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Examining the validity of Kramers-Kronig relations for the magnetic permeability

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We critically analyze the anomalies in the frequency dispersion of the magnetic permeability, showing that it may be sometimes—without contradicting causality—inconsistent with the Kramers-Kronig relations for passive materials, as formulated by Landau and Lifshitz, even at extremely low frequencies where the permeability has definitely a very precise physical meaning. This suggests that in general the permeability may not satisfy the Kramers-Kronig formulas for passive materials, and an alternative set of relations to link the real and imaginary parts of the permeability in the frequency region where the permeability retains its physical meaning is proposed.

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I. INTRODUCTION

One of the most exciting developments in electromagnetism in the last decade was the discovery of novel media with unusual electromagnetic properties mainly determined by the microstructure, and not directly by the chemical composition. In particular, it was shown that by structuring conventional bulk metals (with no intrinsic magnetism) it is possible to induce a strong magnetic response.[1] However, one of the most debated features of the permeability of metamaterials is that sometimes, even in the case of very low loss, the permeability may exhibit an antiresonant response, which is inconsistent with the Kramers-Kronig (KK) formulas for passive media (e.g., Refs. 2 and 3). Up to now, it has been believed that such pathology is invariably a consequence of the effects of spatial dispersion and of higher-order multipoles.[3,4] Here, we present examples of metamaterials with negligible spatial dispersion and such that the magnetic permeability is characterized by an anomalous dispersion (i.e., with an antiresonant response) for arbitrarily low frequencies, even when material absorption is vanishingly small. It is argued that these findings put into question, without contradicting the causal response of the materials, the application of the standard KK relations for passive media to the magnetic permeability.

II. PERMEABILITY WITH ANOMALOUS DISPERSION

In the first example, we consider a metamaterial formed by long rods standing in air and oriented along the z direction, arranged in a triangular (hexagonal) lattice with lattice constant a. We restrict our attention to the case where the direction of propagation and the electric field are in the xoy plane, so that, for simplicity, in this first example the geometry is intrinsically two dimensional. The rods have radius $R = 0.4a$, and a plasmonic-type electrical response characterized by a Drude dispersion model $\varepsilon_{\text{inc}} = 1 - \omega_p^2/\omega(\omega + i\Gamma)$ and $\mu = 1$ (the time variation $e^{-i\omega t}$ is assumed). The emergence of artificial magnetism in metamaterials formed by particles with a plasmonic-type response is well documented in the literature.[5–9] In particular, a metamaterial with a structure similar to the one that we consider here was studied in Ref. 6, and the coherent-potential approximation (CPA) was used to extract the effective parameters. In simple terms, the idea of the CPA is to embed a single cell of the metamaterial into a uniform isotropic unbounded medium with parameters ε_e and μ_e. If ε_e and μ_e are chosen in such a way that they are coincident with the effective parameters of the metamaterial, then the scattering from the metamaterial cell under plane wave incidence should vanish. Based on these simple and intuitive physical ideas, it is demonstrated in Ref. 6 that ε_e and μ_e can be obtained by numerically solving the pair of equations given by Eq. (2) of Ref. 6 with $m = 0, 1$. This ensures that the scattering from the electric and magnetic dipoles induced in the "unit cell" vanishes. For more details the reader is referred to Ref. 6.

In Fig. 1 we plot the effective permeability (along the z direction) calculated with the coherent-potential approximation in the limit of vanishingly small loss ($\Gamma = 0^+$) and for a plasma frequency such that $\omega_p a/c = 10.0$. We plot in the same figure the effective permeability calculated using the Clausius-Mossotti (CM) formula $\mu_{\text{CM}} = 1 + \frac{1}{A_{\text{cell}}}\frac{1}{\alpha_{m,zz}^{-1} - C_{\text{int},zz}}$, where $\alpha_{m,zz}$ is the magnetic polarizability of the rods (per unit of length),[10] and $C_{\text{int},zz} \approx (\frac{\omega}{c})^2[-i\frac{1}{4} + \frac{1}{2\pi}\ln(\frac{\omega}{c}\frac{\sqrt{A_{\text{cell}}}}{4\pi})]$ is an interaction constant.[11] The interaction constant takes into account the frequency dispersion. It should be mentioned that the interaction constant does not reduce to the usual value $C_{\text{int},xx} = C_{\text{int},yy} = 1/(2A_{\text{cell}})$ in the static limit, because in the two-dimensional case $C_{\text{int},zz} \neq C_{\text{int},xx}$ (Ref. 10).

Notwithstanding the absence of loss mechanisms, it is seen in Fig. 1 that both the CPA and the CM formula predict that the permeability decreases monotonically with the frequency, clearly violating the KK relations which, for passive materials, completely preclude any anomalous dispersion effects in such circumstances. In contrast with the results for the permeability, the dispersion of the in-plane permittivity of the material is completely consistent with the KK formulas (inset of Fig. 1).

The first reaction to these unsettling results is to argue that the CPA and the CM theories break down. To shed some light on this matter, we have calculated the effective permeability using the full wave method of Ref. 12, and the corresponding results are shown in Fig. 2. Specifically, the curves associated with the labels $\mu_{\text{ef}}^{(1)}$, $\mu_{\text{ef}}^{(2)}$, and $\mu_{\text{ef}}^{(3)}$ were obtained from the second-order derivatives of the nonlocal dielectric function with respect to the wave vector,[12] and the corresponding formulas are given in Ref. 13. As discussed in Ref. 13, in order that the material's response is effectively local and the effects of higher-order multipoles is negligible, it

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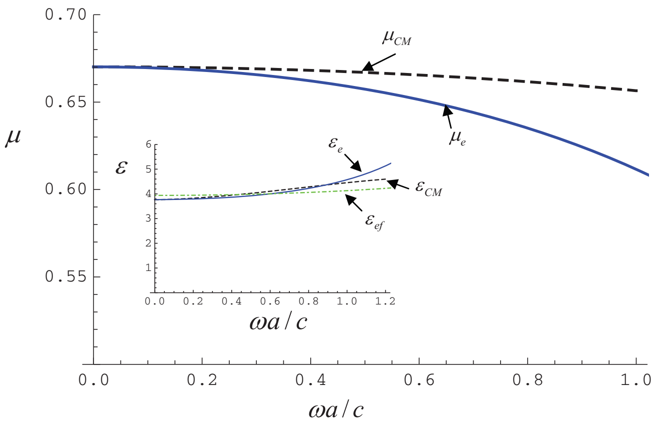

FIG. 1. (Color online) Effective permeability as a function of frequency in the limit of vanishingly small loss for an array of plasmonic-type particles arranged in a triangular lattice. Inset: effective permittivity as a function of frequency. μ_{CM} and $\varepsilon_{\mathrm{CM}}$ were obtained with the Clausius-Mossotti formula (taking into account the dispersion of the interaction constant), μ_e and ε_e were obtained using the CPA (see Ref. 6), and $\varepsilon_{\mathrm{ef}}$ using the full wave method of Ref. 13.

is necessary that to a good approximation, $\mu_{\mathrm{ef}}^{(1)} = \mu_{\mathrm{ef}}^{(2)} = \mu_{\mathrm{ef}}^{(3)}$. From Fig. 2 it can be seen that this condition is observed up to $\omega a/c \approx 1.0$. For larger frequencies, the values of$\mu_{\mathrm{ef}}^{(i)}$ ($i = 1,2,3$) may be significantly different from one another, indicating the presence of some spatial dispersion, particularly around $\omega a/c \approx 1.8$. Quite surprisingly, the results of the full wave simulations are very consistent with those of Fig. 1, and instead of showing that the CPA and CM theories break down, they further support that despite the loss being vanishingly small, the permeability really decreases with frequency. In particular, for low frequencies, the results obtained with the CM formula match well the curves $\mu_{\mathrm{ef}}^{(i)}$, as can be seen in Fig. 2.

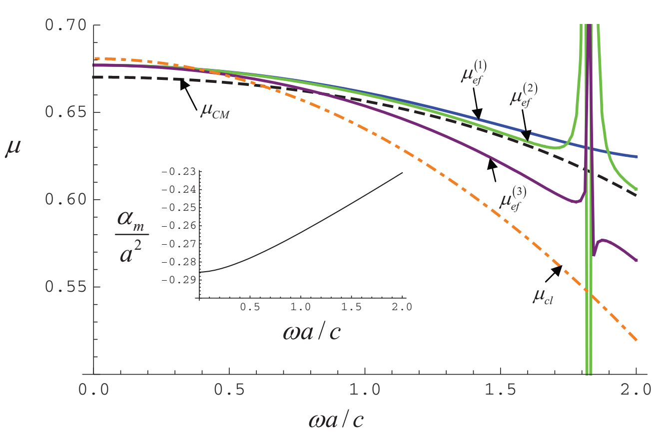

FIG. 2. (Color online) Effective permeability as a function of frequency in the limit of vanishingly small loss for an array of plasmonic-type particles arranged in a triangular lattice. μ_{CM} was obtained with the Clausius-Mossotti formula, $\mu_{\mathrm{ef}}^{(i)}$ using the full wave method of Ref. 13, and μ_{cl} using the classical definition of the magnetization vector (see the main text). Inset: magnetic polarizability of a single inclusion as a function of frequency in the limit of vanishingly small loss.

To further confirm that for $\omega a/c < 1.0$, $\mu_{\mathrm{ef}}^{(i)}$ may, indeed, characterize the magnetic response of the material, we have also calculated the permeability using the conventional definition of the magnetization vector: $\mathbf{M} = \frac{1}{A_{\mathrm{cell}}}\int_\Omega \frac{1}{2}\mathbf{r}\times\mathbf{j}_d dx\,dy$, where $\mathbf{j}_d = -i\omega\varepsilon_0(\varepsilon_{\mathrm{inc}}-1)\mathbf{e}$ represents the current density in the inclusions, Ω is the unit cell (of the triangular lattice), and $A_{\mathrm{cell}} = a^2\sin 60^\circ$. Specifically, in a numerical simulation the unbounded metamaterial was excited by an external distributed macroscopic current of the form $\mathbf{j}_{\mathrm{ext}} = \hat{\mathbf{y}}e^{ik_x x}$, such that k_x has a very small value,[14] and the corresponding induced microscopic electric field $\mathbf{e}$ was numerically calculated.[13] Then, consistent with the usual meaning of the magnetic permeability, we define $\mu_0\mu_{\mathrm{cl}} = B_z/(B_z\mu_0^{-1} - M_z)$, where $B_z = \frac{1}{A_{\mathrm{cell}}}\int_\Omega b_z e^{-ik_x x}dx\,dy$ is the average induction field. As seen in Fig. 2 the result of such calculation concurs very well with $\mu_{\mathrm{ef}}^{(i)}$ ($i = 1,2,3$) at low frequencies (most likely the small offset is a consequence of numerical errors), giving further evidence that $\mu_{\mathrm{ef}}^{(i)}$ may be regarded as the permeability for $\omega a/c < 1.0$, and indicating that theories based on the CPA and CM formulas may be physically sound.

How can we understand this surprising result? First of all, let us note that the anomalous dispersion of the permeability is necessarily a consequence of the lattice interactions between the inclusions. In fact, as shown in the inset of Fig. 2, $\mathrm{Re}\{\alpha_{m,zz}\}$ ($\mathrm{Re}\{\alpha_{m,zz}^{-1}\}$) is an increasing (decreasing) function of frequency, and thus if the interaction between the particles were negligible, the permeability would necessarily increase with frequency. Within the framework of the CM model, the anomalous dispersion effect is explained by the fact that the real part of the interaction constant, $C'_{\mathrm{int},zz}$, decreases with frequency, compensating for the normal dispersion of the magnetic polarizability, so that $\mathrm{Re}\{\alpha_{m,zz}^{-1}\} - C'_{\mathrm{int},zz}$ is an increasing function of frequency.

To study what happens in the presence of a stronger loss, in Fig. 3 we depict the calculated effective permeability $\mu = \mu' + i\mu''$ ($\mu_{\mathrm{ef}}^{(i)}$, and μ_{cl}) for the case where the collision frequency is $\Gamma = 0.01\omega_p$ (in real metals the collision frequency may be actually smaller, e.g., for silver $\Gamma \approx 0.002\omega_p$). As seen, the real part of the permeability μ' is still characterized

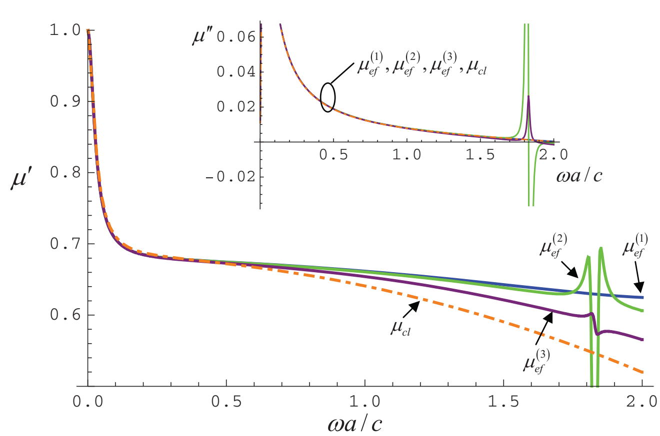

FIG. 3. (Color online) Real part of the effective permeability μ' as a function of frequency for an array of plasmonic-type particles arranged in a triangular lattice when $\Gamma = 0.01\omega_p$. Inset: imaginary part μ'' of the permeability.

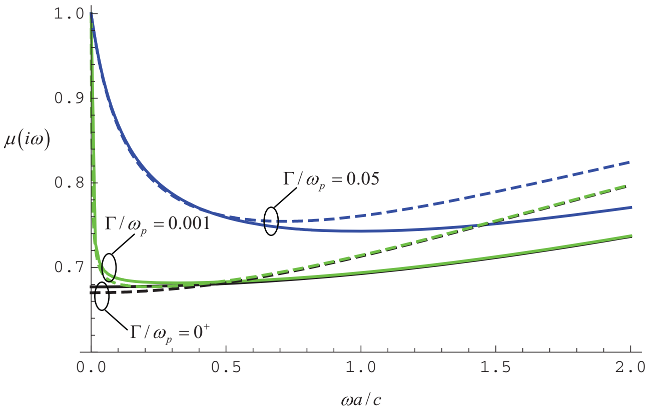

FIG. 4. (Color online) Effective permeability evaluated along the imaginary frequency axis for an array of plasmonic-type particles arranged in a triangular lattice and different values of the collision frequency. Solid lines: full wave method of Ref. 13; dashed lines: CPA (see Ref. 6).

by an anomalous dispersion, whereas the imaginary part μ'' is quite small, particularly in the band $0.5 < \omega a/c < 1.0$. The results of Fig. 3 are consistent with those reported in Ref. 6, but in that work neither the effect of anomalous dispersion nor the limit of vanishingly small loss was discussed. Notice that in the static limit the permeability is unity because the effect of absorption—no matter how small—precludes any magnetic effects in the static limit in metal-dielectric metamaterials.[15] It is worth noting that near $\omega = 0^+$ the permeability drops very steeply because the induction field is pushed away from the metal. For smaller values of loss the transition near $\omega = 0^+$ is even more abrupt, and in the limit of vanishingly small loss this transition occurs in an extremely narrow frequency band, and that is why it is not seen in the scale of the plots of Figs. 1 and 2.

We have also studied the dispersion of the magnetic permeability along the imaginary frequency axis. According to the so-called third Kramers-Kronig formula,[16] $\mu(i\omega)$ is completely determined by the imaginary part of the permeability, μ'', over the real frequency axis. In particular, for passive materials $\mu(i\omega)$ should be a strictly decreasing function of ω along the imaginary axis.[16] In Fig. 4 we plot $\mu(i\omega)$ along the imaginary axis for a metamaterial with the same geometry as in the previous examples and for different values of loss: $\Gamma/\omega_p = 0^+$, 0.001, 0.05. The solid lines correspond to full wave simulations[12,13] and the dashed lines were obtained using the CPA method.[6] As seen, in contradiction to the KK relations for passive materials, $\mu(i\omega)$ exhibits a nonmonotonic behavior and may increase with frequency. Very differently, the electric permittivity $\varepsilon(i\omega)$ is a strictly decreasing function of frequency (not shown).

As illustrated by the second example (described next), the anomalous dispersion may also be observed in the three-dimensional case. To illustrate this, we consider a body centered cubic (bcc) lattice of spherical particles with radius R. The unit cell of the material can be taken as a cube with side a with two particles per cell: one at the center of the cube and one-eighth of the other particle centered at each vertex of the cube. In Fig. 5 we depict the effective permittivity and permeability of the metamaterial calculated

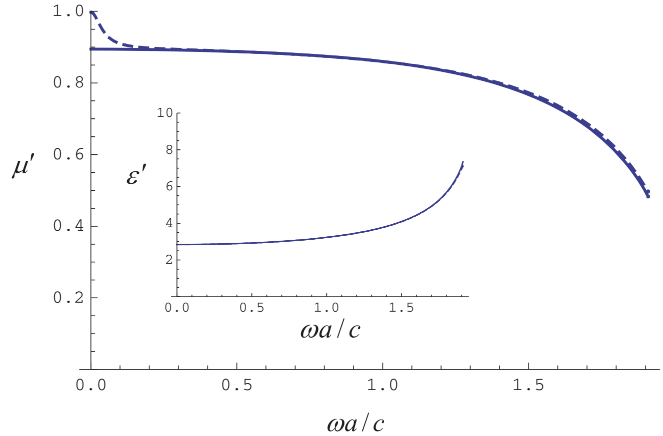

FIG. 5. (Color online) Real part of the effective permeability μ' as a function of frequency for an array of plasmonic spheres arranged in a bcc lattice. The plasma frequency is $\omega_p a/c = 6.0$ and the radius of the particles is $R = 0.35a$. Solid lines: limit of vanishingly small loss; dashed lines: $\Gamma = 0.01\omega_p$. Inset: real part of the effective permittivity ε' as a function of frequency.

using a time-domain implementation of the full wave method of Ref. 12, assuming that the radius of the spheres is $R = 0.35a$ and that $\omega_p a/c = 6.0$.[17] The real part of the permeability is completely consistent with the results of the two-dimensional case. As seen in Fig. 5, even in the limit of vanishing loss (solid line) the permeability decreases monotonically with the frequency. Absorption causes only a mild perturbation in the effective parameters (dashed line, associated with $\Gamma = 0.01\omega_p$). These results further support the possibility of having anomalous dispersion in the permeability, even in the lossless limit.

In general, the anomalous dispersion effects are more pronounced for larger particles and lower values of ω_p. For example, if the normalized plasma frequency is increased to $\omega_p a/c = 10.0$ the anomalous dispersion effects become somewhat weaker, as illustrated in Fig. 6 (solid blue line

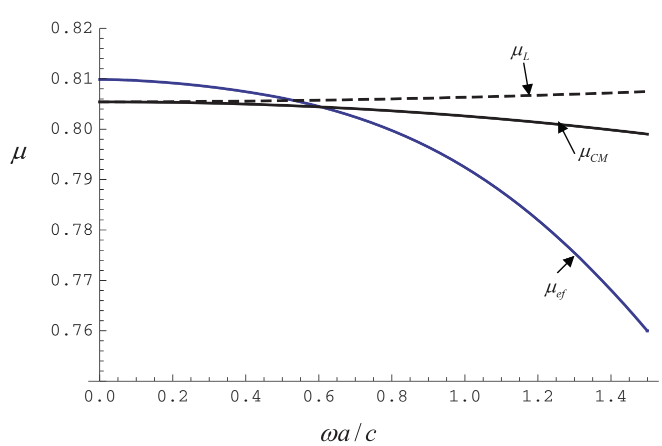

FIG. 6. (Color online) Effective permeability μ as a function of frequency for an array of plasmonic spheres arranged in a bcc lattice in the limit of vanishingly small loss. The plasma frequency is $\omega_p a/c = 10.0$ and the radius of the particles is $R = 0.35a$. Blue line ($\mu_{\rm ef}$): full wave result; solid black line ($\mu_{\rm CM}$): CM formula with dispersive interaction constant; dashed line (μ_L): Lewin's formula.

computed using the method of Ref. 12). It is interesting to mention that the anomalous dispersion effect identified here may be predicted by the Clausius-Mossotti formula

$$\mu_{\mathrm{CM}} = 1 + \frac{1}{V_{\mathrm{cell}}} \frac{1}{\alpha_m^{-1} - C_{\mathrm{int}}} \quad (1)$$

provided the frequency dispersion of the interaction constant is considered.[18,19] In fact, the magnetic polarizability α_m of spherical metallic particles with radius R, permeability $\mu_i = 1$, and permittivity ε_i can be estimated as (neglecting radiation loss)[20,21]

$$\alpha_m^{-1} = \frac{1}{4\pi R^3} \frac{F(\theta)+2}{F(\theta)-1}, \quad F(\theta) = \frac{2(\sin\theta - \theta\cos\theta)}{(\theta^2-1)\sin\theta + \theta\cos\theta}, \quad (2)$$

where $\theta = (\omega R/c)\sqrt{\varepsilon_i}$. The (real part of the) interaction constant C'_{int} is typically taken equal to $1/(3V_{\mathrm{cell}})$. However, the interaction constant is actually a function of frequency due to retardation effects and the finite speed of light.[18,19] Following Ref. 19, in the case of a bcc lattice we can estimate that $C'_{\mathrm{int}} \approx \frac{1}{V_{\mathrm{cell}}}[\frac{1}{3} - 0.1(\frac{\omega}{c}a)^2]$, where $V_{\mathrm{cell}} = a^3/2$ is the volume of the unit cell. The effective permeability calculated using the CM formula and frequency-dependent interaction constant is represented by the curve with label μ_{CM} in Fig. 6. Consistent with the full wave results, it is found that the permeability decreases with frequency. If one had neglected the frequency dependence of C_{int} and assumed $C'_{\mathrm{int}} \approx \frac{1}{3V_{\mathrm{cell}}}$, the permeability would be described by the curve with label μ_L in Fig. 6, which grows with frequency (such approximation is actually completely equivalent to the well-known Lewin's formula[20]). This further emphasizes that the anomalous dispersion effect is a consequence of the lattice interactions between the particles. It is also evident from Fig. 6 that the CM formula provides only a very rough estimate of the effective permeability, especially in the case of relatively large filling ratios.

It should be noted that in the continuum limit, supposing that $a \to 0$ (and assuming that the magnetic polarizability per unit of volume is kept invariant in this limit process), the interaction constant approaches the static value $C'_{\mathrm{int}} \approx \frac{1}{3V_{\mathrm{cell}}}$. Alternatively, if the size of the cell is small compared to the wavelength, the multipoles cannot depend on the time derivatives of the fields [22] (i.e., the relation between the local field and the average macroscopic field is instantaneous in time, and C'_{int} has no frequency dispersion). Thus, as long as the CM formula applies and provided α_m^{-1} satisfies the basic constraints of causality, in the continuum limit it is impossible to have anomalous dispersion effects in the limit of vanishing loss, and the KK formulas for passive media should apply. However, the continuous case is an idealization, and in practice all matter has some intrinsic granularity.

III. KRAMERS-KRONIG RELATIONS FOR PASSIVE MEDIA REVISITED

To explain the reason for the incompatibility between the results reported in the previous section and the KK relations for passive media, next we briefly revisit the derivation of the latter. The KK formulas establish a relation between the real and imaginary parts of an analytical function $\chi(\omega) = \chi'(\omega) + i\chi''(\omega)$. Provided $\chi(\omega)$ is regular both in the upper half plane and in the real axis, except possibly for a pole of order one at the origin, it follows from the application of Cauchy's theorem to a closed contour that contains the segment $-\omega_{\max} < \omega < \omega_{\max}$ of the real axis and a semicircumference C of radius $\omega_{\max}$ oriented counterclockwise in the upper half plane, that for ω real valued (below "P.V." stands for the principal value of the integral):

$$\chi(\omega) = \frac{1}{\omega}[\chi(x)x]_{x=0} + \frac{1}{\pi i}\mathrm{P.V.}\int_{-\omega_{\max}}^{\omega_{\max}} \frac{\chi(x)}{x-\omega}dx + \chi_C(\omega). \quad (3)$$

The first term in the right-hand side of Eq. (3) corresponds to the contribution of a possible pole of $\chi(\omega)$ at $\omega = 0$. Assuming that $\chi(\omega)$ represents the response of some physical system, it follows that $\chi(\omega) = \chi^*(-\omega)$ and the term $[\chi(x)x]_{x=0}$ is necessarily purely imaginary. The function $\chi_C(\omega) = \chi'_C(\omega) + i\chi''_C(\omega)$ represents the contribution of the semicircumference with radius $\omega_{\max}$ and is given by

$$\chi_C(\omega) = \frac{1}{\pi i}\int_C \frac{\chi(z)}{z-\omega}dz = \frac{1}{\pi}\int_0^{\pi} \frac{\chi(\omega_{\max}e^{i\theta})}{1 - e^{-i\theta}\omega/\omega_{\max}}d\theta. \quad (4)$$

Splitting the contributions of real and imaginary parts in Eq. (3), and neglecting the contribution of the term χ_C, we obtain

$$\chi'(\omega) = \frac{1}{\pi}\mathrm{P.V.}\int_{-\omega_{\max}}^{\omega_{\max}} \frac{\chi''(x)}{x-\omega}dx, \quad (5a)$$

$$\chi''(\omega) = \frac{1}{i\omega}[\chi(x)x]_{x=0} - \frac{1}{\pi}\mathrm{P.V.}\int_{-\omega_{\max}}^{\omega_{\max}} \frac{\chi'(x)}{x-\omega}dx. \quad (5b)$$

In the particular case $\omega_{\max} = +\infty$ these are the usual KK relations. The reason why usually we can drop the term χ_C is that the response of the system should vanish for large frequencies, and thus $\chi(\omega) \to 0$ as $\omega \to \infty$ (along any direction in the upper half plane). However, as discussed in Ref. 16 (p. 283), if one tries to apply these relations to $\chi = \mu - 1$, in general it is not possible to take $\omega_{\max} = +\infty$. Indeed, arguing that for large frequencies the magnetic response of the materials ceases, it is found from Eq. (5a) that $\mu'(\omega) - 1 = \frac{1}{\pi}\mathrm{P.V.}\int_{-\infty}^{+\infty}\frac{\mu''(x)}{x-\omega}dx$. However, for passive materials, described uniquely by a local permittivity and a local permeability, it is necessary that $\mu''(\omega) > 0$ for $\omega > 0$, because otherwise the medium could generate electromagnetic energy.[3,16,23,24] Thus, since $\mu''(\omega)/\omega > 0$ for arbitrary ω, it follows from the KK relations for passive media that $\mu(0) > 1$, i.e., if the KK formulas apply, then the material is necessarily paramagnetic in the static limit, as also discussed in a recent work.[25] This would contradict the existence of diamagnetic materials in the static limit. To avoid this absurd, in the book of Landau and Lifshitz (Ref. 16, p. 283), it is argued that because the magnetic permeability may lose its physical meaning at relatively low frequencies, the derivation of the KK formulas should be done by considering only the range of frequencies $|\omega| \leqslant \omega_{\max}$ in the upper half ω plane, where $\omega_{\max}$ is such that the permeability is essentially constant and real valued in the semicircumference $|\omega| = \omega_{\max}$ (we note here parenthetically that in case of metamaterials it is not necessarily true that the permeability loses its meaning at very small frequencies, as discussed in Ref. 26). In such conditions we can apply formulas (5) to the function $\chi = \mu - \mu_1$ with $\mu_1 = \mu(\omega_{\max})$, which yields the relation

$$\mu'(\omega) - \mu_1 = \frac{1}{\pi}\text{P.V.}\int_{-\omega_{\max}}^{+\omega_{\max}} \frac{\mu''(x)}{x-\omega}dx. \tag{6}$$

Since μ_1 can be an arbitrary real number such a procedure effectively solves the inconsistency related to the existence of diamagnetic materials in the static limit.

However, even the modified KK relations proposed by Landau and Lifshitz (which from hereafter are denoted by KK-LL) cannot explain the results of Figs. 1 and 2. Indeed, Eq. (6) predicts that in a transparency window where the effect of loss is vanishingly small [such as $\mu''(\omega) \approx 0$], we have[16]

$$\begin{aligned}\frac{d\mu}{d\omega} &= \frac{1}{\pi}\int_{-\omega_{\max}}^{+\omega_{\max}} \frac{\mu''(x)}{(x-\omega)^2}dx \\ &= \frac{4\omega}{\pi}\int_{0}^{+\omega_{\max}} \frac{\mu''(x)x}{(x^2-\omega^2)^2}dx > 0.\end{aligned} \tag{7}$$

Therefore, the permeability should be a strictly increasing function of frequency in a transparency window, in flagrant contradiction with Figs. 1, 2, and 4.

Similarly, the third Kramers-Kronig formula (in the Landau and Lifshitz framework) for the magnetic permeability,[16]

$$\mu(i\omega) - \mu_1 = \frac{2}{\pi}\int_{0}^{\omega_{\max}} \frac{x\mu''(x)}{x^2+\omega^2}dx. \tag{8}$$

implies that the permeability over the imaginary axis, $\mu(i\omega)$, is a strictly decreasing function of frequency, in disagreement with Fig. 4.

Let us critically analyze the arguments that led to the derivation of Eq. (6). As mentioned before, the Landau and Lifshitz argument is based on the assumption that $\mu(\omega_{\max}e^{i\theta}) \approx \mu_1 =$ const. for *any* angle $0 \leqslant \theta \leqslant \pi$. However, it is unlikely that in general the magnetic response may be effectively constant in the pertinent semicircumference. Indeed, it is expected that the value of the permeability will be larger for complex frequencies $\omega = \omega_{\max}e^{i\theta}$ that are closer to the resonances (poles) of the permeability. As is well known, in the case of low loss the poles of the permeability are very close to the real axis. Hence, we can conclude that unless $\omega_{\max} \gg \omega_{\text{res}}$, where ω_{res} is the largest resonant frequency of μ, the permeability cannot be considered constant in the semicircumference $\omega = \omega_{\max}e^{i\theta}$. For example, if the permeability has a pole close to $\omega_{\max}$ (which is the typical case in metamaterials), it seems quite likely that the value of $\mu(\omega_{\max}e^{i\theta}) - 1$ over the imaginary axis ($\theta = \pi/2$) has a much smaller amplitude than its value $\mu_1 - 1$ over the real axis. Therefore, we can estimate that $|\chi(\omega_{\max}e^{i\theta})| \sim |\mu_1 - 1|$ for values of θ not too close to either $\theta = 0$ or $\theta = \pi$, with $\chi = \mu - \mu_1$. On the other hand, the function $\chi_C(\omega)$ can be dropped in Eq. (3), if and only if it is much smaller than the first two terms in the right-hand side, or equivalently if $|\chi_C(\omega)| \ll |\chi(\omega)|$. But, using Eq. (4), it is possible to estimate that for $\omega/\omega_{\max} < 1/2$ we have $|\chi_C(\omega)| \sim \frac{1}{\pi}\int_0^{\pi}|\chi(\omega_{\max}e^{i\theta})|d\theta \sim |\mu_1 - 1|$. Thus, we have demonstrated that unless $\omega_{\max} \gg \omega_{\text{res}}$ is satisfied, the Landau and Lifshitz theory applies only for frequencies ω such that $|\mu_1 - 1| \ll |\mu(\omega) - \mu_1|$. It should be obvious that except in the case $\mu_1 = 1$ (which as mentioned before is incompatible with the existence of diamagnetic materials) this restriction may be difficult to be fulfilled in practice, and clearly puts into question whether in general we can actually drop the term $\chi_C(\omega)$ in Eq. (3).

IV. PROPOSED RELATIONS TO LINK μ' AND μ''

As shown next, it is possible to relax the assumptions of Landau and Lifshitz and still link the real and imaginary parts of the magnetic permeability in the frequency region where it retains its physical meaning. Indeed, suppose that as usual the permeability $\mu(\omega)$ is regular at the origin and consider the function $\chi = (\mu - \mu_s)/\omega^2$, with $\mu_s = \mu(0)$. Clearly, χ is an analytic function in the upper half plane and in the real axis (loss, even if vanishingly small, is necessarily present), with the exception of the point $\omega = 0$, where it has a pole of order one. Let us suppose, as in the usual approach, that the permeability has physical meaning up to some frequency $\omega_{\max}$. Proceeding as before, we can now estimate that $|\chi_C(\omega)| \sim \frac{1}{\pi}\int_0^{\pi}|\chi(\omega_{\max}e^{i\theta})|d\theta \sim |\frac{\mu_s-1}{\omega_{\max}^2}|$ for $\omega/\omega_{\max} < 1/2$. Hence, we can drop the term $\chi_C(\omega)$ in Eq. (3) when $|\frac{\mu_s-1}{\omega_{\max}^2}| \ll |\frac{\mu-\mu_s}{\omega^2}|$. Clearly, this condition can be easily satisfied by μ provided $\omega \ll \omega_{\max}$. In other words, the factor $1/\omega^2$ enhances the low-frequency spectrum of the magnetic response of the material, and thus also the contribution of the terms associated with the integral over the real axis in Eq. (3). On the other hand, it depresses the high-frequency content of μ, and thus, as compared to the usual formulation, also the contribution of the term $\chi_C(\omega)$ in Eq. (3). Therefore the requirements for the function $(\mu - \mu_s)/\omega^2$ satisfying the KK-LL relations are much relaxed compared to the case of the function $\mu - \mu_1$. The KK-LL relations for $(\mu - \mu_s)/\omega^2$ link μ' and μ'' as follows:

$$\frac{\mu'(\omega) - \mu_s}{\omega^2} = \frac{1}{\pi}\text{P.V.}\int_{-\omega_{\max}}^{\omega_{\max}} \frac{\mu''(x)}{x^2}\frac{1}{x-\omega}dx, \tag{9a}$$

$$\frac{\mu''(\omega)}{\omega^2} = \frac{1}{i\omega}\left(\left.\frac{d\mu}{d\omega}\right|_{\omega=0}\right) - \frac{1}{\pi}\text{P.V.}\int_{-\omega_{\max}}^{\omega_{\max}} \frac{\mu'(x)-\mu_s}{x^2}\frac{1}{x-\omega}dx. \tag{9b}$$

From the previous discussion it should be clear that these relations are increasingly accurate for smaller values of $\omega/\omega_{\max}$, and they are exact in the limit $\omega_{\max} \to \infty$ (if this limit is meaningful). Equations (9a) and (9b) are the formulas that we propose here as an alternative to the standard KK-LL relations for the permeability function. These formulas are also derived in the Appendix using a different train of thought. Moreover, as an alternative to the third KK-LL relation [Eq. (8)], we propose

$$\frac{\mu(i\omega) - \mu_s}{\omega^2} = -\frac{2}{\pi}\int_{0}^{\omega_{\max}} \frac{\mu''(x)}{x}\frac{1}{x^2+\omega^2}dx. \tag{10}$$

Let us study the implications of Eqs. (9a) and (9b) in what concerns the dispersion of the permeability in a transparency window. To begin with, we note that if $\chi''(\omega) \approx 0$ we find from

Eq. (5a) that

$$\frac{d\chi}{d\omega}(\omega) = \frac{1}{\pi}\int_{-\omega_{\max}}^{+\omega_{\max}} \frac{\chi''(x)}{(x-\omega)^2}dx$$
$$= \frac{4\omega}{\pi}\int_{0}^{+\omega_{\max}} \frac{\chi''(x)x}{(x^2-\omega^2)^2}dx. \quad (11)$$

Therefore, applying the above relation to $\chi = (\mu - \mu_s)/\omega^2$ and noting that $\chi'' = \mathrm{Im}\{\mu - \mu_s\}/\omega^2 = \mu''/\omega^2 > 0$, for $\omega > 0$, we conclude that

$$\frac{d}{d\omega}\left(\frac{\mu - \mu_s}{\omega^2}\right) > 0 \quad \text{if} \quad \mu''(\omega) \approx 0, \quad (12)$$

i.e., in the presence of vanishingly small loss it is necessary that $\chi = (\mu - \mu_s)/\omega^2$ is an increasing function of frequency. Since we can write $\mu = \mu_s + \chi\omega^2$, it follows that $d\mu/d\omega = \omega(2\chi + \omega d\chi/d\omega)$, or equivalently $\frac{d\mu}{d\omega} = \frac{2}{\omega}[(\mu - \mu_s) + \frac{\omega^3}{2}\frac{d\chi}{d\omega}]$. Thus, since $d\chi/d\omega > 0$ in a transparent material, we can immediately conclude that if $\mu > \mu_s$ in the considered frequency range, then $d\mu/d\omega > 0$. However, if the permeability of the material is less than μ_s, the derivative of the permeability is a sum of two terms with opposite signs, and thus the sign of $d\mu/d\omega$ is undetermined! Thus, the relations (9) effectively allow for a regime of anomalous dispersion with no loss when $\mu < \mu_s$, where the permeability function may decrease with frequency. Such a result is completely consistent and may explain, indeed, the dispersion of the permeability of the metamaterials formed by plasmonic-type inclusions described in Sec. II. To further support this assertion, we depict in Fig. 7 the function $(\mu - 1)c^2/\omega^2$ as a function of frequency for a material with the same parameters as in Fig. 2. Consistent with Eq. (12), it is seen that $(\mu - 1)c^2/\omega^2$ is a strictly increasing function of frequency. Notice that we took $\mu_s = 1$, because even for vanishingly small loss (but not exactly zero, $\Gamma = 0^+$) the static permeability is unity, as already discussed in Sec. II.

Similarly, Eq. (10) is less demanding than the usual third KK-LL relation [Eq. (8)], as it does not require that $\mu(i\omega)$ is a strictly decreasing function of frequency over the imaginary axis, but rather that $[\mu_s - \mu(i\omega)]/\omega^2$ is a strictly decreasing function of ω. This restriction only implies that $\mu(i\omega)$ decreases with frequency over the imaginary axis when $\mu(i\omega) > \mu_s$. Actually, the condition $\mu(i\omega) > \mu_s$ can never occur because from Eq. (10) we see that $\mu(i\omega) < \mu_s$ because $\mu''(\omega) > 0$ in the real axis. Hence, we conclude that Eq. (10) enables a regime of anomalous dispersion in the imaginary frequency axis, and this may explain the results of Fig. 4.

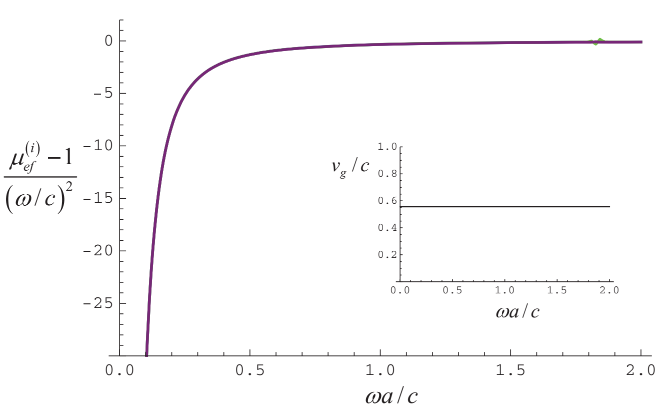

FIG. 7. (Color online) Plot of $(\mu_{\mathrm{ef}}^{(i)} - 1)/(\omega/c)^2$ as a function of frequency for $i = 1, 2, 3$ (the three curves are virtually coincident in the scale of the plot). Inset: group velocity as a function of frequency.

One question that may be raised about our theory is whether the anomalous dispersion of the permeability can be truly consistent with causality. The answer is yes, it is possible to have anomalous dispersion with no loss without violating any fundamental physical principle. For example, in Ref. 27 it was shown that a metamaterial formed by arrays of crossed wires may be characterized by a strong and broadband anomalous dispersion of the index of refraction even for vanishingly small loss. In fact, we believe that the only restriction on the effective response of a lossless material is that the group velocity is less than the velocity of light in vacuum, $|v_g| < c$. Thus, for an isotropic material characterized by the index of refraction $n = \sqrt{\mu\varepsilon}$ it is necessary that $|\frac{d\omega}{dk}| < c$, with $k = \frac{\omega}{c}n$, i.e., $-1 < \frac{1}{n+\omega dn/d\omega} < 1$. Let us discuss specifically the case where both the permittivity and permeability are simultaneously positive so that $0 \leqslant v_g < c$ and thus it is required that $1 < n + \omega dn/d\omega$, and show that such a scenario is compatible with a permeability having anomalous dispersion. Indeed, if the index of refraction $n = \sqrt{\mu\varepsilon}$ of the material is less than unity, the anomalous dispersion of the permeability can always be compensated for by the positive dispersion of the permittivity so that $dn/d\omega > 0$, and $n + \omega dn/d\omega$ remains greater than unity. Alternatively, we can have $dn/d\omega < 0$ (due to the anomalous dispersion of the permeability), but then n should be sufficiently large in the region of anomalous dispersion. In particular, in the latter case it is necessary—but not sufficient—that $\varepsilon > 1/\mu > 1$.

The group velocity for a material with the same parameters as in Fig. 2 is plotted in the inset of Fig. 7. The result is clearly in accordance with the restriction $|v_g| < c$. Somehow, surprisingly, it is seen that the dispersion of the index of refraction is virtually none. Therefore, in this example the positive dispersion of the permittivity apparently compensates perfectly for the anomalous dispersion of the permeability.

To conclude this section, we study the implications in the static limit of the relations (9) for the permeability. It is tempting to put $\omega = 0$ in the right-hand side of Eq. (9a) and use the fact that $\mu''(\omega)/\omega > 0$ to conclude that $\lim_{\omega\to 0}[\mu'(\omega) - \mu_s]/\omega^2 > 0$. However, such reasoning is flawed because if $\omega = 0$ the integral in the right-hand side of Eq. (9a) does not converge. In fact, as shown in Fig. 7 the $\lim_{\omega\to 0}[\mu'(\omega) - \mu_s]/\omega^2$ can be negative, and thus we need to proceed more carefully.[28] A detailed discussion is out of the scope of this work, but it is a simple exercise to show that since the imaginary part of the permeability is an odd function of frequency, $\mu''(x) = -\mu''(-x)$, the $\lim_{\omega\to 0}[\mu'(\omega) - \mu_s]/\omega^2 \equiv C$ [understood as the limit of the right-hand side of Eq. (9a)] always exists. However, it is emphasized that the operator $\lim_{\omega\to 0}$ cannot be interchanged with the integration operator, as discussed above, and thus C can be an arbitrary real number. Hence, it follows that in the quasistatic limit we have $\mu'(\omega) \approx \mu_s + C\omega^2$. Thus, we can conclude that the value of

$\mu_s = \mu(0)$ is a free parameter, and is not restricted in any manner by Eq. (9). In particular, the existence of diamagnetic materials is totally compatible with such formulas, even if $\omega_{\max} = +\infty$.

V. FURTHER DISCUSSION AND CONCLUSIONS

Let us summarize and critically analyze the findings of this work. We have seen that metamaterials formed by inclusions with a plasmonic-type response may have a magnetic response characterized by anomalous dispersion, both in the real and in the imaginary frequency axes, even when loss is vanishingly small; and we have discussed that the usual KK-LL relations for the permeability (assuming passivity, i.e., $\mu'' > 0$) cannot possibly explain this property.

On the other hand, it is unquestionable that such metamaterials have a causal response, which can certainly be measured or calculated with numerical methods. Does causality always imply that the KK relations hold? Here we have to be very careful. In truth, the response of *any* causal linear system satisfies the KK relations with $\omega_{\max} = +\infty$. Thus, for example, if one conceives some device to measure the permeability and defines the magnetic permeability as the result of the measurement with such device, such permeability *will* unquestionably satisfy the KK relations. How can we reconcile this basic property with our results?

The subtle point is that the magnetic permeability μ cannot be regarded as a true physical response for all frequencies. By this we mean that if we conceive two different physical devices to measure μ most likely the results of the measurements will be consistent only up to some frequency $\omega_{\max}$, after which the permeability loses meaning. In fact, it is important to keep in mind that $\mu - 1$ links the magnetization vector and the magnetic field, and since for $\omega \neq 0$ the magnetization vector $\mathbf{M} = \frac{1}{V}\int \frac{1}{2}\mathbf{r} \times \mathbf{j}_d d^3\mathbf{r}$ depends on the origin of the coordinate system, the permeability only has a strict definition (with physical meaning) in the quasistatic limit. Still, a more attentive reader might protest—with reason—that this does not solve our problem. Indeed, such reader might very well argue that even if the response of the two devices does not agree, each measured μ is still a physical response on its own, and thus the result of each measurement should satisfy individually the KK relations. This is certainly true. However, the key point is that since beyond the frequency $\omega_{\max}$ the permeability μ loses its usual physical meaning, its imaginary part is not any more constrained by the passivity condition $\mu'' > 0$. Therefore, if one insists in defining μ as the result of the measurement with some physical device (such that the measured μ will indeed satisfy the KK relations), one must be ready to accept that for some $\omega > \omega_{\max}$ (i.e., in the spectral region where the permeability has no physical meaning, e.g., because of the emergence of strong spatial dispersion) we may have $\mu'' < 0$. It is important to make clear that we totally reject the possibility of a permeability with $\mu'' < 0$ in the region where it retains physical meaning: the condition $\mu'' < 0$ violates passivity.[3,16,23,24] However, if we consider the analytical continuation of μ beyond the region where it retains the usual physical meaning, it may very well happen that in such a region, $\mu'' < 0$. Similarly, if we define μ as the physical response of some device, we must also be ready to accept that the condition $\mu'' < 0$ may occur in a spectral region where the measured μ cannot be interpreted as a magnetic permeability with its usual physical meaning. What do these things show? They demonstrate that any conclusions obtained from the KK relations based on the assumption that $\mu'' > 0$ for $\omega > 0$, for example, that $\mu(\omega = 0) > 1$ (i.e., that all materials are paramagnetic in the static regime), may not apply. In other words, μ as a physical response always satisfies the KK relations with $\omega_{\max} = +\infty$. However, conclusions obtained from the KK relations taking into account that the material is also passive may in general be incorrect, because the condition $\mu'' > 0$ is only ensured in the spectral region where μ has its usual meaning.

In this work, similar to what was done by Landau and Lifshitz[16] we have tried to obtain restrictions on the dispersion of the magnetic permeability considering *exclusively* the frequency region where it retains physical meaning and is consistent with passivity ($\mu'' > 0$ for $\omega > 0$). We argued that the conditions under which the permeability function satisfies the KK relations as formulated by Landau and Lifshitz (KK-LL) may be too strong and are incompatible with a regime of anomalous dispersion that apparently can be observed in realistic metamaterials. Thus in general μ may not satisfy the KK-LL relations (with $\omega_{\max}$ finite and $\mu'' > 0$), even for arbitrarily small frequencies where certainly the permeability function has a very precise physical meaning. It was proven that this regime of lossless anomalous dispersion is, however, compatible with $(\mu - \mu_s)/\omega^2$ satisfying the KK-LL relations (with finite $\omega_{\max}$ and passivity ensured) provided $\mu < \mu_s$ in such regime. These results suggest that in general it is the function $(\mu - \mu_s)/\omega^2$ that satisfies the KK-LL relations rather than the permeability function, as is conventionally assumed, because the former theory is based on weaker assumptions than the latter.

It is important to underline that our results do not contradict causality and that the reason why the corollaries of the KK relations for passive materials are inapplicable (e.g., that anomalous dispersion is not allowed in the case of vanishing loss) is related to the fact that ideally the KK relations for passive materials require that the magnetic response of the material is defined for all frequencies in the upper-half frequency plane (with $\mu'' > 0$ for $\omega > 0$), whereas in general the permeability can only be defined unambiguously for long wavelengths. Indeed, in general $\mu - 1$ cannot be a true physical response for all frequencies (ensuring $\mu'' > 0$ for $\omega > 0$), because otherwise it would be impossible to have diamagnetic materials. Even though this problem can be circumvented in part if one assumes that μ has physical meaning up to a frequency $\omega_{\max}$ (as done in Ref. 16), i.e., that μ is a physical response for signals with a sufficiently slow time variation, the application of the KK-LL relations to μ is only possible if $\mu(\omega) - \mu(\omega_{\max}) \approx 0$ for all complex frequencies (in the upper half plane) such that $|\omega| = \omega_{\max}$. However, as discussed in Sec. III, this condition may be too strong and is certainly not enforced by causality, and thus in general it may not be observed.

Our findings do not contradict, but rather complement, the theory of Landau and Lifshitz. As mentioned in a footnote in Ref. 16 (p. 283), in natural magnetic materials the parameter $\omega_{\max}$ should be sufficiently large so that $\omega_{\max}\tau \gg 1$, where τ is the shortest relaxation time for ferromagnetic and paramagnetic processes. This condition can be adapted to metamaterials as $\omega_{\max} \gg \omega_{\mathrm{res}}$, where ω_{res} is the largest resonance frequency of μ. However, in practice this condition may be impossible to fulfill in a metamaterial. For example, the metamaterials considered in Sec. II have a strong magnetic response from the static regime up to some frequency where the permeability breaks down due to the effects of spatial dispersion (e.g., at $\omega a/c = 1.8$ for Fig. 2). Clearly, in such circumstances it is impossible to define a $\omega_{\max}$ consistent with the definition of Landau and Lifshitz (actually the only possibility would be $\omega_{\max} = 0$).

Moreover, in continuous media (limit case where $a \to 0$) the KK-LL relations are expected to apply. This is indicated by the fact that in this limit the interaction constant is $C'_{\mathrm{int}} \approx \frac{1}{V_{\mathrm{cell}}}\frac{1}{3}$, and thus, at least within the framework of the Clausius-Mossotti formula, it is impossible to have anomalous dispersion effects in the limit of vanishing loss. This indicates that the key reason for the KK-LL relations breaking down is the intrinsic granularity of matter.

Finally, we note that unlike the permeability, the electric permittivity can be defined unambiguously for arbitrarily large frequencies. Indeed, $\varepsilon - 1$ links the averaged polarization currents and the average electric field, and such quantities can be defined unequivocally at any frequency (independent of the origin of the coordinate system), no matter how large, and hence, in principle, the findings of this work apply only to the magnetic permeability function. Our theory can be generalized in a trivial manner to the case of anisotropic magnetic materials.

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APPENDIX

It is interesting to point out that the arguments that we have used to conclude that $(\mu - \mu_s)/\omega^2$ satisfies the KK-LL relations apply word for word to the function $(\mu_s^{-1} - \mu^{-1})/\omega^2$, which therefore also satisfies formulas analogous to Eqs. (9). As shown in this appendix, in the case of metal-dielectric metamaterials, it is possible to reach the same conclusion using a quite different train of thought.

In fact, it is well known that the effective electromagnetic response of a metamaterial can in general be characterized by a nonlocal dielectric function of the form $\overline{\overline{\varepsilon}}_{\mathrm{eff}}(\omega,\mathbf{k})$, where $\mathbf{k}$ is the wave vector.[12,29] Such framework is much more rigorous than the more conventional effective medium approach based on the definition of an effective permittivity ε and an effective permeability μ. In particular, unlike the effective permeability, which may lose its meaning at relatively small frequencies, in principle the nonlocal dielectric function $\overline{\overline{\varepsilon}}_{\mathrm{eff}}(\omega,\mathbf{k})$ can be defined unambiguously over a wide range of frequencies,[12] and thus we can assume that $\lim_{\omega\to\infty} \overline{\overline{\varepsilon}}_{\mathrm{eff}}(\omega,\mathbf{k}) = 1$, i.e., for all purposes we can take $\omega_{\max} = +\infty$. Therefore, as demonstrated in Ref. 29 (p. 13), $\overline{\overline{\varepsilon}}_{\mathrm{eff}}(\omega,\mathbf{k})$ satisfies the KK relations for arbitrary $\mathbf{k}$ fixed. On the other hand, a hypothetical isotropic metamaterial characterized by an effective permittivity ε and an effective permeability μ can be as well characterized—at least in the frequency range where μ has physical meaning—by a nonlocal dielectric function such that[12,30]

$$\overline{\overline{\varepsilon}}_{\mathrm{eff}}(\omega,\mathbf{k}) = \varepsilon\overline{\overline{\mathbf{I}}} + \frac{c^2}{\omega^2}(\mu^{-1} - 1)\mathbf{k} \times \overline{\overline{\mathbf{I}}} \times \mathbf{k}. \tag{A1}$$

But it should be evident that $\overline{\overline{\varepsilon}}_{\mathrm{eff}}(\omega,\mathbf{k})$ can satisfy the KK relations for arbitrary $\mathbf{k}$ fixed if, and only if, both ε and $(1 - \mu^{-1})/\omega^2$ satisfy the same relations. This finding is in complete harmony with our previous assertion that $(\mu_s^{-1} - \mu^{-1})/\omega^2$ should satisfy the KK relations, because the static effective permeability associated with any metal-dielectric metamaterial is necessarily trivial, $\mu_s = 1$, as discussed at length in Ref. 15.

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