present case we analyze simultaneous response of both electric and magnetic field … and dissipated energy is given as a sum. The condition $Q>0$ does not require that $\mathrm{Im}\epsilon$ and $\mathrm{Im}\mu$ must be simultaneously positive."

I disagree with this criticism because the *linear* responses like ϵ and μ are the properties of a medium and they are independent of the configuration of electric and magnetic fields in the medium. For example, the FDT permits to obtain these responses by studying fluctuations in an equilibrium system without any external forces. I believe that KMSS considered only a case of a plane wave, propagating in the medium. In this case $\mathbf{H}=\sqrt{(\epsilon/\mu)}[\mathbf{n}\times\mathbf{E}]$ and indeed only one condition for $\mathrm{Im}\epsilon$ and $\mathrm{Im}\mu$ follows from $Q>0$. However, the condition $Q>0$ should be fulfilled for any configuration of fields in a given medium. Let us place the medium in a small but macroscopic capacitor. Then for a small enough distance between plates, the magnetic field, created by the displacement current, is negligible, and condition $Q>0$ demands $\mathrm{Im}\epsilon>0$. Now let us place the same medium in a small but macroscopic solenoid. Then the electric field, induced by the magnetic one, will be small [compare [4], argumentation before (79.4)] and dissipation will be positive only if $\mathrm{Im}\mu>0$. This is a simple interpretation of the LLP derivation. Thus, I believe that any violation of Eq. (1) contradicts the second law of thermodynamics. I am grateful to Professor Pitaevskii, one of the authors of Ref. [4], for detailed discussion of the text above.

(ii) Application of ϵ and μ to photonic crystal. This point is, probably, closely connected with the first one. KMSS write "Because the resonant frequency of a typical metamaterial element implies a free space wavelength much longer than the unit cell size, an effective medium approach has been applied that results in a characterization of metamaterials in terms of bulk ϵ and μ. The square root of the product $\epsilon\mu$ is the refractive index, which must be consistent with that determined from the dispersion diagram." I disagree with both sentences above. First, to introduce macroscopic $\epsilon(\omega)$ and $\mu(\omega)$, the wavelength *inside the material* must be much longer than the unit-cell size. Therefore, this approximation is not valid for photonic crystal if the quasi-wave-vector $\mathbf{k}$ is deep in the Brillouin zone or at the zone edge, as in the case considered by KMSS. Indeed, $\omega(k)$ as obtained from the equation

$$\omega^2 = \frac{c_0^2 k^2}{\epsilon(\omega)\mu(\omega)} \tag{3}$$

is an isotropic function of k^2 and it is not a periodic function of $\mathbf{k}$. Therefore, the only way to describe photonic crystal macroscopically is to introduce tensors $\epsilon_{\alpha\beta}(\omega,\mathbf{k})$ and $\mu_{\alpha\beta}(\omega,\mathbf{k})$, i.e., to take into account nonlocality. Note that in this case ϵ and μ are tensors even in a cubic crystal (see Ref. [4], Chap. 12). Following KMSS, one can introduce the "effective" refractive index $n_{\mathrm{ef}}(\mathbf{k})$ as $\omega(\mathbf{k})=c_0 k/n_{\mathrm{ef}}(\mathbf{k})$ using the dispersion diagram $\omega(\mathbf{k})$ as obtained from microscopic computations. However, this effective index is not "the square root of the product $\epsilon\mu$" since ϵ and μ are tensors. The macroscopic equations for $\omega(\mathbf{k})$ in this case are similar to the Fresnel equations in the optics of crystals. Thus, I think that Eq. (7) of the KMSS paper is not valid near the boundary of the Brillouin zone.

It is important to note that Veselago theory of the left-handed media is not valid in the case of the spatial dispersion, and it hardly can be generalized for this case. However, if the photonic crystal mode is nondegenerate at the Γ point of any Brillouin zone, Eq. (3) is valid in the vicinity of this point. Indeed, the microscopic field can be written in the form of a Bloch function $e_j=\exp(i\mathbf{k}\cdot\mathbf{r})U_{j,k}(\mathbf{r})$. If k is much smaller than the reciprocal vector of photonic crystal, the macroscopic field $E_j=\langle e_j\rangle=E_j^0\exp(i\mathbf{k}\cdot\mathbf{r})$, where $E_j^0=\langle U_{j,0}(\mathbf{r})\rangle$ and $\langle\rangle$ means averaging over the unit cell. Thus in a photonic crystal near the Γ point, a macroscopic field is a plane wave with a quasi-wave-vector of the Bloch function. In this case, ϵ and μ are k-independent and the dispersion law is given by Eq. (3). Obviously, this averaging procedure is impossible at large k. This indicates nonlocal macroscopic electrodynamics.

It has been recently shown [5,6] that if the group velocity of a mode is negative in the vicinity of the Γ point, there is a frequency interval where photonic crystal is the left-handed material of Veselago-type with negative $\epsilon(\omega)$ and $\mu(\omega)$. This is valid even if the material of the photonic crystal is a dielectric with $\mu=1$.

Finally, I think that Eq. (1) is correct, and that in the present formulation the results of Ref. [1] contradict the second law of thermodynamics. My second Comment may help to reformulate the computational results to avoid the contradiction, because a mathematical formulation of the increase of entropy due to electromagnetic losses in the case of spatial dispersion should have a more complicated form than Eq. (1).

[1] T. Koschny, P. Markos, D. R. Smith, and C. M. Soukoulis, Phys. Rev. E **68**, 065602 (2003).
[2] H. B. Callen and T. A. Welton, Phys. Rev. **83**, 34 (1951).
[3] L. D. Landau and E. M. Lifshitz, *Statistical Physics*, 3rd ed. (Butterworth Heinemann, Oxford, 1980), Pt. 1, p. 384.
[4] L. D. Landau, E. M. Lifshitz, and L. P. Pitaevskii, *Electrodynamics of Continuous Media* (Butterworth Heinemann, Oxford, 1984), p. 274.
[5] A. L. Pokrovsky and A. L. Efros, Solid State Commun. **124**, 283 (2002).
[6] A. L. Efros and A. L. Pokrovsky, Solid State Commun. **129**, 643 (2004).