Tunable plasmon polaritons in arrays of interacting metallic nanoparticles

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We consider a simple cubic array of metallic nanoparticles, each supporting a localized surface plasmon, and study theoretically the properties of the propagating plasmon polaritons resulting from the near-field dipolar interaction between the nanoparticles and the coupling to light. Remarkably, we show that the polaritonic band gap, the dielectric function and the reflection coefficient of the metamaterial can be significantly modulated by the polarization of light. We unveil how such an anisotropic behavior in a plasmonic metamaterial is crucially mediated by the dipolar interactions between the nanoparticles despite the symmetry of the underlying lattice.

Our results thus pave the way towards the realization of tunable quantum plasmonic metamaterials presenting interaction-driven birefringence.

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Introduction.—The ability to manipulate light at subwavelength scales beyond the diffraction limit of traditional optics is at the very heart of the present research in plasmonic metamaterials [1, 2] and transformation optics [3]. Metamaterials have already been shown to exhibit exciting new properties such as negative refractive index [4–6], perfect lensing [7, 8], electromagnetic invisibility cloaking [9–11], “trapped rainbow” slow light [12], and the ability to perform mathematical operations (“metamaterial analog computing”) [13].

In this paper we explore the possibility to design novel quantum plasmonic metamaterials with a highly tunable optical response based on arrays of interacting metallic nanoparticles.

While isolated metallic nanoparticles have been already successfully employed to confine electromagnetic radiation down to the nanometer scale, the focus is now shifting to the exploration of ordered plasmonic arrays of metallic nanoparticles. In these systems the interactions between the nanoparticles lead to dramatic changes in the collective plasmonic properties of the array as compared to those of the individual nanoparticles, opening up new perspectives for confining and guiding light at subwavelength scales [14–20]. Along this direction, we have recently exploited the tunability of the near-field interactions in two-dimensional honeycomb arrays of metallic nanoparticles. This system exhibits remarkable effects, including the emergence of tunable chiral Dirac-like collective plasmons [21] that mimic some of the unique features of electrons in graphene, such as a nontrivial Berry phase [22, 23] and the absence of backscattering off impurities [24].

The optical response of plasmonic metamaterials is determined by their dielectric function, which results in the reflection and transmission coefficients. In order to calculate the dielectric function it is crucial to identify the eigenmodes responsible for transporting electromagnetic radiation in arrays of metallic nanoparticles. These modes stem from the coupling between light and collective plasmons which gives rise to new quasiparticles termed plasmon polaritons. First studied in the late 1950’s by Fano [25] and Hopfield [26] in the context of excitons in bulk solids, polaritons were shown to represent the relevant modes to describe the light-matter interaction and the subsequent absorption processes in periodic atomic arrays. In the context of plasmonics, surface plasmon polaritons along metal-dielectric interfaces were widely studied [1, 2]. In contrast, the physical properties of plasmon polaritons in arrays of metallic nanoparticles are still to be explored. Such plasmonic metamaterials can be realized using electron beam [18] or nanosphere lithography [20] resulting in one- and two-dimensional arrays of nanoparticles, or by means of self-organization of nanoparticles capped with molecular linkers such as thiol chains [27] and DNA [28] for three-dimensional structures. Undoubtedly, there is a need for investigating plasmon polaritons in arrays of interacting metallic nanoparticles, where the control of the microscopic interactions is expected to lead to dramatic effects which would otherwise be absent in noninteracting systems. This is the purpose of the present theoretical paper.

As a proof of concept, we hence explore plasmon polaritons in a simple cubic array of metallic nanoparticles. We show that, due to interaction effects, an anisotropic response of the plasmonic array becomes apparent. Specifically, we unveil analytically the plasmon polariton band structure and show that it is highly tunable with the polarization of light, resulting in a plasmon polariton band gap that can be tuned by about 50%.

Our prediction should thus be clearly observable in the frequency- and wavevector-dependent dielectric function of the metamaterial, resulting in an interaction-driven birefringence despite the symmetric lattice structure of the array. Our analytical treatment can be easily generalized to other three-dimensional lattices that are thus expected to exhibit a similar tunable optical response.

Arrays of interacting metallic nanoparticles.—We consider an ensemble of identical spherical metallic nanoparticles of radius $r$, each containing $N_e$ valence electrons, and forming a simple cubic lattice with $N$ lattice sites and lattice constant $a$. Each nanoparticle supports a localized surface plasmon which can be modelled as a collective dipolar excitation of the electronic center of mass at the Mie frequency $\omega_0$ [29].
For a nanoparticle in vacuum, the latter takes the simple form
\[ \omega_0 = (N_e e^2 / 4 \pi \epsilon_0 m_e \omega_0^3)^{1/2}, \]
where \(-e \) and \( m_e \) are the electron charge and mass, respectively, and where \( \epsilon_0 \) is the vacuum permittivity. The noninteracting part of the Hamiltonian describing the independent localized surface plasmons on the cubic lattice sites reads [30–32]
\[ H_0 = \sum_{\mathbf{R}} \left[ \frac{\Pi^2(\mathbf{R})}{2M} + \frac{M}{2} \omega_0^2 h^2(\mathbf{R}) \right], \tag{1} \]
where \( h(\mathbf{R}) \) denotes the electronic center-of-mass displacement corresponding to a nanoparticle located at position \( \mathbf{R} \), \( \Pi(\mathbf{R}) \) is the conjugated momentum to \( h(\mathbf{R}) \) and \( M = N_e m_e \) is the total electronic mass per nanoparticle.

If the wavelength associated with the resonance frequency of each localized surface plasmon is much larger than the interparticle distance \( a \) and when \( r \lesssim a/3 \), each localized plasmon can be considered as a point dipole with dipole moment \( \mathbf{p} = -Q h(\mathbf{R}) \mathbf{p} \) and total charge \( Q = N_e e \) which interacts with the neighboring ones through dipole-dipole interactions [16, 19]. In the quasistatic (near-field) approximation, the interaction between two dipoles \( \mathbf{p} \) and \( \mathbf{p}' \) located at \( \mathbf{R} \) and \( \mathbf{R}' \), respectively, reads
\[ V_{\text{dip}} = \frac{\mathbf{p} \cdot \mathbf{p}' - 3(\mathbf{p} \cdot \mathbf{n})(\mathbf{p}' \cdot \mathbf{n})}{4\pi \epsilon_0 |\mathbf{R} - \mathbf{R}'|^3}, \tag{2} \]
with \( \mathbf{n} = (\mathbf{R} - \mathbf{R}')/|\mathbf{R} - \mathbf{R}'| \). In what follows, we impose that, due to the electric field associated with light, all nanoparticles are polarized in the same direction \( \mathbf{p} = \sin \theta \cos \varphi \mathbf{x} + \sin \theta \sin \varphi \mathbf{y} + \cos \theta \mathbf{z} \), where \( \theta \) is the angle between \( \mathbf{p} \) and \( \mathbf{z} \), and \( \varphi \) the angle between the projection of \( \mathbf{p} \) in the \( xy \) plane and \( \mathbf{x} \). From Eq. (2) the interaction Hamiltonian between localized surface plasmons thus reads
\[ H_{\text{int}} = \frac{Q^2}{8\pi \epsilon_0 a^3} \sum_{\mathbf{R}} \sum_{j=1}^{3} C_j h(\mathbf{R}) \left[ h(\mathbf{R} + \mathbf{e}_j) + h(\mathbf{R} - \mathbf{e}_j) \right], \tag{3} \]
with \( C_j = 1 - 3[\sin^2 \theta (\delta_{j1} \cos^2 \varphi + \delta_{j2} \sin^2 \varphi) + \delta_{j3} \cos^2 \theta] \) and where \( \mathbf{e}_1 = a \mathbf{x} \), \( \mathbf{e}_2 = a \mathbf{y} \) and \( \mathbf{e}_3 = a \mathbf{z} \). Only interactions between nearest neighbors are taken into account in the Hamiltonian (3) since the interactions beyond nearest neighbors do not qualitatively change the collective plasmon dispersion [33], as is the case for metallic nanoparticle arrays with other geometries [16, 21].

Collective plasmon dispersion.—Introducing the bosonic operator \( b_{\mathbf{R}} = (M \omega_0/2\hbar)^{1/2} h(\mathbf{R}) + i \Pi(\mathbf{R})/(2M \omega_0)^{1/2} \) which annihilates a localized surface plasmon at lattice site \( \mathbf{R} \) and its momentum space representation \( b_\mathbf{q} \) through \( b_{\mathbf{R}} = \sum_{\mathbf{q}} \exp(i \mathbf{q} \cdot \mathbf{R}) b_\mathbf{q} \), the Hamiltonian representing the collective plasmons, \( H_{\text{pl}} = H_0 + H_{\text{int}} \) [cf. Eqs. (1) and (3)], transforms into
\[ H_{\text{pl}} = \hbar \sum_\mathbf{q} \left[ (\omega_0 + 2\Omega f_\mathbf{q}) b_\mathbf{q}^\dagger b_\mathbf{q} + \Omega f_\mathbf{q} (b_\mathbf{q}^\dagger b_{-\mathbf{q}} + b_{-\mathbf{q}} b_\mathbf{q}) \right], \tag{4} \]
with \( \omega_{\text{pl}} = \omega_0 \sqrt{1 + 4 \Omega/\omega_0} f_\mathbf{q} \). \tag{8} \]

It is important to realize that the dispersion in Eq. (8) can be tuned by the polarization of the localized surface plasmons which enters the function \( f_\mathbf{q} \) in Eq. (5). This is illustrated in Fig. 1 which shows the collective plasmon dispersion from Eq. (8) along the high symmetry axes in the first Brillouin zone, for various polarization angles \( (\theta, \varphi) \). In the figure, \( r = a/3 \). The inset shows one eighth of the cubic first Brillouin zone together with the high symmetry points.

\[ f_\mathbf{q} = \sum_{j=1}^{3} C_j \cos(\mathbf{q} \cdot \mathbf{e}_j). \tag{5} \]

The purely plasmonic problem represented by the Hamiltonian (4) can be diagonalized by a Bogoliubov transformation. Introducing the new bosonic operator
\[ b_\mathbf{q} = C_1 b_\mathbf{q} - C_2 b_{-\mathbf{q}}^\dagger - C_3 b_{-\mathbf{q}}^\dagger b_{-\mathbf{q}} \tag{6} \]

with \( C_1 = \cosh \vartheta_0 b_\mathbf{q} - \sinh \vartheta_0 b_{-\mathbf{q}}^\dagger \), and when \( \vartheta_0 = 2^{-1/2} [(1 + 2\Omega f_\mathbf{q}/\omega_0)/(1 + 4\Omega f_\mathbf{q}/\omega_0)^{1/2} + 1]^{1/2} \) and \( \sinh \vartheta_0 = -2^{-1/2} \text{sign}(f_\mathbf{q}) [(1 + 2\Omega f_\mathbf{q}/\omega_0)/(1 + 4\Omega f_\mathbf{q}/\omega_0)^{1/2} - 1]^{1/2} \) leads to
\[ H_{\text{pl}} = \sum_\mathbf{q} \hbar \omega_{\text{pl}} b_\mathbf{q}^\dagger b_\mathbf{q}, \tag{7} \]
with the collective plasmon dispersion
\[ \omega_{\text{pl}} = \omega_0 \sqrt{1 + 4 \Omega/\omega_0} f_\mathbf{q}. \tag{8} \]
Coupling to light.—The collective plasmons discussed above can be triggered by light. As realized by Fano [25] and Hopfield [26], the correct description of the coupling of elementary excitations to light in periodic systems can only be achieved by quantizing light in a cavity that has the same size as the crystal. This is a direct consequence of the translational invariance of the nanoparticle array and the subsequent conservation of photonic and plasmonic momenta. As a result, energy oscillates back and forth between the two subsystems, such that the semiclassical view of absorption processes is not appropriate. Hence, we describe the photonic modes in the cavity of volume $V = N a^3$ by the Hamiltonian

$$H_{ph} = \sum_q \hbar \omega_q^{ph} c_q \dagger c_q,$$

(9)

where $c_q$ ($c_q \dagger$) annihilates (creates) a photon with momentum $q$ and transverse polarization $\epsilon$ ($q \cdot \epsilon = 0$) and where $\omega_q^{ph} = \epsilon(q)$ is the photon dispersion with $c$ the speed of light [34]. Notice that in the expression (9), the summation over photon polarizations has been made implicit in order to simplify the notation in the sequel of the paper.

In the long-wavelength limit (dipolar approximation), the localized surface plasmons interact with the light modes through the Hamiltonian

$$H_{pl-ph} = \sum_R \left[ \frac{Q^2}{M} \mathbf{A}(R) \cdot \mathbf{A}^\dagger(R) \right] + \frac{Q^2}{2M} \mathbf{A}^\dagger(R) \mathbf{A}(R),$$

(10)

where

$$\mathbf{A}(R) = \sum_q \sqrt{\frac{\hbar}{2e_0 V \omega_q^{ph}}} \left( c_q e^{i\mathbf{q} \cdot \mathbf{R}} + c_q \dagger e^{-i\mathbf{q} \cdot \mathbf{R}} \right),$$

(11)

is the vector potential at the location $\mathbf{R}$ of the nanoparticles. The plasmon-photon interaction term (10) thus takes the form

$$H_{pl-ph} = \hbar \omega_0 \sum_q \left[ i \xi_q (\cosh \vartheta_q - \sinh \vartheta_q) \beta_q - \dagger \beta_q \right] c_q c_q \dagger c_q \dagger c_q - c_q \dagger c_q - c_q \dagger c_q - c_q \dagger c_q \dagger,$$

(12)

in terms of the Bogoliubov modes $\beta_q$ diagonalizing the purely plasmonic problem [cf. Eq. (6)]. In Eq. (12), $\xi_q = (\pi \omega_0 / \omega_q^{pl})^{1/2} (r/a)^{3/2}$ and we used $\varphi = \epsilon$. Plasmon polariton dispersion.—The total Hamiltonian $H = H_{pl} + H_{ph} + H_{pl-ph}$ [cf. Eqs. (7), (9) and (12)] can now be diagonalized by introducing the annihilation operator associated with plasmon polaritons $\gamma_q = w_q c_q + x_q \beta_q + y_q c_q \dagger - z_q \beta_q \dagger$, with $w_q$, $x_q$, $y_q$ and $z_q$ complex numbers, and imposing the diagonal form of the Heisenberg equation of motion [$\gamma_q$, $H$] = $\hbar \omega_q^{pp} \gamma_q$ [26]. This procedure yields the frequency- and wavevector-dependent dielectric function

$$\epsilon(q, \omega) = \frac{c^2 q^2}{\omega^2} = 1 + \frac{8 \pi \omega_0}{\omega_0 + 4 \pi \omega_0 f_q - \omega^2},$$

(13)

which constitutes the central result of this paper.

The plasmon polariton dispersion (14) is shown in Fig. 2 along the $\Gamma X$ direction in the first Brillouin zone for transverse light polarization (i.e., in the $yz$ plane, see the inset in Fig. 1). For wavenumbers close to the edge of the first Brillouin zone, the + and – branches of the plasmon polariton dispersion (14) asymptotically approach the light and the collective plasmon dispersion, respectively. When $|q|$ → 0, however, the + branch goes to $\omega_0^{ph} \to 0$, while the – branch tends to $\omega_0^{pp} |q| \to 0$, + $\omega_0 \sqrt{1 + 8 \pi \omega_0 / |q|}$. Thus the strong plasmon-photon coupling results in a gap of the order of $\Delta \simeq 4 \pi \omega_0$ in the plasmon polariton dispersion. This has important consequences on the optical properties of our simple cubic array of nanoparticles. Indeed, for frequencies within the band gap, no plasmon polariton can propagate in the metamaterial, such that the reflectivity of the latter is equal to one. We estimate that for an interparticle distance $a = 3 r$, the plasmonic gap $\Delta$ is about 25% of the Mie frequency $\omega_0$. For noble-metal nanoparticles, the latter lies usually in the visible range ($\omega_0 \simeq 2 - 3 eV$), yielding a plasmonic gap of about $\Delta \simeq 0.5 - 0.75 eV$. Plasmonic damping, that may mask the above gap, is evaluated [21] to be of the order of 0.1 eV for silver nanoparticles with radius $r \sim 10 n m$, such that the gap should be clearly observable in an experiment.

Remarkably, the plasmon polariton dispersion (14) can be tuned by the polarization of light through the modifica-
tion of the collective plasmon dispersion $\omega_{pl}^q$ [cf. Eq. (8) and Fig. 1]. This is illustrated in Fig. 3, which shows the plasmon polariton dispersion along the $\Gamma M$ direction in the first Brillouin zone (see the inset in Fig. 1) for two polarization angles $\xi$ defined through the transverse polarization $\hat{\epsilon} = \cos \xi \hat{\epsilon}_1 + \sin \xi \hat{\epsilon}_2$, with $\hat{\epsilon}_1 = \hat{z} \times \hat{q} / |\hat{z} \times \hat{q}|$ and $\hat{\epsilon}_2 = \hat{q} \times \hat{\epsilon}_1 / |\hat{q} \times \hat{\epsilon}_1|$. As can be seen from Fig. 3, the $-\pi$ branch of the plasmon dispersion is significantly modulated by the polarization of light. This effect results from the dependence on polarization of the collective plasmon dispersion [dashed lines in Fig. 3]. Consequently, the polaritonic band gap, defined as $\Delta = \omega_{pl}^q = \omega_{0,PP} - \max(\omega_{q,PP}^{-})$, can be significantly modulated (by about 50%) by tilting the polarization of light, as shown in the inset of Fig. 3. Considering the amplitude of the effect, this feature should be clearly measurable in an experiment. Thus the dielectric function of the metamaterial in Eq. (13) and its reflectivity are strongly sensitive to the polarization of incoming light [35]. This effect is a direct consequence of the anisotropic dipolar interaction between the metallic nanoparticles, resulting in a birefringence of the plasmonic metamaterial despite the symmetric lattice structure of our array. This is in stark contrast with conventional birefringence observed in crystals, which is usually associated with strongly asymmetric lattice structures [36].

**Conclusion.**—We have studied plasmon polaritons in a simple cubic array of interacting metallic nanoparticles. Remarkably, these plasmon polaritons resulting from the strong coupling between light and collective plasmonic excitations present a dispersion that can be significantly modulated by the polarization of light. As a result, the dielectric function and the reflection coefficient of the metamaterial can be tuned by changing the polarization of the incoming light. The consequent optical birefringence is a direct consequence of the anisotropic dipolar interactions between nanoparticles despite the symmetric lattice structure of the metamaterial.

Our results can be easily extended beyond the simple cubic lattice to other types of metastructures, such as bcc, fcc, or hcp lattices of metallic nanoparticles, paving the way to quantum plasmonic metamaterials with fully tunable optical properties.

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COLLECTIVE PLASMON DISPERSION WITH DIPOLE-DIPOLE INTERACTION BEYOND NEAREST NEIGHBORS

For simplicity of treatment and to highlight the main physical concepts behind our work, in the main text we only discuss the effects of interactions between nearest neighboring nanoparticles in the simple cubic lattice. However, as the dipole-dipole interaction decays as one over the cube of the interparticle distance, it is important to check the robustness of our results against the effect of interactions beyond nearest neighbors. In the following, we show that the plasmon dispersion discussed in the main text is not qualitatively modified by interactions beyond nearest neighbors. Specifically, we compute the collective plasmon interaction decays as one over the cube of the interparticle distance, it is important to check the robustness of our results against the effects of interactions between nearest neighboring nanoparticles in the simple cubic lattice. However, as the dipole-dipole interaction decays as one over the cube of the interparticle distance, it is important to check the robustness of our results against the effect of interactions beyond nearest neighbors. In the following, we show that the plasmon dispersion discussed in the main text is not qualitatively modified by interactions beyond nearest neighbors. Specifically, we compute the collective plasmon dispersion including next, third and fourth nearest neighbors and show that the interaction between the nearest neighbors alone captures the relevant physics of the problem.

Using the same notations as in the main text, the purely plasmonic Hamiltonian \( H_{pl} \) has to be supplemented by three extra terms \( H_{int}^{(2)}, H_{int}^{(3)} \) and \( H_{int}^{(4)} \) when the dipole-dipole interaction between next, third and fourth nearest neighbors are taken into account, viz.

\[
H_{pl} = H_0 + H_{int} + \sum_{n=2}^{4} H_{int}^{(n)},
\]

with

\[
H_{int}^{(2)} = \frac{Q^2}{8\pi\varepsilon_0(\sqrt{2}a)^3} \sum_{R} \sum_{j=1}^{3} \sum_{\sigma=\pm} C_{j\sigma}^{(2)} h(R) \left[ h(R + e_{j\sigma}^{(2)}) + h(R - e_{j\sigma}^{(2)}) \right],
\]

\[
H_{int}^{(3)} = \frac{Q^2}{8\pi\varepsilon_0(\sqrt{3}a)^3} \sum_{R} \sum_{j=1}^{2} \sum_{\sigma=\pm} C_{j\sigma}^{(3)} h(R) \left[ h(R + e_{j\sigma}^{(3)}) + h(R - e_{j\sigma}^{(3)}) \right],
\]

\[
H_{int}^{(4)} = \frac{Q^2}{8\pi\varepsilon_0(2a)^3} \sum_{R} \sum_{j=1}^{3} C_{j} h(R) \left[ h(R + 2e_j) + h(R - 2e_j) \right].
\]

Here, \( H_{int}^{(n)} \) represents the dipole-dipole interaction Hamiltonian between the \( n \)-th nearest neighbors. Besides the notation introduced in the main text, we define

\[
C_{1\sigma}^{(2)} = 1 - \frac{3}{2} \sin^2 \theta (\cos \varphi + \sigma \sin \varphi)^2,
\]

\[
C_{2\sigma}^{(2)} = 1 - \frac{3}{2} (\sin \theta \sin \varphi + \sigma \cos \theta)^2,
\]

\[
C_{3\sigma}^{(2)} = 1 - \frac{3}{2} (\sin \theta \cos \varphi + \sigma \cos \theta)^2,
\]

\[
C_{1\sigma}^{(3)} = 1 - (\sin \theta \cos \varphi + \sin \theta \sin \varphi + \sigma \cos \theta)^2,
\]

\[
C_{2\sigma}^{(3)} = 1 - (\sin \theta \cos \varphi - \sin \theta \sin \varphi + \sigma \cos \theta)^2,
\]

as well as the vectors \( e_{1\sigma}^{(2)} = e_1 + \sigma e_2, e_{2\sigma}^{(2)} = e_2 + \sigma e_3, e_{3\sigma}^{(2)} = e_1 + \sigma e_3, e_{1\sigma}^{(3)} = e_1 + e_2 + \sigma e_3, \) and \( e_{2\sigma}^{(3)} = e_1 - e_2 + \sigma e_3. \) Introducing the bosonic ladder operator in momentum space \( b_q \) and its adjoint defined in the main text, the plasmonic Hamiltonian including dipole-dipole interactions up to the fourth nearest neighbors then reads

\[
H_{pl} = \hbar \sum_{q} \left\{ \left[ \omega_0 + 2\Omega \left( f_q + \sum_{n=2}^{4} f_q^{(n)} \right) \right] b_q^\dagger b_q + \Omega \left( f_q + \sum_{n=2}^{4} f_q^{(n)} \right) \left( b_q^\dagger b_{-q} + b_{-q} b_q \right) \right\},
\]
with

\[ f_q^{(2)} = \frac{1}{2\sqrt{2}} \sum_{j=1}^{3} \sum_{\sigma=\pm} C_{j\sigma}^{(2)} \cos (q \cdot e_{j\sigma}^{(2)}), \]  
(S5a)

\[ f_q^{(3)} = \frac{1}{3\sqrt{3}} \sum_{j=1}^{2} C_{j\sigma}^{(3)} \cos (q \cdot e_{j\sigma}^{(3)}), \]  
(S5b)

\[ f_q^{(4)} = \frac{1}{8} \sum_{j=1}^{3} C_j \cos (2q \cdot e_j). \]  
(S5c)

As explicited in the main text, the Hamiltonian (S4) above can be diagonalized by a Bogoliubov transformation, leading to the collective plasmon dispersion

\[ \omega_{pl}^{(4)} = \omega_0 \sqrt{1 + \frac{4}{\omega_0} \left( f_q + \sum_{n=2}^{4} f_q^{(n)} \right)}. \]  
(S6)

The dispersion relation (S6) is shown in Fig. S1 for various polarization angles of the localized surface plasmons (solid lines). For comparison, we also show in Fig. S1 the plasmon dispersion

\[ \omega_{pl}^{(4)} = \omega_0 \sqrt{1 + \frac{4}{\omega_0} f_q} \]  
(S7)

that only includes nearest neighbor interactions (dotted lines), as well as the dispersion

\[ \omega_{pl}^{(2)} = \omega_0 \sqrt{1 + \frac{4}{\omega_0} \left( f_q + f_q^{(2)} \right)} \]  
(S8)

and

\[ \omega_{pl}^{(3)} = \omega_0 \sqrt{1 + \frac{4}{\omega_0} \left( f_q + \sum_{n=2}^{3} f_q^{(n)} \right)} \]  
(S9)

that include the next nearest and third nearest neighbors (dashed and dashed-dotted lines in Fig. S1, respectively). As can be seen from the figure, dipole-dipole interactions beyond the nearest neighbor contribution do not lead to dramatic qualitative changes in the collective plasmon dispersion relation. For clarity, in the main text we thus limit ourselves to the discussion of dipole-dipole interaction effects between nearest neighbors only.
FIG. S1. Collective plasmon dispersion relation with dipole-dipole interaction including nearest (dotted), next nearest (dashed), third nearest (dashed-dotted) and fourth nearest neighbors (solid lines) along the high symmetry axes in the first Brillouin zone, for various polarization angles \((\theta, \varphi)\). In the figure, \(r = a/3\).