Anisotropic ac Electrical Permittivity of a Periodic Metal-Dielectric Composite Film in a Strong Magnetic Field

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The ac magnetoelectric properties of a metal-dielectric composite are considered in the quasistatic regime. When the metal constituent is not in the form of a dilute suspension, the magnetoplasma resonances are altered in a way that reflects the microstructure. When the microstructure is periodic with more than one characteristic length scale, the electric permittivity can exhibit a strong dependence on both the magnitude and the direction of the applied dc magnetic field. This will occur in the vicinity of one of those resonances if $\omega\tau > 1$. The possibility of observing this effect in a suitably synthesized composite film is considered in detail. [S0031-9007(97)05127-2]

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Recent advances in thin film fabrication techniques permit the synthesis of made-to-order composite thin semiconducting films with a microstructure that is quite complicated [1]. In particular, periodic microstructures where the size scales of lateral inhomogeneities are comparable to the film thickness can be made routinely. Such structured films are, in some sense, a new form of matter, and the study of their physical properties is being pursued by a rapidly growing number of research groups. When such films are mesoscopic in character and held at low temperatures (i.e., liquid He temperature or lower), they exhibit a variety of ballistic and quantum phenomena in electrical transport. However, even at higher temperatures such systems sometimes exhibit interesting new modes of behavior, some of which can be understood within the framework of classical continuum transport theory. Very recently, the phenomenon of induced anisotropic dc magnetotransport in a composite conductor with a periodic microstructure was first predicted and studied theoretically and numerically [2–5], and then verified experimentally [6]. Here we discuss the possibility of observing analogous behavior in the ac electric permittivity of a metal-dielectric composite with a periodic microstructure in the presence of a strong magnetic field.

When an external static or quasistatic electric field $\mathbf{E}_0$ is applied to a finite sample of dielectric or conducting material, the induced dipole moment (and higher moments when the shape is nonspherical) is responsible for the depolarization effect, i.e., the internal field differs from $\mathbf{E}_0$. In the case of an ac electric field of angular frequency ω, applied to a metallic particle that is smaller than the skin depth, this has the effect of changing the plasma resonance frequency from its intrinsic value $\omega_p/\sqrt{\epsilon_1}$, where $\omega_p \equiv (4\pi ne^2/m)^{1/2}$ (n is the density of charge carriers, e the charge of one carrier, m its effective mass, ϵ_1 the dielectric constant of the underlying ionic lattice), to $\omega_p\sqrt{L/\epsilon_1}$, where L is the appropriate depolarization factor. (L usually depends on the direction of $\mathbf{E}_0$ and on the dielectric constant of the surrounding medium ϵ_2. When $\epsilon_2 = \epsilon_1$ and the inclusion is spherical, then $L = 1/3$ for all directions of $\mathbf{E}_0$.)

In the presence of a static magnetic field $\mathbf{B}$, this so called "surface plasmon resonance" of an isolated metal sphere is split into two resonances. Since the internal electric field is uniform even in the presence of $\mathbf{B}$, the problem is essentially solvable in closed form [7–10]. When the magnetic field is small so that $\omega_c \ll \omega_p$ ($\omega_c \equiv e|\mathbf{B}|/mc$ is the cyclotron frequency), then the two resonances occur at

$$\omega_\pm \cong \frac{\omega_p}{\sqrt{3\epsilon_1}} \pm \frac{\omega_c}{2} + \frac{3\sqrt{3\epsilon_1}}{8}\frac{\omega_c^2}{\omega_p}, \qquad (1)$$

while when the magnetic field is strong so that $\omega_c \gg \omega_p$, they occur at

$$\omega_- \cong \frac{\omega_p^2}{3\epsilon_1\omega_c} \quad (\text{"magnetoplasma resonance"}), \qquad (2)$$

$$\omega_+ \cong \omega_c + \frac{\omega_p^2}{3\epsilon_1\omega_c} \quad (\text{"magnetoplasma shifted cyclotron resonance"}). \qquad (3)$$

The factor 3 in these expressions stands for the dimensionality and embodies the effect of depolarization on the magnetoplasma phenomenon in an isolated sphere. In order to observe these resonances, the conductivity relaxation time τ must satisfy $\omega\tau > 1$. Such resonances were studied both experimentally and theoretically many years ago [11,12] on small semiconducting samples of various shapes. That and related work are also discussed in the review article on cyclotron resonance by Lax and Mavroides [13]. These resonances can also be observed in a metal-dielectric composite in which a low density of small metal spheres (radii must be smaller than the electromagnetic skin depth) are embedded in a dielectric host.

When we consider a nondilute metal-dielectric composite, these resonances will be affected by electromagnetic interactions between different inclusions (this is analogous to multiple scattering), and the properties will depend on the microstructure. We now consider such systems for the case where the metallic constituent is a simple, free electron metal. In this case, the electric permittivity tensor $\hat{\epsilon}_m$ has the following form:

$$\hat{\epsilon}_m = \epsilon_1 - \frac{4\pi}{i\omega\hat{\rho}_m}, \tag{4}$$

where (we assume $\mathbf{B} \parallel z$)

$$\hat{\rho}_m = \frac{4\pi}{\omega_p^2\tau}\begin{pmatrix} 1 - i\omega\tau & -\omega_c\tau & 0 \\ \omega_c\tau & 1 - i\omega\tau & 0 \\ 0 & 0 & 1 - i\omega\tau \end{pmatrix} \tag{5}$$

is the ac resistivity tensor. The magnetic field enters through the cyclotron frequency ω_c, which appears only in the antisymmetric off-diagonal elements of $\hat{\rho}_m$.

If the microstructure is isotropic and if the metal volume fraction is not too large, then a Clausius-Mossotti-type (CMT) expression can provide a good approximation for the bulk effective permittivity tensor $\hat{\epsilon}_e$. Assuming that the dielectric constituent is itself isotropic, with a scalar dielectric tensor $\hat{\epsilon}_d = \epsilon_2$, and that the metallic constituent is in the form of spherical inclusions, the CMT approximation for $\hat{\epsilon}_e$ is given by [7] ($\langle\,\rangle$ denotes a volume average)

$$\hat{\epsilon}_e = \epsilon_2 + \langle(d\epsilon_2 - \delta\hat{\epsilon})^{-1}\rangle^{-1}\langle(d\epsilon_2 - \delta\hat{\epsilon})^{-1}\delta\hat{\epsilon}\rangle, \tag{6}$$

where

$$\delta\hat{\epsilon}(\mathbf{r}) = \begin{cases} \epsilon_1 - \epsilon_2 - \frac{4\pi}{i\omega\hat{\rho}_m} & \text{inside the metal constituent,} \\ 0 & \text{in the dielectric constituent,} \end{cases} \tag{7}$$

and d is the dimensionality, equal to 3 in the case of spherical inclusions, but equal to 2 in the case of infinitely long and parallel cylindrical inclusions. Using this approximation we can get closed form expressions for the elements of $\hat{\epsilon}_e$. However, because those are somewhat complicated, we will assume, for simplicity, that $\epsilon_1 = \epsilon_2 = \epsilon$. To further simplify the results, and since we are interested in the case where $\omega\tau > 1$, we will in fact consider only those expressions in the extreme limit $\omega\tau \gg 1$.

In that case we get ($p_m, p_d = 1 - p_m$ are the volume fractions of the metal and the dielectric constituents, respectively)

$$\epsilon_{xy}^{(e)} = -\epsilon_{yx}^{(e)} = -ip_m d\epsilon y[x^2 - x(y + 2) + 1]/D, \tag{8}$$

$$\epsilon_{xx}^{(e)} = \epsilon_{yy}^{(e)} = \epsilon - p_m d\epsilon(x - p_d) \times [x^2 - x(y + 2) + 1]/D, \tag{9}$$

$$\epsilon_{zz}^{(e)} = \epsilon - p_m d\epsilon/(x - p_d), \tag{10}$$

$$\epsilon_{zx}^{(e)} = \epsilon_{xz}^{(e)} = \epsilon_{zy}^{(e)} = \epsilon_{yz}^{(e)} = 0, \tag{11}$$

where

$$x \equiv \frac{d\epsilon\omega^2}{\omega_p^2} \propto \frac{\omega^2}{n}, \qquad y \equiv \frac{d\epsilon\omega_c^2}{\omega_p^2} \propto \frac{\mathbf{B}^2}{n}, \tag{12}$$

$$D \equiv [x^2 - x(y + 2) + 1][x^2 - x(y + 2p_d) + p_d^2] + p_m^2 y(x - y). \tag{13}$$

When the magnetic field is nonzero, i.e., $y > 0$, then both $\epsilon_{xy}^{(e)}$ and $\epsilon_{xx}^{(e)}$ diverge when x is equal to one of the zeros of D, of which there are usually four. When $p_m \to 0$, these divergences occur at the zeros of $x^2 - x(2 + y) + 1$, namely, when the frequency is equal to one of the resonance frequencies $\omega_\pm$ of an isolated metallic spherical inclusion

$$2\omega_\pm^2 = \omega_c^2 + \frac{2\omega_p^2}{d\epsilon} \pm \left(\omega_c^4 + \frac{4\omega_c^2\omega_p^2}{d\epsilon}\right)^{1/2} \quad \text{for } \omega_\pm\tau \gg 1. \tag{14}$$

This expression reduces to (1), (2), (3) in the appropriate limits. However, when p_m is finite the resonance frequencies depend upon it; i.e., they depend on the microstructure.

If $\mathbf{B}$ is not too small (i.e., y not much smaller than 1), then $\epsilon_{xx}^{(e)}$ goes through 0 at a different frequency, given by one of the zeros of

$$[x^2 - x(y + 2) + 1] \times [x^2 - x(y + 2p_d + dp_m) + p_d(p_d + dp_m)] + p_m^2 y(x - y). \tag{15}$$

At the same time, $\epsilon_{xy}^{(e)} = -\epsilon_{yx}^{(e)}$ remains finite and imaginary. Thus, we will have in our possession a composite medium where the dielectric behavior exhibits negligible dissipation (this is a result of considering the extreme limit $\omega\tau \gg 1$), while the ratio $\epsilon_{xy}^{(e)}/\epsilon_{xx}^{(e)}$ has a large magnitude.

Recent studies of the bulk effective dc conductivity tensor $\hat{\sigma}_e$ of conducting composites have shown that when in at least one constituent the antisymmetric off-diagonal terms of the local conductivity tensor are large, i.e., $|\sigma_{xy}/\sigma_{xx}| \gg 1$ and $|\sigma_{xy}/\sigma_{yy}| \gg 1$, interesting behavior can ensue: First of all, a strong dependence of the bulk effective Ohmic resistivity components $\rho_{xx}^{(e)}, \rho_{yy}^{(e)}, \rho_{zz}^{(e)}$ on magnetic field strength $|\mathbf{B}|$ appears, even when those resistivity components were field independent in each of the homogeneous constituents. Second, in the case of a periodic microstructure, those resistivity components all become very anisotropic, i.e., their values depend strongly on the precise orientation of $\mathbf{B}$ and of the volume averaged current density $\langle\mathbf{J}\rangle$ with respect to the microstructure [2–6].

Because the magnetoelectric behavior of a metal-dielectric composite in the quasistatic regime depends upon the microstructure in the same way as the dc magnetotransport behavior described above, we expect that similar effects will be observed. In particular, we expect that the diagonal components of the bulk effective ac permittivity tensor $\epsilon_{xx}^{(e)}$, $\epsilon_{yy}^{(e)}$, $\epsilon_{zz}^{(e)}$ will acquire a strong dependence on $|\mathbf{B}|$ even when the bulk effective dc conductivity vanishes, and that in the case of a periodic microstructure, a strong anisotropy of those permittivities will appear. Unfortunately, such behavior can never be found within the CMT approximation, where the periodic character of the microstructure is entirely ignored. Therefore we need to perform a more precise calculation of $\hat{\epsilon}_e$, where the details of the microstructure are taken into consideration. A practical method for doing this was developed earlier in the context of dc magnetotransport [3]. That method is based on solving a truncated set of linear algebraic equations for the Fourier expansion coefficients of the local electric field in the periodic composite.

In order to have a system where $\hat{\epsilon}_e$ depends on $\mathbf{B}$ as described above, we need to have a dielectric host for which the electric permittivity tensor $\hat{\epsilon}$ has $\mathbf{B}$-dependent antisymmetric components that are large compared to the diagonal components. In fact, for our purposes the strong field regime holds whenever $|\epsilon_{xy}/\epsilon_{xx}| > 1$ and $|\epsilon_{xy}/\epsilon_{yy}| > 1$. Such a material is not commonplace. However, we could use a random, isotropic suspension of metal spheres in a standard uniform dielectric host with a real, positive, scalar electric permittivity tensor. This composite would then serve as the nonstandard dielectric host, where $\epsilon_{xy} = -\epsilon_{yx}$ can be much greater than ϵ_{xx} and ϵ_{yy} when ω and $\mathbf{B}$ are chosen appropriately, as shown above in our discussion of the CMT approximation. Near a resonance, the ratios $\epsilon_{xy}/\epsilon_{xx} > 1$, $\epsilon_{xy}/\epsilon_{yy} > 1$ can become large even for modest fields $\mathbf{B}$, since ϵ_{xx} and ϵ_{yy} become very small. In this nonstandard dielectric host we would embed a periodic array of much larger inclusions which would again be made of a standard dielectric material, with scalar $\hat{\epsilon}$. The strong dependence of the bulk effective $\hat{\epsilon}_e$ upon $\mathbf{B}$ would appear when ω is near the value where ϵ_{xx} of the random suspension (the nonstandard dielectric host) vanishes.

It is actually possible to make such a nonstandard composite host in other ways so as to have a large ratio $\epsilon_{xy}/\epsilon_{xx}$: This will occur in the vicinity of any sharp quasistatic resonance of the composite host. Such sharp resonances are found in a composite when it is made in the form of a dilute suspension of identically shaped and identically oriented inclusions [14], as in the example discussed above of spherical inclusions. Such sharp resonances are also found in nondilute composites when the microstructure is periodic [14].

Combining all of the above considerations, we can expect that a strong dependence of $\epsilon_{xx}^{(e)}$ on both the magnitude and the direction of $\mathbf{B}$ will appear if we use a periodic array of identically shaped metallic inclusions embedded in a standard dielectric host, and if the microstructure has at least *two characteristic length scales:* a small length scale region to mimic the nonstandard dielectric host, and a large length scale region to mimic the array of large standard dielectric inclusions. We have tried out this idea by computing the magnetoelectric properties of a periodic composite with the unit cell shown in Fig. 1(a), using the method described briefly above (see Ref. [3]). This is a two dimensional microstructure, where each of the small squares represents an infinitely long metal rod of square cross section, which lies parallel to the z axis. In Fig. 1(b) we show a polar plot of the real and imaginary parts of the transverse diagonal component of $\hat{\epsilon}_e$ for the case where both $\mathbf{B}$ and $\langle \mathbf{E} \rangle$ are in the x, y plane and remain perpendicular to each other as $\mathbf{B}$ is rotated in that plane. The dimensionless parameters used to obtain these results are indicated in the figure caption. Those parameters can be realized by using intrinsic (undoped) InAs as the dielectric host, i.e., $\epsilon_2 = 10$, and Si-doped InAs rods as the metal inclusions, with $\epsilon_1 = 10$, a negative charge carrier density $n = 6 \times 10^{16}$ cm^{-3}, an effective mass $m = 0.023 m_e$ (m_e is the bare mass of a free electron), and a mobility $\mu = 3$ m^2(V s)$^{-1}$ (note that $\omega_c \tau = \mu|\mathbf{B}|$, and that this is equal to the Hall-to-Ohmic resistivity ratio), which translate into a bulk plasma frequency $\omega_p \cong 0.9 \times 10^{14}$ s^{-1}, an Ohmic conductivity $\sigma \cong 280$ (Ω cm)$^{-1}$, a relaxation time $\tau \cong 4 \times 10^{-13}$ s, and a mean free path $\ell \cong 220$ nm. With these values, the assumed value of $\omega_c \tau = 5$ can be obtained by

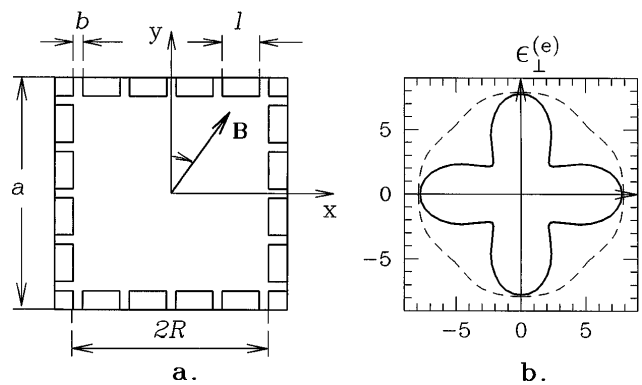

FIG. 1. (a) A square unit cell of the periodic microstructure used in our calculations: the small squares represent a cross section of the infinitely long metal rods that lie parallel to the z axis. The size parameters were $l/a = 0.16$, $b/a = 0.04$, $R/a = 0.4$. The two important size scales in this system are represented, we believe, by b and $2R$, which differ by a factor of 20. (b) Polar plot of the bulk effective transverse electric permittivity $\epsilon_{\perp}^{(e)}$ of the periodic composite with unit cell shown in (a)—solid line shows the real part, dashed line the imaginary part. The magnetic field $\mathbf{B}$ has a fixed magnitude and is rotated in the x, y plane. The angular frequency of the applied electric field is $\omega = 0.3\omega_p$, and lies in the resonance region, where the ratio $\epsilon_{xy}^{(e)}/\epsilon_{xx}^{(e)}$ would be large if the composite consisted entirely of a square array of square rods of cross section $l \times l$ and with a nearest neighbor spacing b, as in (a). The dielectric constants were taken to be $\epsilon_2 = \epsilon_1 = 10$, while the electric permittivity tensor of the inclusions was taken to have the form of (4) and (5) with $\omega_c \tau = \mu|\mathbf{B}| = 5$ and $\omega_p \tau = 35$.

using a magnetic field of 1.7 T, and the angular frequency of the electric field needs to be about 2.7×10^{13} s^{-1}; thus $\omega\tau \cong 10$. This translates into a wavelength in vacuum $\lambda_0 \cong 70$ μm, a wavelength inside the semiconductor $\lambda_s \cong 22$ μm, and a skin depth $\delta \cong 1.5$ μm, which are all large compared to ℓ and to the typical de Broglie wavelength $\lambda_e \cong 50$ nm of the charge carriers. This is necessary in order that our complete neglect of ballistic effects and quantum properties of the charge carriers be a good approximation [6]. To this end, it is also necessary that the system be held at a temperature T that is not too low, namely, $k_B T > \hbar\omega_c$, which means $T > 90$ K. It is also necessary that the length scales which characterize the relevant aspects of the microstructure lie in between the two length scales mentioned above. This means that the length parameters l, a, and $2R$ of Fig. 1(a) must satisfy

$$\ell, \lambda_e \ll l, a, 2R \ll \lambda_s, \delta \,. \qquad (16)$$

Finally, our experience with dc magnetotransport in composites has shown that, in practice, one can use film shaped specimens that are quite thin [6]: Even when the film thickness h is equal only to the cross sectional size $2R$ or l of a single inclusion, for strong **B** one already gets a magnetoelectric effect that is of the same order of magnitude as the effect in a film of infinite thickness. The requirement that must be satisfied in order to be effectively in the $h = \infty$ limit is $\omega_c\tau \gg R/h$ [4].

In summary, by using the CMT approximation we first showed that in a nondilute metal-dielectric composite medium the magnetoplasma resonances depend upon the microstructure. Near such a resonance, it is possible to achieve large values for the ratio $\epsilon_{xy}/\epsilon_{xx} = -\epsilon_{yx}/\epsilon_{xx}$. Motivated by this observation and by results of previous studies of dc magnetotransport in composite conductors, we then performed a numerical study of the ac magnetoelectric properties of a particular metal-dielectric periodic composite structure which had two characteristic length scales, determined by the particular nonuniform distribution of the metallic inclusions. When the frequency was in the vicinity of one of the sharp resonances, there appeared a strong dependence of the transverse electric permittivity on both the magnitude and the direction of the applied static magnetic field. This is presumably related to the appearance of large values for the ratio $\epsilon_{xy}/\epsilon_{xx} = -\epsilon_{yx}/\epsilon_{xx}$ near the resonance. We hope that the results presented here will stimulate experimental study aimed at verification of our predictions and continued exploration of the magneto-optical properties of such systems.

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