

Faraday rotation, band splitting, and one-way propagation of plasmon waves on a nanoparticle chain

N. A. Pike and D. Stroud

Citation: [Journal of Applied Physics](#) **119**, 113103 (2016); doi: 10.1063/1.4943647

View online: <http://dx.doi.org/10.1063/1.4943647>

View Table of Contents: <http://scitation.aip.org/content/aip/journal/jap/119/11?ver=pdfcov>

Published by the [AIP Publishing](#)

Articles you may be interested in

[Overcoming the adverse effects of substrate on the waveguiding properties of plasmonic nanoparticle chains](#)

J. Appl. Phys. **119**, 043101 (2016); 10.1063/1.4940415

[Plasmonic enhancement of photocurrent in carbon nanotube by Au nanoparticles](#)

Appl. Phys. Lett. **102**, 103102 (2013); 10.1063/1.4794937

[Plasmonic structures of metal nanoparticles: Chains and arrays](#)

AIP Conf. Proc. **1475**, 9 (2012); 10.1063/1.4750078

[Colored porous silicon as support for plasmonic nanoparticles](#)

J. Appl. Phys. **111**, 084302 (2012); 10.1063/1.3703469

[Tunable localization and oscillation of coupled plasmon waves in graded plasmonic chains](#)

J. Appl. Phys. **106**, 113307 (2009); 10.1063/1.3268459



INNOVATION LEADERSHIP RELIABILITY

VALVES

GLOBAL LEADER

MODULES

CONCEPT TO PRODUCT

BELLOWS

LEADING TECHNOLOGY

SERVICES

24/7 GLOBAL SUPPORT

VAT Booth #731

APS March Meeting, Baltimore, MD

www.vatvalve.com

Faraday rotation, band splitting, and one-way propagation of plasmon waves on a nanoparticle chain

N. A. Pike^{1,2} and D. Stroud^{2,a)}

¹Department of Physics, University of Liege, 4000 Liege, Belgium

²Department of Physics, The Ohio State University, Columbus, Ohio 43210, USA

(Received 8 October 2015; accepted 29 February 2016; published online 15 March 2016)

We calculate the dispersion relations of plasmonic waves propagating along a chain of semiconducting or metallic nanoparticles in the presence of both a static magnetic field \mathbf{B} and a liquid crystalline host. The dispersion relations are obtained using the quasistatic approximation and a dipole-dipole approximation to treat the interaction between surface plasmons on different nanoparticles. For plasmons propagating along a particle chain in a nematic liquid crystalline host with both \mathbf{B} and the director parallel to the chain, we find a small, but finite, Faraday rotation angle. For \mathbf{B} perpendicular to the chain, but director still parallel to the chain, the field couples the longitudinal and one of the two transverse plasmonic branches. This coupling is shown to split the two branches at the zero field crossing by an amount proportional to $|\mathbf{B}|$. In a cholesteric liquid crystal host and an applied magnetic field parallel to the chain, the dispersion relations for left- and right-moving waves are found to be different. For some frequencies, the plasmonic wave propagates only in one of the two directions.

© 2016 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4943647>]

I. INTRODUCTION

Ordered arrays of metal particles in dielectric hosts have many remarkable properties.^{1–6} For example, they support propagating modes which are linear superpositions of the so-called surface or particle plasmons. In dilute suspensions of such nanoparticles, these surface plasmons give rise to the characteristic absorption peaks, in the near infrared or visible, which play an important role in their optical response, and which have recently been observed in semiconductor nanoparticles as well as metallic ones.^{7,8} For ordered chains, if both the particle dimensions and the interparticle separation are much smaller than the wavelength of light, one can readily calculate the dispersion relations for both transverse (T) and longitudinal (L) waves propagating along the chain, using the quasistatic approximation, in which the curl of the electric field is neglected.

In a previous paper, we calculated these dispersion relations for metallic chains immersed in an anisotropic host, such as a nematic or cholesteric liquid crystal (NLC or CLC).⁵ The liquid crystalline host serves as an anisotropic dielectric medium whose alignment and dielectric tensor can be controlled by varying the direction and magnitude of an external static electric field, or temperature. Thus, we can tune the dispersion relations of the plasmonic waves by adjusting these control parameters. The effects of an isotropic host material, such as air or vacuum, can be found, for example, in Refs. 5 and 9.

It is worth briefly summarizing some of the new effects generated by the anisotropic host, in comparison to an isotropic host such as air or vacuum. With an isotropic host, there are three branches of plasmonic waves propagating along the chain: one longitudinal (L) and two degenerate transverse (T)

branches. For a uniaxially anisotropic host, such as an NLC, these three branches still exist, and the two T branches remain degenerate as long as the symmetry axis of the host dielectric tensor (i.e., the director of an NLC) is parallel to the chain. The degeneracy persists because the host does not break the symmetry between the two T branches. The three branches are, of course, shifted up or down because the host has a different dielectric tensor than an isotropic host. Furthermore, the L branch continues to cross the doubly degenerate T branch at a particular wave vector, just as in the isotropic case. If, however, the director of the NLC is perpendicular to the particle chain, the symmetry between the two T branches is broken, and it is found that they are no longer degenerate. This is a feature special to the *anisotropic* host and was obtained numerically in Ref. 5.

In this article, we consider the additional effects of a static magnetic field applied either parallel or perpendicular to a chain of nanoparticles in an anisotropic host. In order to obtain a larger effect from the magnetic field, we will also consider doped semiconducting nanoparticles, besides the metallic particles considered previously. Such nanoparticles have a much lower electron density than typical metallic nanoparticles. For example, the electron density in semiconductor nanoparticles, such as the Cu_{2-x}S nanoparticles whose optical properties have recently been studied,⁷ can be adjusted over a broad range from 10^{17} to 10^{22} cm^{-3} or even lower. The largest effects are obtained with electron densities towards the lower end of this range. We find, for a parallel magnetic field orientation, that a linearly polarized T wave undergoes a Faraday rotation as it propagates along the chain. For a field of 2 T and a suitably low electron density, this Faraday rotation can be at least 1° per ten interparticle spacings. For the case of very small nanoparticles with diameters (denoted $2a$) of 3–5 nm,⁷ and spacing $d = 3a$, this rotation corresponds to a 1° for every 45–75 nm of chain length.

^{a)}Electronic mail: Stroud@physics.osu.edu

In this case, for the parallel field orientation, the NLC quantitatively modifies the amount of Faraday rotation, but there would still be rotation without the NLC host.

As mentioned, our calculations are carried out in the quasistatic approximation, which is suitable when both the particle size and the separation between particles are small compared to the wavelength of light in the medium. If both these conditions are satisfied, the relevant quantity for calculating the plasmon dispersion relation is the ratio a/d between the particle radius a and center-to-center separation d between particles. As is shown below, the plasmon waves typically propagate at frequencies of order $0.4\omega_p$, where ω_p is the plasma frequency. For particles of typical metallic electron density, ω_p is in the near ultraviolet, so the particles should be spaced of order 20–40 nm or less, in order to satisfy this condition. However, if semiconducting nanoparticles are used, the spacing (and particle radius) can be much larger. Our calculations are all carried out in terms of the dimensionless quantity kd , so they are, in principle, valid for a range of particle sizes and separations, so long as the ratio a/d is fixed. As far as chain length is concerned, our dispersion relations are computed assuming an infinitely long chain of particles. Of course, a realistic chain in a physical system will be a finite number of perhaps 50–100 particles. The “end effects” associated with the finite chain length could be calculated, in principle, though they are not considered in this paper, but the infinite-chain calculation should provide a good approximation, and can easily be carried out using Bloch’s theorem.

We also consider the propagation of plasmonic waves along a nanoparticle chain but with a CLC host. In this case, if the magnetic field is parallel to the chain and the director rotates about the chain axis with a finite pitch angle, we show that the frequencies of left- and right-propagating waves are, in general, not equal. Because of this difference, it is possible, in principle, that for certain frequencies, a linearly polarized wave can propagate along the chain only in one of the two possible directions. Indeed, for sufficiently low electron concentration, we do find one-way propagation in certain frequency ranges. This realization of one-way propagation is quite different from other proposals for one-wave wave-guiding.^{10–14}

The remainder of this article is organized as follows: First, we use the formalism of Ref. 5 to determine the dispersion relations for the L and T waves in the presence of an anisotropic host and a static magnetic field. Next, we give simple numerical examples and finally we provide a brief concluding discussion.

II. FORMALISM

We consider a chain of identical metallic or semiconducting nanoparticles, each a sphere of radius a , arranged in a one-dimensional periodic lattice along the z axis, with lattice spacing d , so that the n th particle is assumed centered at $(0, 0, nd)$ ($-\infty < n < +\infty$). The propagation of plasmonic waves along such a chain of nanoparticles has already been considered extensively for the case of isotropic metal particles embedded in a homogeneous, isotropic medium.⁹ In the present work, we calculate, within the quasistatic

approximation, how the plasmon dispersion relations are modified when the particle chain is immersed in both an anisotropic dielectric, such as an NLC or CLC, and a static magnetic field. We thus generalize earlier work in which an anisotropic host is considered without the magnetic field.^{5,6}

In the absence of a magnetic field, the medium inside the particles is assumed to have a scalar dielectric function. If there is a magnetic field \mathbf{B} parallel to the chain (which we take to lie along the z axis), the dielectric function of the particles becomes a tensor, $\hat{\epsilon}$. In the Drude approximation, the diagonal components are $\epsilon(\omega)$, while $\epsilon_{xy} = -\epsilon_{yx} = iA(\omega)$ and all other components vanish. In this case, the components of the dielectric tensor take the form

$$\epsilon(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i/\tau)} \rightarrow 1 - \frac{\omega_p^2}{\omega^2} \quad (1)$$

and

$$A(\omega) = -\frac{\omega_p^2\tau}{\omega} \frac{\omega_c\tau}{(1 - i\omega\tau)^2} \rightarrow \frac{\omega_p^2\omega_c}{\omega^3}, \quad (2)$$

where ω_p is the plasma frequency, τ is a relaxation time, and ω_c is the cyclotron frequency, and the second limit is applicable when $\omega\tau \rightarrow \infty$. We will use Gaussian units throughout. While this approximation may be somewhat crude, especially for semiconducting nanoparticles, it should be a reasonable first approximation.

The dielectric function of the liquid crystal host, for either the NLC or CLC case, is taken to be that described in Ref. 5. The dispersion relations for the surface plasmon waves are determined within the formalism of Ref. 5. Specifically, we write down a set of self-consistent equations for the coupled dipole moments; these are given in Ref. 5 as Eq. (9), and repeated here for reference

$$\mathbf{p}_n = -\frac{4\pi a^3}{3} \hat{\mathbf{t}} \sum_{n' \neq n} \hat{\mathcal{G}}(\mathbf{x}_n - \mathbf{x}_{n'}) \mathbf{p}_{n'}. \quad (3)$$

Here,

$$\hat{\mathbf{t}} = \delta\hat{\epsilon}(\hat{\mathbf{1}} - \hat{\mathbf{\Gamma}}\delta\hat{\epsilon})^{-1} \quad (4)$$

is a “t-matrix” describing the scattering properties of the nanoparticle spheres in the surrounding material, $\hat{\mathcal{G}}$ and $\hat{\mathbf{\Gamma}}$ are a 3×3 Green’s function and depolarization matrix given in Ref. 5, $\hat{\mathbf{1}}$ is the 3×3 identity matrix, and $\delta\hat{\epsilon} = \hat{\epsilon} - \hat{\epsilon}_h$, where $\hat{\epsilon}_h$ is the dielectric tensor of the liquid crystal host.

A. Nematic liquid crystal

We first consider a chain of such particles placed in an NLC host with $\mathbf{B} \parallel \hat{z}$ and parallel to the liquid crystal director \hat{n} . Using the formalism of Ref. 5, combined with Eq. (3), we obtain two coupled sets of linear equations for the transverse (T) components of the polarization, p_{nx} and p_{ny} . The solutions are found to be left- and right-circularly polarized transverse waves with frequency ω and wave number k_{\pm} , where the frequencies and wave numbers are connected by

the dispersion relations in the nearest-neighbor approximation

$$1 = -\frac{2a^3 \epsilon_{\parallel}}{3d^3 \epsilon_{\perp}^2} \left(\frac{\epsilon(\omega) - 1}{\epsilon(\omega) + 2} \mp \frac{3A(\omega)}{(\epsilon(\omega) + 2)^2} \right) \cos(k_{\pm}d), \quad (5)$$

where we use the notation of Ref. 5. These equations are accurate to first order in $A(\omega)$. The longitudinal (L or z) mode is unaffected by the magnetic field. Since the frequency-dependences of both $\epsilon(\omega)$ and $A(\omega)$ are assumed known, these equations represent implicit relations between ω and k_{\pm} for these T waves.

If $\mathbf{B} \parallel \hat{x}$ while both \hat{n} and the chain of particles are parallel to \hat{z} , then the y and z polarized waves are coupled. The dispersion relations are obtained as solutions to the coupled equations

$$\begin{aligned} p_{0y} &= \frac{-2a^3}{3d^3} \left[\frac{\epsilon_{\parallel}}{\epsilon_{\perp}^2} t_{yy} p_{0y} - \frac{2}{\epsilon_{\perp}} t_{yz} p_{0z} \right] \cos(kd), \\ p_{0z} &= \frac{-2a^3}{3d^3} \left[-\frac{\epsilon_{\parallel}}{\epsilon_{\perp}^2} t_{yz} p_{0y} - \frac{2}{\epsilon_{\perp}} t_{zz} p_{0z} \right] \cos(kd). \end{aligned} \quad (6)$$

These y and z modes are uncoupled from the x modes.

If we solve this pair of equations for p_{0y} and p_{0z} for a given k , we obtain a nonzero solution only if the determinant of the matrix of coefficients vanishes. For a given real frequency ω , there will, in general, be two solutions for $k(\omega)$ which decay in the $\pm z$ direction. These correspond to two branches of propagating plasmon (or plasmon polariton) waves, with dispersion relations which we may write as $k_{\pm}(\omega)$. The frequency dependence appears in t_{yz} , t_{yy} , and t_{zz} , which depend on ω [through $\epsilon(\omega)$ and $A(\omega)$]. However, unlike the case where the magnetic field is parallel to the z axis, the waves are elliptically rather than circularly polarized.

B. Cholesteric liquid crystal

We now consider immersing the chain of semiconductor nanoparticles in a CLC in the presence of a static magnetic field with $\mathbf{B} \parallel \hat{z}$ and the chain. A CLC can be thought of as an NLC whose director is perpendicular to a rotation axis (which we take to be \hat{z}), and which spirals about that axis with a pitch angle α per interparticle spacing. For a CLC, if we include only interactions between nearest-neighbor dipoles, the coupled dipole equation [Eq. (3)] takes the form

$$\tilde{p}_n = -\frac{4\pi a^3}{3} \left[\hat{\mathbf{R}}^{-1}(z_1) \hat{\mathbf{t}} \hat{\mathcal{G}} \cdot \tilde{p}_{n+1} + \hat{\mathbf{R}}(z_1) \hat{\mathbf{t}} \hat{\mathcal{G}} \cdot \tilde{p}_{n-1} \right], \quad (7)$$

as is shown in Refs. 5 and 6. Here, $\tilde{p}_n = \mathbf{R}_n(z) p_n$ and $\mathbf{R}_n(z)$ is a 2×2 rotation matrix for the director $\hat{n}(z)$. If $\mathbf{B} \parallel \hat{z}$, the two T branches are coupled. One can write a 2×2 matrix equation for the coupled dipole equations in the rotated x and y directions. This equation is found to be

$$\tilde{\mathbf{p}}_0 = -\frac{2a^3}{3d^3} \hat{\mathbf{M}}(k, \omega) \cdot \tilde{\mathbf{p}}_0, \quad (8)$$

where $\tilde{\mathbf{p}}_0$ is the rotated two-component column vector whose components are \tilde{p}_{x0} and \tilde{p}_{y0} . The components of the matrix $\hat{\mathbf{M}}(k, \omega)$ are found to be

$$\begin{aligned} M_{xx} &= \epsilon_1 [t_{xx} \cos(kd) \cos(\alpha d) + it_{xy} \sin(kd) \sin(\alpha d)], \\ M_{yy} &= \epsilon_2 [t_{yy} \cos(kd) \cos(\alpha d) + it_{xy} \sin(kd) \sin(\alpha d)], \\ M_{xy} &= \epsilon_2 [t_{xy} \cos(kd) \cos(\alpha d) - it_{yy} \sin(kd) \sin(\alpha d)], \\ M_{yx} &= \epsilon_1 [it_{xx} \sin(kd) \sin(\alpha d) - t_{xy} \cos(kd) \cos(\alpha d)], \end{aligned} \quad (9)$$

where $\epsilon_1 = \frac{\epsilon_{\parallel}^{1/2}}{\epsilon_{\perp}^{3/2}}$ and $\epsilon_2 = \frac{1}{\sqrt{\epsilon_{\perp} \epsilon_{\parallel}}}$. The dispersion relations for the two T waves are the non-trivial solutions to the secular equation formed from Eqs. (8) and (9).

The most interesting result emerging from Eqs. (8) and (9) is that the dispersion relations are *non-reciprocal*, i.e., $\omega(k) \neq \omega(-k)$, in general. The magnetic field appears only in the off-diagonal elements t_{xy} and t_{yx} , which are linear in the field except for very large fields. The terms involving t_{xy} and t_{yx} in Eq. (9) are multiplied by $\sin(kd)$ and thus change sign when k changes sign. Thus, the secular equation determining $\omega(k)$ is not even in k , implying that the dispersion relations are non-reciprocal. The non-reciprocal nature of the dispersion relations disappears at $B = 0$ even though the off diagonal terms of $\mathbf{M}(k, \omega)$ are still nonzero, because $\sin(kd)$ appears only to second order. Also, when the host dielectric is an NLC, the non-reciprocity vanishes because the rotation angle $\alpha = 0$ and all terms proportional to $\sin(kd)$ vanish, even at finite \mathbf{B} .

For a finite \mathbf{B} , the difference in magnitude of wave number between a right-moving or left-moving wave is

$$\Delta k_i(\omega) = |\text{Re}(k_{i,L})| - |\text{Re}(k_{i,R})|, \quad (10)$$

where $i = 1, 2$ for the two elliptical polarizations and L or R denotes the left-moving or right-moving branch. If, for example, $\Delta k(\omega) \neq 0$, then the left- and right-moving waves have different magnitudes of wave numbers for a given frequency and are non-reciprocal.

C. Faraday rotation and ellipticity

By solving for $k(\omega)$ using either Eq. (5) or Eq. (6) for an NLC, or Eq. (8) for a CLC, one finds that the two modes polarized perpendicular to \mathbf{B} and propagating along the nanoparticle chain have, in general, different wave vectors. For the NLC, we denote these solutions $k_+(\omega)$ and $k_-(\omega)$, while for the CLC, we denote them $k_1(\omega)$ and $k_2(\omega)$.

We first discuss the case of an NLC host and $\mathbf{B} \parallel \hat{z}$. Then, the two solutions represent left- and right-circularly polarized waves propagating along the chain. A linearly polarized mode therefore represents an equal-amplitude mixture of the two circularly polarized modes. This mixture undergoes a *rotation* of the plane of polarization as it propagates down the chain and is analogous to the usual Faraday effect in a *bulk* dielectric. The angle of rotation per unit chain length may be written as

$$\theta(\omega) = \frac{1}{2} [k_+(\omega) - k_-(\omega)]. \quad (11)$$

In the absence of damping, θ is real. If τ is finite, the electrons in each metal or semiconductor particle will experience damping, leading to an exponential decay of the plasmonic waves propagating along the chain. This damping is automatically included in the above formalism and can be seen most easily if only nearest neighbor coupling is included. The quantity,

$$\theta(\omega) = \theta_1(\omega) + i\theta_2(\omega), \quad (12)$$

is the *complex* angle of rotation per unit length of a linearly polarized wave propagating along the chain of particles. $\text{Re}[\theta(\omega)]$ represents the angle of rotation of a linearly polarized wave (per unit length of chain), and $\text{Im}[\theta(\omega)]$ is the corresponding Faraday ellipticity, i.e., the amount by which the initially linearly polarized wave becomes elliptically polarized as it propagates along the chain.

In the case of a CLC host, neither of the two T modes is circularly polarized in general. Thus, the propagation of a linearly polarized wave along the chain cannot be simply interpreted in terms of Faraday rotation.

III. NUMERICAL ILLUSTRATIONS

We now numerically evaluate the dispersion relations presented in Sec. II assuming the host is the liquid crystal known as E7. This liquid crystal was described by Müller *et al.*,¹⁵ from whom we take the dielectric constants ϵ_{\parallel} and ϵ_{\perp} . We first consider the case of an NLC host with both the director and an applied magnetic field parallel to the chain axis \hat{z} . To illustrate the predictions of our simple expressions, we take $a/d = 1/3$, and assume a magnetic field such that the ratio $\omega_c/\omega_p = 0.07$ or 0.007 as indicated in the figures. For a typical plasma frequency of $\sim 10^{13} \text{ s}^{-1}$, the ratio of 0.007 would correspond to a magnetic induction of $B \sim 2T$. We consider both the undamped and damped cases; in the latter, we choose $\omega_p\tau = 100$. For propagating waves, we choose solutions for which $\text{Im}[k_{\pm}] > 0$ so that these waves decay to zero, as expected, when $z \rightarrow \infty$ when $\text{Re } k > 0$.

The calculated dispersion relations for the two circular polarizations of plasmonic wave are shown in Fig. 1 with and without single-particle damping. The splitting between the two circularly polarized T waves is too small to be seen on the scale of this plot. The difference can be seen through its effect on the Faraday rotation angle, which is shown in Fig. 2. In this, and all subsequent plots, we have calculated far more points than are shown in the Figure, so that effectively the entire range $0 < kd < \pi$ is included.

In Fig. 2, we plot the corresponding quantity $\theta(k)d$, the rotation angle for a distance equal to one interparticle spacing. When there is no damping, we find that the real part of θd is very small and that the imaginary part is zero. Both become larger when damping is included, as we do here by setting $\omega_p\tau = 100$. Even in this case, neither $\text{Re}[\theta(\omega)d]$ nor $\text{Im}[\theta(\omega)d]$ exceed about 0.005 rad, showing that a linear incident wave is rotated only slightly over a single particle spacing (by about $1/4^\circ$ per interparticle spacing for the chosen parameters). If we assume that the wave *intensity* has an exponential decay length of no more than around 20 interparticle spacings in realistic chains, the likely Faraday rotation

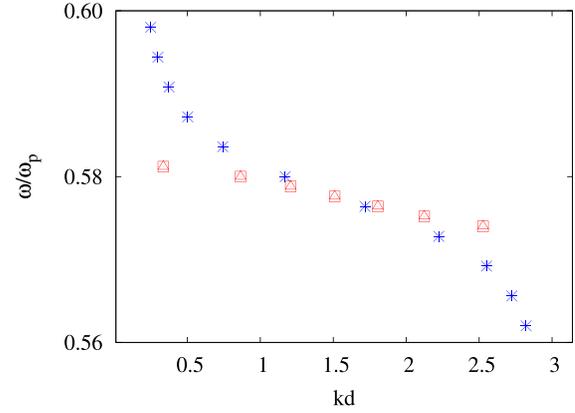


FIG. 1. Blue symbols (x 's and $+$'s): Dispersion relations for left- (x) and right- $(+)$ circularly polarized T plasmon waves propagating along a chain of nanoparticles immersed in a NLC with both the director and a magnetic field parallel to a chain. The particles are described by a Drude dielectric function with $\omega_p\tau = 100$ and $\omega_c/\omega_p = 0.07$. Red symbols (open squares and triangles): Same as the blue symbols, but assuming no single-particle damping ($\omega_p\tau = \infty$). In both cases, the splitting between left and right circularly polarized waves is not visible on the scale of the figure (but the rotation is visible in Fig. 2). For $\omega_p = 5.0 \times 10^{12} \text{ s}^{-1}$, the chosen ω_c/ω_p corresponds to $B \sim 2T$.

of such a wave will only be 3° – 4° over this distance. The present numerical calculations also suggest that $\theta(k)d$ is very nearly linear in B for a given k , so a larger rotation could be attained by increasing B ; it can also be increased if the electron density is reduced.

For a chain of Drude particles in an NLC where $B \perp \hat{z}$, we find, using the same parameters and requirements as the previous case, that the two non-degenerate waves (one an L and the other a T wave) become mixed when $B \neq 0$. The dispersion relations, again with and without damping, are plotted in Fig. 3. When compared to the previous work in Ref. 5 without the presence of damping, the dispersion relations in Fig. 3 are modified because of the finite damping and presence of the magnetic field. Decreasing the electron density of the metal or semiconductor at fixed \mathbf{B} increases the interaction of the coupled L and T mode near their crossing point

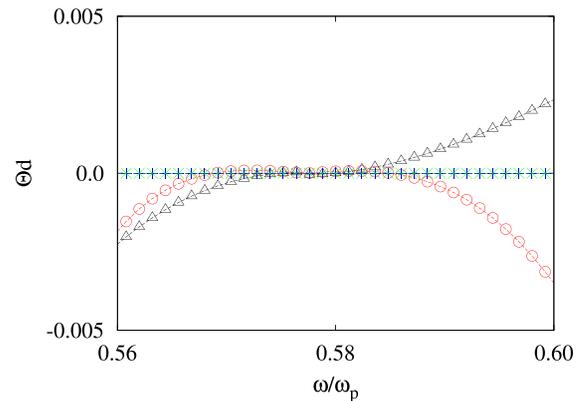


FIG. 2. Real and imaginary parts of θd , the rotation angle per interparticle spacing (in radians), as a function of frequency, assuming $\omega_c/\omega_p = 0.07$. Blue $+$'s (real part) and green x 's (imaginary part of θd): Drude model with no damping ($\omega_p\tau \rightarrow \infty$). Black triangles (real part) and red circles (imaginary part of θd): Drude model with finite damping ($\omega_p\tau = 100$). In both cases, the magnetic field and the director of the NLC are assumed parallel to the chain axis, as in Fig. 1. The dotted lines merely connect the points.

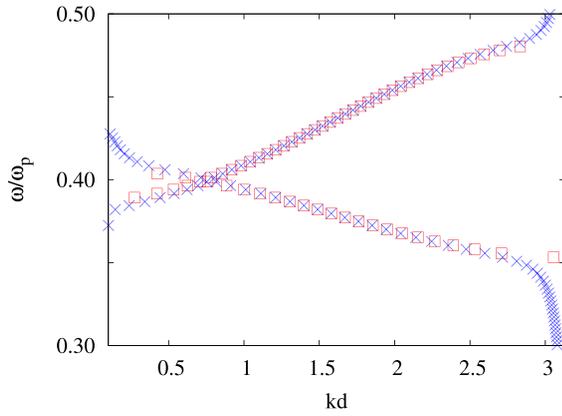


FIG. 3. Red open squares: dispersion relations for plasmon waves elliptically polarized in the yz plane and propagating along a chain of nanoparticles described by a Drude dielectric function and assuming no damping. The chain is assumed immersed in an NLC with director parallel to the chain (\hat{z} , $\mathbf{B} \parallel \hat{x}$ and $\omega_c/\omega_p = 0.007$). Blue x 's: same as red open squares, but assuming single-particle damping corresponding to $\omega_p\tau = 100$. For $\omega_p = 1.0 \times 10^{13} \text{ s}^{-1}$, the chosen ω_c/ω_p corresponds to about 2 T.

$kd = 0.7$, although this is not visible in the figure for the chosen parameters.

We find that the magnetic field appears to “repel” the two dispersion relations near their crossing point, although this is again not visible in the figure for a magnetic field of 2 T. For fixed electron density n_e , we find that this band gap between the mixed L and T waves increases monotonically with increasing magnetic field. Also, for a fixed magnetic field, the band gap *decreases* monotonically with increasing electron density. In the [Appendix](#), we show analytically that the band gap varies linearly with the magnitude of the magnetic field. We have not obtained a simple analytical relationship between the gap and the electron density for fixed magnetic field, but we do show numerically in the [Appendix](#) that this gap does increase monotonically with decreasing electron density.

Finally, we discuss the case of a chain parallel to the z axis, subjected to a magnetic field along the z axis, and immersed in a CLC whose twist axis is also parallel to \hat{z} . Using the same host dielectric constants given above and a twist angle of $\alpha d = \pi/6$, we show in Fig. 4(a) the resulting dispersion relations, i.e., ω/ω_p plotted against $|kd|$, for the two transverse branches. In particular, we show both transverse branches for a right-moving wave, displayed as black (+) symbols, and a left-moving wave, displayed as red (x) symbols, giving a total of 4 plots shown in Fig. 4(a). The separation between the two T branches is on the order of $0.05 \omega/\omega_p$ for all k .

In Fig. 4(b), we plot the corresponding difference in wave number between the left- and right-moving waves as $\Delta k_i(\omega)d$. Since $\Delta k_i(\omega)d$ is nonzero in a wide frequency range, the wave propagation is indeed non-reciprocal in this range. One-way wave propagation clearly does occur in part of this range. Such propagation occurs when, at particular frequencies, waves can propagate only in one of the two directions. From Fig. 4(a), we can see that for the upper dispersion relation, only the right-hand-moving wave can propagate near $kd = \pi$, whereas for the lower one, only the

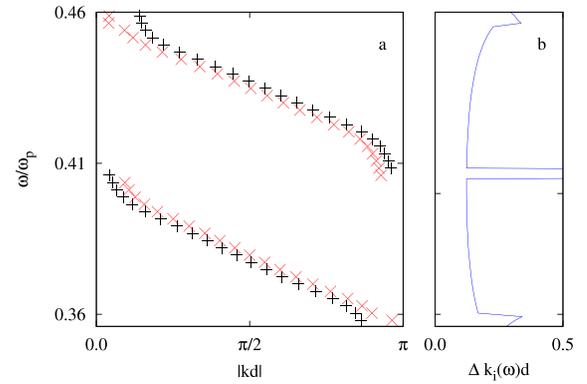


FIG. 4. (a). Black + symbols: the two dispersion relations for right-moving transverse plasmon waves propagating along a chain of Drude nanoparticles immersed in a CLC host with $\mathbf{B} \parallel \hat{z}$, plotted as a function of $|kd|$. Red x 's: same quantities but for left-moving plasmon waves. We assume that $\omega_c/\omega_p = 0.007$, $\omega_p\tau = 100$, and the twist angle $\alpha d = \pi/6$. (b). The absolute value $|\Delta k_i d|$ of the difference $\Delta k_i d = |Re(k_{iL}d)| - |Re(k_{iR}d)|$, between the normalized wave vectors for left-propagating and right-propagating modes of the two branches, as given by Eq. (10), as functions of frequency. L, R refer to the left-moving or right-moving waves, and i ($i = 1, 2$) labels the two branches for each direction. Note that a non-zero value of $|\Delta k_i(\omega)d|$ implies that for a given frequency the left- and right-traveling waves have different wave vectors. Numerically, we find that $|\Delta k_i(\omega)d|$ is independent of i . There is a gap between the two curves in Fig. 4(b). This gap corresponds to a region where the waves propagate in a single direction only. These *one-way modes* occur in the upper branch near $|kd| = \pi$, where there is a small region where only the red branch has solutions, and in the lower branch near $|kd| = 0$, where only the black branch has solutions. One-way wave propagation occurs only in the region between the two horizontal lines in Fig. 4(b).

left-hand-moving wave propagates near $kd = 0$. Thus, there is a gap in the plot of $\Delta k_i(\omega)d$ near $\omega/\omega_p = 0.41$, within which there is only one-way wave propagation. In Fig. 4(b), the boundaries of the frequency band for one-way propagation are indicated by the two horizontal lines.

IV. DISCUSSION

The present numerical calculations omit several potentially important factors which could alter the numerical results. The first of these are the effects of particles beyond the nearest neighbors on the dispersion relations.⁹ We believe that these further neighbors will mainly modify the details of the dispersion relations without changing the qualitative features introduced by the magnetic field and the NLC or CLC host. Another omitted factor is the (possibly large) influence of the particles in disrupting the director orientation of the liquid crystalline host,^{16–20} whether NLC or CLC. This could be quite important in modifying the dielectric properties of the host liquid near the particles, and could also cause the positions of the particles themselves to be disturbed, depending on whether they are somehow held in place. Even though these effects could be quite substantial, we believe that the qualitative effects found in the present calculations, notably the regime of one-way propagation found for certain frequencies in a CLC host, should still be present. We hope to investigate these effects in future work. Finally, it is known that radiative damping,²¹ not included in the quasistatic approximation, can have a significant effect on the dispersion relations at special frequencies. Once again, however, we believe that the qualitative effects

discussed in this paper should still be present even if radiation damping is included. Thus, we believe that our calculations do qualitatively describe the combined effects of a liquid crystalline host and an applied magnetic field on the surface plasmon dispersion relations.

It should be noted that the magnetic field effects described in this paper are numerically small, for the parameters investigated. The smallness is caused mainly by the small value of the ratio ω_c/ω_p , taken here as 0.07 or 0.007 depending on the electron density used in the calculation. To increase this ratio, one could either increase ω_c (by raising the magnetic field strength) or decrease ω_p (by reducing the free carrier density in the particle). For the case of a particle chain in a CLC host, any change which increases ω_c/ω_p will increase Δk , leading to a broader frequency range for one-way wave propagation.

In summary, we have calculated the dispersion relations for plasmonic waves propagating along a chain of semiconducting or metallic nanoparticles immersed in a liquid crystal and subjected to an applied magnetic field. For a magnetic field parallel to the chain and director axis of the NLC, a linearly polarized wave is Faraday-rotated by an amount proportional to the magnetic field strength. For a CLC host and a magnetic field parallel to the chain, the transverse wave solutions become non-reciprocal (left- and right-traveling waves having different dispersion relations) and there are frequency ranges in which waves propagate only in one direction. Thus, plasmonic wave propagation can be tuned, either by a liquid crystalline host or a magnetic field, or both. In the future, it may be possible to detect some of these effects in experiments, and to use some of the predicted properties for applications, e.g., in optical circuit design.

ACKNOWLEDGMENTS

This work was supported by the Center for Emerging Materials at The Ohio State University, an NSF MRSEC (Grant No. DMR-1420451) and N.A.P. was additionally supported by the Belgian Fonds National de la Recherche Scientifique FNRS under Grant No. PDR T.1077.15-1/7.

APPENDIX: ANALYSIS OF BAND GAP IN FIG. 3

In this Appendix, we show that the two bands shown in Fig. 3, which in zero magnetic field cross near $kd = 0.7$, are “repelled” in a finite magnetic field $\mathbf{B} \parallel \hat{x}$ resulting in the opening of a band gap which, at fixed electron density n_e , is proportional to $|\mathbf{B}|$. We will also show numerically that this band gap at fixed $|\mathbf{B}|$ is monotonically increasing with decreasing n_e .

The dispersion relations for the coupled y and z polarized waves are obtained from the two coupled equations (6). They have non-trivial solutions when the determinant of the matrix of coefficients vanishes, i.e.,

$$\left[1 + \frac{2a^3 \epsilon_{\parallel}}{3d^3 \epsilon_{\perp}^2} t_{yy}(\omega) \cos(kd) \right] \left[1 - \frac{4a^3}{3d^3 \epsilon_{\perp}} t_{zz}(\omega) \cos(kd) \right] - \frac{8a^6 \epsilon_{\parallel}}{9d^6 \epsilon_{\perp}^3} t_{yz}^2(\omega) \cos^2(kd) = 0. \quad (\text{A1})$$

We first consider the case of zero magnetic field. In this case, the off-diagonal components of the t -matrix, namely, $t_{yz} = -t_{zy}$, both vanish. The dispersion relations are then given by

$$F_1(k, \omega) \equiv 1 + \frac{2a^3 \epsilon_{\parallel}}{3d^3 \epsilon_{\perp}^2} t_{yy}(\omega) \cos(kd) = 0 \quad (\text{A2})$$

and

$$F_2(k, \omega) \equiv 1 - \frac{4a^3}{3d^3 \epsilon_{\perp}} t_{zz}(\omega) \cos(kd) = 0. \quad (\text{A3})$$

The two bands will be degenerate when $F_1(k, \omega) = F_2(k, \omega)$, or equivalently

$$\frac{\epsilon_{\parallel}}{\epsilon_{\perp}^2} t_{yy}(\omega) + \frac{1}{\epsilon_{\perp}} t_{zz}(\omega) = 0. \quad (\text{A4})$$

Equation (A4) gives the frequency of the degeneracy, which we denote ω_0 . The corresponding wave vector k_0 of the degeneracy is determined by either

$$F_1(k_0, \omega_0) = 0 \quad (\text{A5})$$

or

$$F_2(k_0, \omega_0) = 0. \quad (\text{A6})$$

Now, we consider Eq. (A1) with non-zero magnetic field, i.e., finite $t_{yz}(\omega)$. For $k = k_0$, assuming that the band energies ω are close to their zero-field value ω_0 , we can expand F_1 and F_2 in Taylor series as $F_i(k_0, \omega) \sim (\omega - \omega_0) F'_i(k_0, \omega_0)$ for $i = 1, 2$, where $F'_i(k_0, \omega_0) = [\partial F_i(k_0, \omega) / \partial \omega]_{\omega=\omega_0}$. Again to lowest order in B , we can write $t_{yz}(\omega) \sim t_{yz}(\omega_0)$. Then, the solutions to Eq. (A1) are given by

$$F'_1(k_0, \omega_0) F'_2(k_0, \omega_0) (\omega - \omega_0)^2 = \frac{8a^6 \epsilon_{\parallel}}{9d^6 \epsilon_{\perp}^3} t_{yz}^2(\omega_0) \cos^2(k_0 d) \quad (\text{A7})$$

or

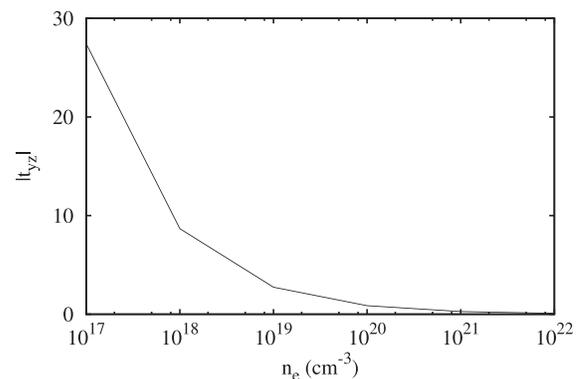


FIG. 5. Semilog plot of the (dimensionless) matrix element $|t_{yz}|$ as a function of electron density n_e (in cm^{-3}) for a fixed magnetic field of $B = 2T$. The orientations of the magnetic field and chain are the same as in Fig. 3. Since the gap equals a density-independent constant multiplied by $|t_{yz}|$, this plot shows that the gap also is monotonically decreasing with increasing n_e at fixed $|\mathbf{B}|$. In all cases, we assume $\omega_p \tau = 100$.

$$\omega(B) - \omega_0 = \pm \left\{ \frac{8a^6 \epsilon_{\parallel}}{9d^6 \epsilon_{\perp}^3} t_{yz}^2(\omega_0) \cos^2(k_0 d) / |F'_1(k_0, \omega_0) F'_2(k_0, \omega_0)| \right\}^{1/2}. \quad (\text{A8})$$

Here, $\omega(B)$ represents one of the two band energies at $k = k_0$. Since $t_{yz}(\omega_0)$ is proportional to B (see below), and since the other terms in Eq. (A8) do not depend on magnetic field, Eq. (A8) shows that the splitting between these two band energies at $k = k_0$ is proportional to $|\mathbf{B}|$.

To show that $t_{yz}(\omega_0) \propto B$, we can calculate t_{yz} (and the other components of \mathbf{t}) from Eqs. (1), (2), and (4). The result, to lowest order in $\delta\epsilon_{yz}(\omega)$, is

$$t_{yz}(\omega) = \frac{\delta\epsilon_{yz}(\omega)}{(1 - \Gamma_{yy}\delta\epsilon_{yy}(\omega))(1 - \Gamma_{zz}\delta\epsilon_{zz}(\omega))}. \quad (\text{A9})$$

Since $\delta\epsilon_{yz}(\omega) \propto A(\omega)$, we see that $t_{yz}(\omega) \propto \omega_c \propto B$. Hence, the splitting between the two bands at $k = k_0$ is proportional to $|\mathbf{B}|$.

In addition to the magnetic field dependence, one can also numerically determine the dependence of the band gap on the electron density n_e . In Fig. 5, we show the calculated matrix element $|t_{yz}|$ as a function of n_e for $|\mathbf{B}| = 2T$; it increases monotonically with decreasing n_e over several orders of magnitude of n_e . For the magnitudes of $|\mathbf{B}|$ and n_e considered in Fig. 3, this splitting is not visible on the scale of the Figure, but we have verified numerically that this splitting is definitely present for finite magnetic field and finite electron density.

In the plot of Fig. 5, we assume $\omega_p \tau = 100$ for all densities. Since ω_p depends on n_e , we are effectively assuming that τ also depends on n_e . We have done similar calculations,

however, with τ independent of n_e , and also with no damping ($\tau = \infty$). In all cases, we find the same monotonic decrease in gap with increasing n_e , though quantitatively the behavior is different.

- ¹S. A. Maier, M. L. Brongersma, P. G. Kik, S. Meltzer, A. A. G. Requicha, and H. A. Atwater, *Adv. Mater.* **13**, 1501 (2001).
- ²S. A. Maier, P. G. Kik, H. A. Atwater, S. Meltzer, E. Harel, B. E. Koel, and A. A. G. Requicha, *Nat. Mater.* **2**, 229 (2003).
- ³Z. Y. Tang and N. A. Kotov, *Adv. Mater.* **17**, 951 (2005).
- ⁴S. Y. Park, A. K. R. Lytton-Jean, B. Lee, S. Weigand, G. C. Schatz, and C. A. Mirkin, *Nature* **451**, 553 (2008).
- ⁵N. A. Pike and D. Stroud, *J. Opt. Soc. Am. B* **30**, 1127–1134 (2013).
- ⁶N. A. Pike and D. Stroud, “Plasmonics: Metallic nanostructures and their optical properties XI,” *Proc. SPIE* **8809**, 880910 (2013).
- ⁷J. A. Fauchaux, A. L. Stanton, and P. K. Jain, *J. Phys. Chem. Lett.* **5**, 976 (2014).
- ⁸S.-W. Hsu, K. On, and A. R. Tao, *J. Am. Chem. Soc.* **133**, 19072 (2011).
- ⁹M. L. Brongersma, J. W. Hartman, and H. A. Atwater, *Phys. Rev. B* **62**, R16356 (2000).
- ¹⁰Z. Yu, G. Veronis, Z. Wang, and S. Fan, *Phys. Rev. Lett.* **100**, 023902 (2008).
- ¹¹M. Dixit and D. Stroud, *Appl. Phys. Lett.* **104**, 061604 (2014).
- ¹²Y. Mazor and B. Z. Steinberg, *Phys. Rev. B* **86**, 045120 (2012).
- ¹³Y. Hadad and B. Z. Steinberg, *Phys. Rev. Lett.* **105**, 233904 (2010).
- ¹⁴Z. Wang, Y. D. Chong, J. Joannopoulos, and M. Soljacic, *Phys. Rev. Lett.* **100**, 013905 (2008).
- ¹⁵J. Müller, C. Sönnichsen, H. von Poschinger, G. von Plessen, T. A. Klar, and J. Feldmann, *Appl. Phys. Lett.* **81**, 171 (2002).
- ¹⁶T. C. Lubensky, D. Pettey, N. Currier, and H. Stark, *Phys. Rev. E* **57**, 610 (1998).
- ¹⁷P. Poulin and D. A. Weitz, *Phys. Rev. E* **57**, 626 (1998).
- ¹⁸H. Stark, *Phys. Rep.* **351**, 387 (2001).
- ¹⁹R. D. Kamien and T. D. Powers, *Liq. Cryst.* **23**, 213 (1997).
- ²⁰D. W. Allender, G. P. Crawford, and J. W. Doane, *Phys. Rev. Lett.* **67**, 1442 (1991).
- ²¹W. H. Weber and G. W. Ford, *Phys. Rev. B* **70**, 125429 (2004).