Plasmons in two-dimensional lattices of near-field coupled nanoparticles

François Fernique and Guillaume Weick
Université de Strasbourg, CNRS, Institut de Physique et Chimie des Matériaux de Strasbourg, UMR 7504, F-67000 Strasbourg, France

We consider plasmonic metasurfaces constituted by an arbitrary periodic arrangement of spherical metallic nanoparticles. Each nanoparticle supports three degenerate dipolar localized surface plasmon (LSP) resonances. In the regime where the interparticle distance is much smaller than the optical or near-infrared wavelength associated with the LSPs, the latter couple through the dipole–dipole interaction and form collective plasmonic modes which extend over the whole metasurface. Within a Hamiltonian model which we solve exactly, we derive general expressions which enable us to extract analytically the quasistatic plasmonic dispersion for collective modes polarized within the plane and perpendicular to the plane of the metasurface. Importantly, our approach allows us not only to consider arbitrary Bravais lattices, but also non-Bravais two-dimensional metacrystals featuring nontrivial topological properties, such as, e.g., the honeycomb, Lieb, or kagome lattices. Additionally, using an open quantum system approach, we consider perturbatively the coupling of the collective plasmons to both photonic and particle-hole environments, which lead, respectively, to radiative and nonradiative frequency shifts and damping rates, for which we provide analytical expressions. While it is tempting to make a direct analogy between the various systems which we consider and their electronic tight-binding equivalents, we critically examine how the long-range retarded and anisotropic nature of the dipole–dipole interaction may quantitatively and qualitatively modify the underlying band structures and discuss their experimental observability.

I. INTRODUCTION

The interaction of light with a small metallic particle results in collective electronic modes termed localized surface plasmons (LSPs) [1, 2]. In the case where the wavelength of the incoming light is much larger than the nanostructure itself, the LSP corresponds to a dipolar collective oscillation of the electronic cloud against the inert ionic background. While such a phenomena was empirically discovered centuries ago by late Romans [3], the underlying physics was only theorized by Mie at the beginning of the 20th century, who solved Maxwell’s equations for a metallic sphere embedded in a dielectric medium [4–6]. Since then, plasmonic nanostructures have attracted a surge of interest due to their ability to perform subwavelength optics by confining the electromagnetic field to nanometric regions [7–9]. Due to the extreme sensitivity of the LSP resonance frequency to the nanoparticle size, shape, material, and dielectric environment [2, 10], a wealth of appealing technological applications have risen from the field of nanoplasmonics, such as, e.g., chemical [11] and biological [12] sensors.

When two metallic nanoparticles are positioned in close vicinity of each other (i.e., separated by a distance typically smaller than the LSP wavelength) so that they form a dimer, another factor crucially influencing the resonance frequencies of the latter is the Coulomb interaction between the LSPs. The quasistatic dipole–dipole interaction, which decays as the interparticle distance \( d \) as \( 1/d^3 \), gives rise to coupled modes, often coined “hybridized” modes [13], which correspond to symmetric (in-phase) or antisymmetric (out-of-phase) configurations of the oscillating electric dipolar moments on each nanoparticle. For transverse-polarized modes (with respect to the axis formed by the dimer), the high- (low-energy) plasmonic state corresponds to an in-phase (out-of-phase) configuration. Conversely, for longitudinal modes, the low- (high-)energy state corresponds to aligned (anti-aligned) dipole moments. The splitting in frequencies between these hybridized modes scales with the inter-particle distance as \( 1/d^3 \), and can be spectroscopically resolved as long as the linewidth (which is of both radiative and nonradiative nature) of the two resonance peaks is somewhat smaller than the above-mentioned splitting.

The picture above is valid as long as the two nanoparticles are not too close to each other, so that higher multipolar modes do not mix with the dipolar ones [14–17], and the quantum tunneling of electronic charges between the two particles can be disregarded, such that so-called charge transfer plasmons are irrelevant [18–22]. Since the pioneering work by Ruppin [23], who extended Mie’s theory [4–6] to two nearby metallic spheres embedded in a dielectric medium, hybridized plasmonic modes in nanoparticle dimers have been investigated in numerous experimental [13, 24–29] and theoretical works [18, 30–38].

In periodic arrays of near-field coupled nanoparticles, the dipolar interaction between the LSPs leads to collective modes that are extended over the whole lattice. In chains of regularly-spaced nanoparticles, such collective plasmons were extensively studied both at the theoretical [17, 37, 39–54] and experimental [29, 40, 55–60] levels, since these systems may serve as plasmonic waveguides where plasmon–photon hybrid modes (so-called plasmon polaritons) are laterally confined to subwavelength scales and can possibly propagate over macroscopic distances. The importance of retardation effects in the dipolar in-
teraction, which become relevant for nanoparticles in the chain spaced by a distance of the order of the LSP wavelength, was put forward in Refs. [41–47, 51–54]. In particular, it was shown that retardation leads to a pronounced cusp-like feature in the dispersion relation of the collective plasmons polarized transversely to the chain for wavevectors corresponding to the intersection of the light cone with the quasistatic band structure. The crucial role played by radiative and absorption losses on the propagation of plasmonic waves along the nanoparticle chain was also studied in detail in the previous works of Refs. [17, 39–54]. Notably, Ref. [37] showed that the nonradiative Landau damping, that is, the desintegration of the collective plasmons into particle-hole pairs, is of primarily importance as it dominates the plasmon linewidth for nanoparticles of only a few nanometers in size.

Recently, dimerized [61–66] as well as zig zag chains of nanoparticles [67, 68] were proposed as a plasmonic analogue of the celebrated Su-Schrieffer-Heeger (SSH) model [69–71] presenting nontrivial topologically-protected edge states. In particular, the robustness of such topological states against the long range retarded dipolar interactions was discussed in Refs. [64–66].

The extension of the concepts introduced above to two spatial dimensions offers new exciting possibilities. Metasurfaces, that is, two-dimensional periodic arrangements of subwavelength metallic nanostructures, are indeed at present a very active field of research, as they enable one to tailor light in a way that goes far beyond what can be achieved with conventional optics. Thus far, the vast majority of the literature on plasmonic metasurfaces (see the review articles of Refs. [72, 73] and references therein) focused on the regime where the separation distance between each resonant element is of the order of the LSP wavelength, as this can be experimentally achieved with nowadays nanofabrication techniques. In this regime, the diffractive electromagnetic far fields generated by the essentially noninteracting nanoparticles of the array interfer and give rise to so-called surface lattice resonances (SLRs). The latter are of particular interest since they lead to much narrower absorption lines as that of the individual constituents of the metasurface, as well as angle-dependent dispersions, as was theoretically predicted in Refs. [74, 75] and later experimentally verified in Refs. [76–78]. Further works have demonstrated the use of SLRs in tailoring frequency stop gaps [79] and are of relevance to applications in light emission [80, 81].

Genuinely quantum-mechanical effects [82], such as the exciting perspective of lasing [83–85], as well as Bose–Einstein condensation [86, 87], have also been demonstrated in two-dimensional plasmonic lattices. Notably, recent works [88, 89] combining modelling and experiments have brought attention to the role of the geometrical arrangement of the nanostructures composing the metasurface on the SLR properties.

In the present work, we focus on the less explored case of near-field coupled nanoparticles supporting dipolar LSPs in metasurfaces. In this case, the stronger dipolar coupling between LSPs, as compared to the weak diffractive couplings encountered in SLRs, can exhibit potentially interesting analogies with atomically-thin, two-dimensional materials, such as graphene [90] or transition metal dichalcogenides [91], where the electronic band structures are usually well-described by electronic tight-binding calculations. Metasurfaces composed of near-field nanoparticles may indeed present appealing nontrivial features in their bandstructure, paving the way to topological photonics performed with subwavelength elements [92–94]. For instance, it was theoretically demonstrated that a honeycomb lattice of plasmonic nanoparticles that are near-field coupled present chiral massless Dirac-like bosonic collective excitations [95–97] which behave as electrons in graphene [90]. Exotic, so-called type-II Dirac plasmon-polaritons presenting a fully-tunable tilted conical dispersion, were also recently unveiled in Ref. [98].

Due to the vast number of possible two-dimensional lattices of near-field coupled plasmonic nanoparticles with potentially interesting properties in their band structure, here we develop a general theoretical model which enables us to consider the plasmonic properties of arbitrary metasurfaces. Our open quantum system approach, which builds on previous works on plasmonic dimers [36, 38] and chains [37, 53], allows us to unveil analytical expressions for the quasistatic plasmonic dispersion relations for collective modes polarized parallel and perpendicular to the plane of the metasurface. By considering the coupling of the purely plasmonic modes to photons of the electromagnetic vacuum, we also consider the effects of retardation in the light–matter interaction, and we show that such retardation effects play a crucial role on the plasmonic band structure. Our approach further gives access to the radiative lifetime of the plasmonic modes, which we evaluate analytically. Importantly, we also consider the decay of the collective plasmons into electron-hole pairs and show that the resulting Landau damping can be as significant as it is in single nanoparticles which are only a few nanometers in size.

This paper is organized as follows: Section II presents our Hamiltonian model and the open quantum system approach which we use to study collective plasmons in a generic two-dimensional array of interacting spherical metallic nanoparticles. We provide in Sec. III the diagonalization procedure of the purely plasmonic Hamiltonian, which gives access to the quasistatic dispersion relation of the collective modes. The latter is extensively discussed in Sec. IV, including the cases of Bravais and non-Bravais lattices. In Sec. V, we consider the effects of the photonic environment alone and present our results for the fully-retarded dispersion relation of the plasmonic modes, as well as the corresponding radiative lifetimes. Section VI then analyzes the role played by the electronic environment onto the collective modes and presents our results for their Landau damping decay rates, as well as their associated electronic frequency shifts. In Sec. VII, we discuss the experimental observability of the plas-
monic modes, before we conclude in Sec. VIII. A few technical details, as well as supplementary examples of application of our theoretical model, are presented in four appendixes.

II. MODEL

We consider an ensemble of interacting spherical metallic nanoparticles of radius \( a \) forming an arbitrary two-dimensional Bravais lattice with a basis. The array is characterized by the vectors \( \mathbf{R} = nt_1 + mt_2 \) forming the Bravais lattice. Here, \( t_1 \) and \( t_2 \) are the primitive lattice vectors, while \( n \in [0, N_1] \) and \( m \in [0, N_2] \) are integer numbers with \( N_1 \) \((N_2)\) the number of unit cells in the \( t_1 \) \((t_2)\) direction. The array is composed of \( S \) sublattices, and the nanoparticles belonging to the sublattice \( s = 1, \ldots, S \) are located at \( \mathbf{R}_s = \mathbf{R} + \mathbf{d}_s \), where \( \mathbf{d}_s \) is the vector belonging to the \( xy \) plane and connecting the sublattice \( s \) to \( \mathbf{R} \) (see Fig. 1). By convention, we set \( \mathbf{d}_1 = 0 \) in the remainder of the paper.

Each nanoparticle forming the array supports three degenerate orthogonal dipolar LSP resonances polarized along the \( \sigma = x, y, \) and \( z \) directions and characterized by the frequency \( \omega_0 \). Neglecting the effect of the embedding medium, as well as the screening of the valence electrons by the core electrons, one has \( \omega_0 = \omega_p/\sqrt{3} \), where \( \omega_p = \sqrt{4\pi n_e e^2/m_e} \) is the plasma frequency [2].\(^1\)

Here, \( -e \left( < 0 \right) \) is the electron charge, \( m_e \) its mass, and \( n_e \) the electronic density of the metal.

The dipolar LSPs supported by the nanoparticles in the array interact with their neighbors through the Coulomb interaction. In this work we focus on the subwavelength regime where \( 3a \lesssim d \ll \kappa_0^{-1} \), with \( d \) the center-to-center nearest neighbor interparticle distance and \( \kappa_0 = \omega_0/c \), with \( c \) the speed of light in vacuum.

In this parameter regime [17], the dominant interaction between two nanoparticles is the near-field quasistatic dipole–dipole interaction which decays as \( 1/d^3 \). Such a coupling gives rise to collective plasmonic modes which extend over the whole metasurface [72].

Similarly to individual LSPs, the extended plasmonic modes supported by the metasurface are coupled to both a photonic and an electronic environment. The collective plasmons are indeed coupled to vacuum electromagnetic modes through the light–matter interaction, giving rise to finite radiative lifetimes as well as radiative frequency shifts, stemming from the retarded part of the dipole–dipole interaction [38, 53]. Moreover, such collective modes are coupled to electron-hole excitations inside the nanoparticles, leading to the nonradiative Landau damping and an additional frequency renormalization.

We write the full Hamiltonian of the system as

\[
H = H_{\text{pl}} + H_{\text{ph}} + H_{\text{eh}} + H_{\text{pl-ph}} + H_{\text{pl-eh}},
\]

where \( H_{\text{pl}} \) describes the purely plasmonic degrees of freedom, while \( H_{\text{ph}} \) and \( H_{\text{eh}} \) correspond to the photonic and electronic environments, respectively. In Eq. (1), \( H_{\text{pl-ph}} \) and \( H_{\text{pl-eh}} \) are the coupling Hamiltonians of the plasmonic subsystem to photons and electron-hole pairs, respectively.

Within the Coulomb gauge [99, 100], the plasmonic Hamiltonian

\[
H_{\text{pl}} = H_{\text{pl}}^0 + H_{\text{pl}}^{\text{int}}
\]

describing the near-field coupled LSPs is characterized by a noninteracting and an interacting term [37, 96]. The noninteracting part related to individual nanoparticles reads

\[
H_{\text{pl}}^0 = \sum_s \sum_{\mathbf{R}_s} \sum_\sigma \left\{ \frac{\|\Pi^\sigma_s(\mathbf{R}_s)\|^2}{2N_e m_e} + \frac{N_e m_e}{2} \omega_0^2 \|h^\sigma_s(\mathbf{R}_s)\|^2 \right\},
\]

with \( h^\sigma_s(\mathbf{R}_s) \) the \( \sigma \)-component of the displacement field associated with the dipole moment \( \mathbf{p}(\mathbf{R}_s) = -eN_e \sum_\sigma h^\sigma_s(\mathbf{R}_s) \hat{\sigma} \) of a single LSP located at position \( \mathbf{R}_s \), while \( \Pi^\sigma_s(\mathbf{R}_s) \) is the momentum conjugated to \( h^\sigma_s(\mathbf{R}_s) \). In Eq. (3), \( N_e \) is the total number of valence electrons in each nanoparticle. The interacting term in Eq. (2) arises from the quasistatic dipole–dipole interaction and reads

\[
H_{\text{pl}}^{\text{int}} = \frac{(eN_e)^2}{2} \sum_{ss'} \sum_{\mathbf{R}_s \mathbf{R}_s'} \sum_{\sigma \sigma'} \delta_{\sigma \sigma'} \delta_{\sigma' \sigma} \left[ \frac{\kappa_0}{|\mathbf{R}_s - \mathbf{R}_s'|^3} \right] \chi_{ss'}(\mathbf{R}_s, \mathbf{R}_s'),
\]

where \( \rho_{ss'} = \mathbf{R}_s' - \mathbf{R}_s \). Here and in what follows, hats designate unit vectors.

As we deal with nanoparticles of small sizes, quantum-size effects (such as Landau damping) can be important.

\(^1\) Throughout this paper we use cgs units.
Moreover, a quantum treatment of the plasmonic degrees of freedom provides a self-contained description of the light–matter interaction [38, 53]. In view of the analysis of these effects we thus present the quantized plasmonic Hamiltonian (2) in terms of the bosonic ladder operators that annihilate an LSP at position $\mathbf{R}_s$ on sublattice $s$ with polarization $\sigma = x, y, z$,

$$b^\sigma_s(\mathbf{R}_s) = \sqrt{\frac{N_e m_e \hbar \omega}{2 \hbar}} h^\sigma_s(\mathbf{R}_s) + \frac{i \Pi^\sigma_s(\mathbf{R}_s)}{\sqrt{2N_e m_e \hbar \omega}}$$

and its adjoint $b^{\sigma \dagger}_s(\mathbf{R}_s)$. Together with Eqs. (3) and (4), Eq. (2) thus takes the form

$$H_{pl} = \frac{\hbar \omega}{2} \sum_s \sum\limits_{\mathbf{R}_s} \sum\limits_{\sigma} \sum\limits_{\mathbf{R}'_s} \sum\limits_{\sigma' \sigma''} [b^\sigma_s(\mathbf{R}_s) b^{\sigma' \dagger}_{\sigma''} S(\mathbf{R}_s) + b^{\sigma' \dagger}_{\sigma''} S(\mathbf{R}_s)]$$

$$\times \left[ b^{\sigma''} (\mathbf{R}'_s) + b^{\sigma'' \dagger}_{\sigma''} (\mathbf{R}'_s) \right] \times \delta_{\sigma \sigma'} - 3(\hat{\sigma} \cdot \hat{\rho}_{s\sigma''}) (\hat{\sigma}' \cdot \hat{\rho}_{s\sigma''})$$

$$\times \frac{\hbar \omega}{2} \sum_s \sum\limits_{\mathbf{R}_s} \sum\limits_{\sigma} (\mathbf{R}_s - \mathbf{R}'_s / \langle d \rangle^3),$$

with the coupling constant

$$\Omega = \frac{\omega_0}{2} \left( \frac{a}{d} \right)^3.$$  

Note that $\Omega \ll \omega_0$ since we consider interparticle distances $d \geq 3a$.

The Hamiltonian (6) displays some similarities with a tight-binding Hamiltonian of an electronic two-dimensional system [101]. The first term on the right-hand side of Eq. (6) $\propto b^\sigma_s(\mathbf{R}_s) b^{\sigma'}_{s' \sigma''} (\mathbf{R}'_s)$ corresponds to a fixed onsite energy, while the resonant terms $\propto b^{\sigma'}_{s' \sigma''} (\mathbf{R}'_s) b^{\sigma''} (\mathbf{R}_s)$ in the second term describe the creation of an LSP at the lattice site $\mathbf{R}_s$ together with the destruction of an LSP at the lattice site $\mathbf{R}'_s$, similar to a hopping term.

There are however important differences between the plasmonic Hamiltonian (6) and an electronic tight-binding Hamiltonian. Firstly, plasmons correspond to bosonic excitations, which do not have a finite chemical potential. Secondly, the dipole–dipole interaction responsible for the existence of the collective plasmons is quite different compared to the hopping amplitude in a tight-binding model [101]. On the one hand, the long-range dipolar interaction scales with $1/|\mathbf{R}_s - \mathbf{R}'_s|^{3}$, whereas the hopping amplitude decreases exponentially with the distance. Thus, the dipolar interaction beyond the first neighbors can have important effects. On the other hand, the dipole–dipole interaction depends on the polarization of the excitations, contrarily to those in tight-binding models. Thirdly, there are additional non-resonant terms $\propto b^{\sigma} (\mathbf{R}_s) b^{\sigma}_{s' \sigma''} (\mathbf{R}'_s) + \text{h.c.}$ in Eq. (6) which do not conserve the number of quasiparticles and play a crucial role for physical quantities depending on the plasmonic eigenstates, e.g., the collective mode damping rates [37]. How the above-mentioned differences may crucially affect the plasmonic band structure is extensively discussed in Sec. IV.

The Hamiltonian (1) further describes the coupling of the collective plasmons to vacuum electromagnetic modes in a volume $\mathcal{V}$ described by the Hamiltonian

$$H_{pl} = \sum_{\mathbf{R}_s} \sum\limits_{\mathbf{R}_s} \sum\limits_{\lambda_k} \frac{\hbar \omega}{2} a^\dagger_{\lambda_k} a_{\lambda_k} \mathbf{A}(\mathbf{R}_s),$$

where $a_{\lambda_k}$ ($a_{\lambda_k}^\dagger$) annihilates (creates) a photon with wavevector $\mathbf{k}$, transverse polarization $\lambda_k$ (i.e., $\mathbf{k} \cdot \hat{\lambda}_k = 0$), and conical dispersion $\omega_k = c|\mathbf{k}|$. In the long-wavelength limit $|\mathbf{k}|a \ll 1$, the minimal-coupling Hamiltonian between plasmons and photons in Eq. (1) reads [99, 100]

$$H_{pl-ph} = \sum_s \sum\limits_{\mathbf{R}_s} \left[ \frac{\epsilon_{\mathbf{k}} - \Pi_s(\mathbf{R}_s) \cdot \mathbf{A}(\mathbf{R}_s)}{\mathcal{V} \omega_k} + \frac{N_e e^2}{2 m_e c^2} \mathbf{A}(\mathbf{R}_s)^2 \right],$$

with

$$\Pi_s(\mathbf{R}_s) = i \sqrt{\frac{N_e m_e \hbar \omega}{2}} \sum\limits_{\sigma} \hat{\sigma} \left[ b^\sigma(\mathbf{R}_s) - b^{\sigma \dagger}_{s' \sigma''} (\mathbf{R}_s) \right]$$

and where

$$\mathbf{A}(\mathbf{R}_s) = \sum_{\mathbf{R}_s} \frac{\hat{\lambda}_k}{\mathcal{V} \omega_k} \left( a_{\lambda_k}^\dagger e^{i \mathbf{k} \cdot \mathbf{R}_s} + a_{\lambda_k} e^{-i \mathbf{k} \cdot \mathbf{R}_s} \right),$$

is the vector potential evaluated at the nanoparticle centers.$^2$ Importantly, within the Coulomb gauge, the first term in the right-hand side of Eq. (9) contains the retardation effects stemming from the finite velocity of light.

In addition to the photonic environment, the collective plasmons are coupled to electron-hole excitations described by the Hamiltonian [102, 103]

$$H_{eh} = \sum_s \sum\limits_{\mathbf{R}_s} \sum\limits_{\mathbf{R}_s} c_{\mathbf{R}_s}^\dagger c_{\mathbf{R}_s} c_{\mathbf{R}_s, i},$$

where $c_{\mathbf{R}_s}^\dagger$ ($c_{\mathbf{R}_s}$) annihilates (creates) an electron in the nanoparticle located at $\mathbf{R}_s$ associated with the one-body state $\mathbf{R}_s, i$ with energy $\epsilon_{\mathbf{R}_s, i}$ in the self-consistent potential $V$ of that nanoparticle. Assuming $V$ to be a spherical

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$^2$ Note that, since we consider interparticle separation distances much smaller than the wavelength associated with the LSP resonances, we neglect Umklapp processes in equations (8) and (9).
hard-wall potential with infinite height, the coupling between plasmonic and single-electronic degrees of freedom is [36]

\[ H_{\text{pl-eh}} = \Lambda \sum_s \sum_{R_s} \sum_{\sigma} \sum_i \left( b_s^\sigma(R_s) + b_s^\sigma\dagger(R_s) \right) \times \langle R_s | i | R_{s,j} \rangle c_{R_s,i}^\dagger c_{R_{s,j}}, \]

with

\[ \Lambda = \sqrt{\frac{\hbar m_{\text{el,0}}}{2N_e}}. \]

III. DIAGONALIZATION OF THE PLASMONIC HAMILTONIAN

In this section, we start by focusing on the plasmonic degrees of freedom alone. We present the diagonalization procedure of the Hamiltonian (6), from which we obtain the quasistatic plasmonic band structure which we analyze in detail in the subsequent Sec. IV. The discussion about how the band structure is influenced by retardation effects in the dipole-dipole interaction is relegated to Sec. V.

Since we consider large metasurfaces, we use periodic boundary conditions and move to the wavevector space using the Fourier transform

\[ b_s^\sigma(q) = \frac{1}{\sqrt{N}} \sum_{R_s} e^{-i R_s \cdot q} b_s^\sigma(R_s) \]

of the bosonic ladder operator \( b_s^\sigma(R_s) \) defined in Eq. (5), with \( N = N_1N_2 \gg 1 \) the total number of unit cells in the lattice. The Hamiltonian (6) then reads

\[ H_{\text{pl}} = \hbar \omega_q \sum_q \sum_s \sum_{\sigma} b_{s+q}^\sigma(q) b_s^\sigma(q) \]

\[ + \frac{\hbar \Omega}{2} \sum_q \sum_{ss'} \sum_{\sigma\sigma'} \left\{ f_{s,s'}^{\sigma\sigma'}(q) b_{s'}^\sigma(q) b_s^\sigma\dagger(-q) \right\} \]

\[ \times \left\{ b_{s+q}^\sigma(q) + b_s^\sigma\dagger(-q) \right\} + \text{h.c.}, \]

In Eq. (16), the lattice sum

\[ f_{s,s'}^{\sigma\sigma'}(q) = \sum_{\rho_{s,s'} \neq 0} \left( \frac{d}{\rho_{s,s'}} \right)^{3} e^{i q \cdot \rho_{s,s'}} \times \left[ \delta_{\sigma\sigma'} - 3(\hat{\sigma} \cdot \hat{\rho}_{s,s'}) (\hat{\sigma}' \cdot \hat{\rho}_{s,s'}) \right] \]

takes into account the quasistatic dipolar interaction between each pair of nanoparticles composing the metasurface. Note that for \( s = s' \), \( f_{s,s'}^{\sigma\sigma'}(q) \) is real due to the inversion symmetry of the Bravais lattice. Moreover, \( f_{s,s'}^{\sigma\sigma'}(q) = [f_{s,s'}^{\sigma\sigma'}(q)]^* \) since \( \rho_{s,s'} = -\rho_{s',s} \). In the remainder of the paper, the lattice sum (17) is calculated for a specific metasurface numerically, until satisfactory convergence is obtained.

The plasmonic Hamiltonian \( H_{\text{pl}} \) given in Eq. (16) is quadratic, and can thus be diagonalized exactly by means of a bosonic Bogoliubov transformation. We then introduce a set of bosonic operators

\[ \beta_{s}^\sigma(q) = \sum_{s,\sigma} \left[ u_{s,s'}^{\sigma\sigma'}(q) b_s^\sigma(q) + v_{s,s'}^{\sigma\sigma'}(q) b_s^\sigma\dagger(-q) \right] \]

annihilating a collective plasmon with wavevector \( q \) in the band \( \tau \) with polarization \( \varepsilon \). Notice that, in general, the latter is a \( q \)-and \( \tau \)-dependent quantity but we drop in the remaining of this paper both indexes for notational simplicity. The inverse transformation is given by

\[ b_s^\sigma(q) = \sum_{s,\sigma} \left[ u_{s,s'}^{\sigma\sigma'}(q) \beta_{s'}^\sigma(q) - v_{s,s'}^{\sigma\sigma'}(q) \beta_{s'}^\sigma\dagger(-q) \right]. \]

In Eqs. (18) and (19), \( u_{s,s'}^{\sigma\sigma'}(q) \) and \( v_{s,s'}^{\sigma\sigma'}(q) \) are complex coefficients which are determined by imposing that \( H_{\text{pl}} \) is diagonal in this new basis, i.e.,

\[ H_{\text{pl}} = \sum_q \sum_{\tau,\varepsilon} H_{\tau,\varepsilon}^q(q), \quad H_{\tau,\varepsilon}^q(q) = \hbar \omega^q_{\tau,\varepsilon}(q) \beta^\tau_{\varepsilon}(q) \beta^\tau_{\varepsilon}\dagger(q), \]

where \( \omega^q_{\tau,\varepsilon}(q) \) is the collective plasmon dispersion relation. The Bogoliubov operators \( \beta^\tau_{\varepsilon}(q) \) and \( \beta^\tau_{\varepsilon}\dagger(q) \) act on an eigenstate \( |n^\tau_{\varepsilon}(q)\rangle \) of the Hamiltonian \( H_{\tau,\varepsilon}^q(q) \) corresponding to a collective plasmon in the band \( \tau \) with wavevector \( q \) and polarization \( \varepsilon \) as \( \beta^\tau_{\varepsilon}(q)|n^\tau_{\varepsilon}(q)\rangle = \sqrt{n^\tau_{\varepsilon}(q)}|n^\tau_{\varepsilon}(q) - 1\rangle \) and \( \beta^\tau_{\varepsilon}\dagger(q)|n^\tau_{\varepsilon}(q)\rangle = \sqrt{n^\tau_{\varepsilon}(q) + 1}|n^\tau_{\varepsilon}(q) + 1\rangle \), respectively. Here, \( n^\tau_{\varepsilon}(q) \) is a non-negative integer. Note that the bosonic commutation relations \( [\beta^\tau_{\varepsilon}(q), \beta^\tau_{\varepsilon}\dagger(q')] = \delta_{\tau,\tau'} \delta_{\varepsilon,\varepsilon'} \delta_{qq'} \) and \( [\beta^\tau_{\varepsilon}(q), \beta^\tau_{\varepsilon}'(q')] = 0 \) impose that the Bogoliubov coefficients in Eq. (18) fulfill the relations

\[ \sum_{s,\sigma} \left[ u_{s,s'}^{\sigma\sigma'}(q) u_{s,s'}^{\sigma\sigma'}(q) - v_{s,s'}^{\sigma\sigma'}(q) v_{s,s'}^{\sigma\sigma'}(q) \right] = \delta_{\sigma,\sigma'} \delta_{\varepsilon,\varepsilon'} \]

\[ \sum_{s,\sigma} \left[ u_{s,s'}^{\sigma\sigma'}(q) v_{s,s'}^{\sigma\sigma'}(q) - v_{s,s'}^{\sigma\sigma'}(q) u_{s,s'}^{\sigma\sigma'}(q) \right] = 0, \]

where \( u_{s,s'}^{c\sigma\rho}(q) = u_{s,s'}^{c\sigma\rho}(q) \) and \( v_{s,s'}^{c\sigma\rho}(q) = v_{s,s'}^{c\sigma\rho}(q) \).

The dispersion relation \( \omega^q_{\tau,\varepsilon}(q) \), as well as the coefficients of the Bogoliubov transformation (18), are obtained from the Heisenberg equation of motion

\[ [\beta^\tau_{\varepsilon}(q), H_{\text{pl}}] = \hbar \omega^q_{\tau,\varepsilon}(q) \beta^\tau_{\varepsilon}(q), \]

which yields the system of equations

\[ \left[ \omega_0 - \omega^q_{\tau,\varepsilon}(q) \right] u_{s,s'}^{c\sigma\rho}(q) \]

\[ + \Omega \sum_{s',\sigma'} \left[ u_{s,s'}^{c\sigma\rho}(q) - v_{s,s'}^{c\sigma\rho}(q) \right] f_{s,s'}^{c\sigma\rho}(q) = 0 \]

3 In practice, we perform the summation in Eq. (17) up to \( \rho_{s,s'}^{\text{max}} = 300d \), which yields a relative error of the order of \( 10^{-9} \).
The two expressions above need to be satisfied for all integer $s \in [1, S]$ and for all polarizations $\sigma = x, y, z$, yielding a $6S \times 6S$ eigensystem. Due to the structure of the lattice sum $f_{s\sigma}^{\sigma'}(q)$ defined in Eq. (17), such an eigensystem decouples into a block-diagonal matrix composed of a $4S \times 4S$ and a $2S \times 2S$ block, corresponding to the in-plane (IP, $\sigma = x, y$) and out-of-plane (OP, $\sigma = z$) polarized modes, respectively. Each block then yields a secular equation of order $2S$ and $S$ in $|\omega_\sigma(q)|^2$, respectively, which then gives access to the band structure.

Alternatively to the exact diagonalization procedure presented above, it may be useful to treat the nonresonant terms in Eq. (16) perturbatively, since, for all practical purposes, the coupling constant $\Omega \ll \omega_0$ [cf. Eq. (7)]. Such a procedure has the advantage of dividing by two the dimension of the system of equations leading to the plasmonic band structure, which may be helpful in deriving the spectrum and the associated Bogoliubov coefficients analytically. This perturbative procedure is presented in Appendix A.

Additional insight about the nature of the quasistatic collective plasmons can be obtained from their corresponding eigenstates, from which we can deduce the polarization $\varepsilon$ of the collective modes. Introducing the vector $\mathbf{u}_\varepsilon(q) = (\sum s u_{s\varepsilon}^x(q), \sum_s u_{s\varepsilon}^y(q), \sum_s u_{s\varepsilon}^z(q))$, we define the polarization angle as [104]

$$\phi^\varepsilon(q) = \arccos (|\mathbf{u}_\varepsilon(q) \cdot \hat{q}|). \quad (25)$$

With such a definition, purely longitudinal (transverse) collective plasmons correspond to $\phi^\varepsilon(q) = 0 (\pi/2)$.

IV. QUASISTATIC PLASMONIC BAND STRUCTURE

We now focus on the quasistatic band structure of the collective plasmons arising from the near-field dipolar interaction between LSPs and obtained from the diagonalization procedure presented in Sec. III. We first concentrate in Sec. IV A on metasurfaces composed of nanoparticles arranged in an arbitrary Bravais lattice. We will then consider the more complex problem of lattices with a basis in Sec. IV B.

A. Bravais lattices

1. Arbitrary Bravais lattices

In the case of an arbitrary Bravais lattice (i.e., $S = 1$), the sublattice indexes $s$ and $s'$, as well as the band index $\tau$ are irrelevant and can then be dropped from the system of equations (24). Such a system can be fully analytically solved, yielding for the OP plasmonic modes polarized in the $\varepsilon = z$ direction the dispersion relation

$$\omega^z(q) = \omega_0 \sqrt{1 + \frac{2}{\omega_0} f^{zz}(q)}, \quad (26)$$

and

$$\omega^{\parallel, \pm}(q) = \omega_0 \left[1 + \frac{1}{\omega_0} \left| f^{xx}(q) + f^{yy}(q) \right| \pm \sqrt{\left| f^{xx}(q) - f^{yy}(q) \right|^2 + 4 \omega_0^2} \right]^{1/2}, \quad (27)$$

for the IP modes with polarizations $\varepsilon = \varepsilon_{\parallel, \pm}$. We find for the corresponding coefficients of the Bogoliubov transformation (18)

$$u^{zz}(q) = \frac{\omega^z(q) + \omega_0}{2\sqrt{\omega_0\omega^z(q)}}$$

and

$$v^{zz}(q) = \frac{\omega^z(q) - \omega_0}{2\sqrt{\omega_0\omega^z(q)}}$$

in the case of the OP polarized modes ($\sigma = z$). For the IP modes ($\sigma = x, y$), the condition $\zeta_{\pm}^\varepsilon(q)\zeta_{\pm}^\varepsilon(q) = \left|2\omega_0\Omega f^{xy}(q)\right|^2$ must be fulfilled, with $\zeta_{\pm}^\varepsilon(q) = |\omega^{\parallel, \pm}(q)|^2 - \omega_0^2 - 2\omega_0\Omega f^{\sigma\sigma}(q) (\sigma = x, y)$, so that we obtain

$$u^{\parallel, \pm x}(q) = \frac{\omega^{\parallel, \pm x}(q) + \omega_0}{2\sqrt{\omega_0\omega^{\parallel, \pm x}(q)}} \sqrt{\frac{\zeta_{\pm}^y(q)}{\sum_{\sigma=x,y} \zeta_{\pm}^\varepsilon(q)}}, \quad (30a)$$

$$u^{\parallel, \pm y}(q) = \pm \text{sgn} \left\{f^{xy}(q)\right\} \frac{\omega^{\parallel, \pm x}(q) + \omega_0}{2\sqrt{\omega_0\omega^{\parallel, \pm x}(q)}} \sqrt{\frac{\zeta_{\pm}^y(q)}{\sum_{\sigma=x,y} \zeta_{\pm}^\varepsilon(q)}}, \quad (30b)$$

and

$$v^{\parallel, \pm x}(q) = \frac{\omega^{\parallel, \pm x}(q) - \omega_0}{2\sqrt{\omega_0\omega^{\parallel, \pm x}(q)}} \sqrt{\frac{\zeta_{\pm}^y(q)}{\sum_{\sigma=x,y} \zeta_{\pm}^\varepsilon(q)}}, \quad (31a)$$

$$v^{\parallel, \pm y}(q) = \pm \text{sgn} \left\{f^{xy}(q)\right\} \frac{\omega^{\parallel, \pm x}(q) - \omega_0}{2\sqrt{\omega_0\omega^{\parallel, \pm x}(q)}} \sqrt{\frac{\zeta_{\pm}^y(q)}{\sum_{\sigma=x,y} \zeta_{\pm}^\varepsilon(q)}}, \quad (31b)$$

We point out that neglecting the nonresonant terms in Eq. (16), which corresponds to performing the rotating wave approximation (RWA), yields the same dispersion relations (26) and (27) to first order in $\Omega \ll \omega_0$ and the same Bogoliubov coefficients (28) and (30), while $v^{\sigma}(q) = 0$ within such an approximation (cf. Appendix A).
2. Square lattice

As an example of application of our general method for obtaining the quasistatic band structure of plasmonic modes in generic Bravais lattices, we consider in the following the simple square lattice sketched in Fig. 2(a), whose corresponding first Brillouin zone (1BZ) is depicted in Fig. 2(b). (In Appendix B, we briefly discuss the band structure of two other typical Bravais lattices, such as the rectangular and hexagonal ones.)

The plasmonic dispersion relation (26) for the square lattice is plotted in Fig. 2(c) for the OP polarization as a solid line. For comparison, we also show (dashed line) the plasmonic band structure considering only dipolar interactions between nearest neighbors (nn) in the lattice, for which the lattice sum (17) reduces to \( f_{nn}^{zz}(q) = 2[\cos(q_x d) + \cos(q_y d)] \). As can be seen from Fig. 2(c), the nearest-neighbor approximation qualitatively reproduces the full band structure in most of the 1BZ, except for wavenumbers close to the \( \Gamma \) point. There, the long-range nature of the quasistatic dipolar interaction leads to a pronounced cusp of the dispersion relation.

The behavior of the quasistatic plasmonic dispersion relation for OP polarization close to the \( \Gamma \) point mentioned above can be understood by treating the nearest neighbors in the lattice sum (17) exactly, while averaging the interactions beyond nearest neighbors in the spirit of the mean-field (mf) approximation, leading to

\[
\omega^2(q) \simeq \omega_0 + \Omega \left[ 4 + \sqrt{2} \pi - 2 \pi |q| d + \left( \frac{\pi}{\sqrt{2}} - 1 \right) (|q| d)^2 \right],
\]

so that a cusp appears in the dispersion relation (26). In Fig. 2(c) we show by a dotted line the plasmonic band structure within the mean-field approximation detailed above. As can be seen from the figure, the mean-field approximation accurately describes the cusp of the full band structure in the vicinity of the \( \Gamma \) point, while it tends toward the nearest-neighbor approximation away from the \( \Gamma \) point.

We now turn to the discussion of the plasmonic modes polarized within the plane formed by the square lattice.

![FIG. 2.](image-url) (a) Sketch of a square lattice with primitive lattice vectors \( t_1 = d (1, 0) \) and \( t_2 = d (0, 1) \). (b) Corresponding first Brillouin zone, with primitive reciprocal vectors \( b_1 = \frac{2\pi}{t} (1, 0) \) and \( b_2 = \frac{2\pi}{t} (0, 1) \). (c),(d) Quasistatic plasmonic dispersion relation as a function of the wavevector \( q \) (scaled with the interparticle distance \( d \)) along high-symmetry paths in the first Brillouin zone [cf. panel (b)] for (c) out-of-plane (OP) and (d) in-plane (IP) polarizations. The solid lines represent the full quasistatic dispersion, including long-range couplings, and the color code corresponds to the polarization angle (25), which equals 0 (\( \pi/2 \)) for purely longitudinal (transverse) modes. The dashed and dotted lines correspond, respectively, to the nearest-neighbor and mean-field approximations discussed in the text. In the figure, the interparticle distance \( d = 3a \) (corresponding to \( \Omega = \omega_0/54 \)).

The band structure (27) is plotted in Fig. 2(d) as solid lines. The color code corresponds to the polarization.
angle defined in Eq. (25). While for the high-symmetry axes $\Gamma M$ or $\Gamma X$, the IP collective plasmons are purely longitudinal or transverse, for less-symmetric axes such as the $MX$ direction in the 1BZ, such modes can be of a mixed type. For comparison, we further plot the dispersion relation taking into account nearest-neighbor couplings only, for which the lattice sums in Eq. (27) are replaced by $f_{nn}^{\sigma}(q) = -4 \cos(q_x d) + 2 \cos(q_y d)$, $f_{nn}^{xy}(q) = 2 \cos(q_x d) - 4 \cos(q_y d)$, and $f_{nn}^{xx}(q) = 0$. In contrast to the OP modes [Fig. 2(c)], the long-range nature of the dipolar interaction has a more pronounced effect on the plasmonic band structure for IP polarized modes. For instance, the dipolar interaction lifts the degeneracy of such a singularity in the derivative of $\omega(x)$. The corresponding Bogoliubov coefficients are given by

$$u_{\tau\tau}^{zz}(q) = \frac{\omega(x) + \omega_0}{2\sqrt{2}\omega_0\omega_0^2(q)},$$

and

$$v_{\tau\tau}^{zz}(q) = \frac{\omega(x) - \omega_0}{2\sqrt{2}\omega_0\omega_0^2(q)}.$$  

For IP polarization ($\sigma = z$), the $8 \times 8$ eigenvalue problem can in principle be solved analytically, but provides cumbersome expressions. For practical purposes, we therefore solve for the eigenproblem numerically. To illustrate our method in the special case of bipartite lattices of near-field coupled metallic nanoparticles, we consider the celebrated honeycomb lattice sketched in Fig. 3(a). The corresponding 1BZ is shown in Fig. 3(b). Such a metasurface has been predicted [96, 97] to exhibit Dirac-like collective plasmonic modes at the $\Gamma$ and $K$ points of the 1BZ, with appealing topological properties such as a nontrivial Berry phase (and its related topologically-protected edge states [105]) or the absence of backscattering. Importantly, the results put forward in Refs. [96, 97] rely on short-range dipolar couplings between nearest neighbors alone. Moreover, Refs. [96, 97] considered the case of orientated dipoles, relevant, e.g., for elongated metallic rods, while we consider here the case of spherical nanoparticles.

The two plasmonic bands (37) for $\sigma = z$ are plotted in Fig. 3(c) as solid lines. For comparison, we also show by dashed lines the plasmonic band structure with dipolar interactions between nearest neighbors only [96], given by Eq. (37) and replacing $f_{111}^{zz}(q)$ and $f_{122}^{zz}(q)$ by $f_{nn,11}^{zz}(q) = 0$ and $f_{nn,12}^{zz}(q) = \sum_{m=1}^{3} e_{m}^{zz} \mathbf{e}_{m}$, respectively. Here, the vectors $\mathbf{e}_{1} = d_{2} - t_{2}$, $\mathbf{e}_{2} = d_{2}$, and $\mathbf{e}_{3} = d_{3} - t_{1}$ connect a lattice site belonging to the red sublattice in Fig. 3(a) to its three (blue) nearest neighbors. As can be seen from Fig. 3(c), the presence of a Dirac point at the $\Gamma$ point of the 1BZ located at $\mathbf{K} = \frac{4\pi}{3\sqrt{3}}(1,0)$ is not ruled out by long-range interactions. Indeed, in the vicinity of the $\Gamma$ point, expanding the lattice sums to linear order in $|k|$, where $q = \mathbf{K} + k$ with $|k| \ll |\mathbf{K}|$, yields $f_{111}^{zz}(q) \simeq f_{111}^{zz}(\mathbf{K}) \simeq -0.449$ and $f_{122}^{zz}(q) \simeq -1.16(k_x + k_y)$.

B. Bravais lattices with a basis

Our general method for obtaining quasistatic plasmonic band structures detailed in Sec. III further applies to arbitrary Bravais lattices with a basis. In the following, we start by considering Bravais lattices with a basis of two.

1. Bipartite lattices

In the case of a bipartite lattice ($S = 2$), the $4 \times 4$ matrix resulting from the system of equations (24) for the OP polarization $\sigma = x$ can be straightforwardly solved, yielding the two bands ($\tau = \pm$) with dispersion relations

$$\omega_{\tau}^{x}(q) = \omega_0 \sqrt{1 + \frac{\Omega}{\omega_0} |f_{111}^{xx}(q) + \tau f_{122}^{xx}(q)|}.$$  

The resulting band structure is represented in Fig. 2(d) by a dotted line, and reproduces quite well the full quasistatic dispersion in the vicinity of the $\Gamma$ point. In particular, to first order in $|q| < 1$, we find $\omega^{x,\pm}(q) \simeq \omega_0 + \Omega/2 - \pi/\sqrt{2} + 2\pi(q_x d)$ and $\omega^{x,-}(q) \simeq \omega_0 - \Omega(2 + \pi/\sqrt{2})$, demonstrating the presence (absence) of a cusp for the high- (low-)energy plasmonic branch.
dispersion above with the nearest-neighbor result \[96\] with the group velocity.

\[\mathbf{\sigma} \text{ vector}\]

\[\mathbf{\Gamma} \text{ vector}\]

\[\text{FIG. 3. (a) Sketch of a honeycomb lattice with primitive lattice vectors } \mathbf{t}_1 = d (\sqrt{3}, 0) \text{ and } \mathbf{t}_2 = d (\sqrt{3}, \frac{2}{3}), \text{ and basis vector } \mathbf{d}_2 = d (\sqrt{3}, \frac{1}{2}). \text{ (b) Corresponding first Brillouin zone, with primitive reciprocal vectors } \mathbf{b}_1 = \frac{2\pi}{d}(\sqrt{3}, -1) \text{ and } \mathbf{b}_2 = \frac{2\pi}{d}(0, 1). \text{ (c),(d) Quasistatic plasmonic band structure for (c) out-of-plane (OP) and (d) in-plane (IP) polarizations. The solid lines correspond to the full quasistatic dispersion, and the color code to the polarization angle (25). The dashed lines correspond to the nearest-neighbor approximation. Same parameters as in Fig. 2.}\]

\[\begin{align*}
\omega_{\mathbf{n}, \tau}(\mathbf{k}) & = \omega_0 + \tau v_{\mathbf{n}}^\tau |\mathbf{k}| \quad \text{with } v_{\mathbf{n}}^\tau = 3\Omega d/2, \text{ we see that the intrasublattice coupling } f_{\mathbf{n}}^{\tau |\mathbf{G}} \text{ leads to an inconsequential redshift of the Dirac point frequency, while the intersublattice coupling } f_{\mathbf{n}}^{\tau |\mathbf{G}} \text{ renormalizes the precise value of the group velocity.}
\end{align*}\]

Since the Bogoliubov coefficients \((39)\) are negligible as compared to the coefficients \((38)\) close to the \(K\) point, we can safely disregard the former, which amounts to perform the RWA (cf. Appendix A). Within this limit, the associated effective Hamiltonian reads in terms of the spinor operator \(\hat{\Psi}_\mathbf{k} = (b_\mathbf{f}(\mathbf{k}), b_\mathbf{b}(\mathbf{k}))\) as \(H_{\text{eff}} = \sum_\mathbf{k} \hat{\Psi}_\mathbf{k}^\dagger H_{\text{eff}}^\mathbf{k} \hat{\Psi}_\mathbf{k}\), with the massless Dirac Hamiltonian

\[H_{\text{eff}}^\mathbf{k} = [\hbar \omega_0 - \hbar \Omega f_{\mathbf{n}}^{\tau |\mathbf{G}}(\mathbf{K})] \end{align*}\]

where \(\mathbb{I}_n\) is the \(n \times n\) identity matrix and \(\mathbf{\sigma} = (\sigma_x, \sigma_y, \sigma_z)\) is the vector of Pauli matrices acting on the sublattice pseudospin \(1/2\). Therefore, the long-range character of the dipole–dipole interaction does not rule out the massless Dirac nature of the plasmonic quasiparticles in the vicinity of the \(K\) point, and the nearest-neighbor approximation is sufficient in catching the relevant physics. The same conclusion applies to the inequivalent Dirac point located at \(K'\).

We further observe in Fig. 3(c) the presence of a cusp for the upper \((\tau = +)\) band when all quasistatic interactions are taken into account, while no cusp appears for the lower \((\tau = -)\) band. Notice that the upper (lower) band corresponds to bright (dark) modes, where the two dipolar LSPs are in-phase (out-of-phase) within each unit cell.

We now turn to the description of the IP polarized plasmonic modes. We show in Fig. 3(d) the plasmonic band structure obtained numerically for \(\mathbf{\sigma} = x, y\) as solid lines. The color code corresponding to the polarization angle \((25)\) reveals that two bands correspond to purely transverse plasmons, and two other bands to purely longitudinal plasmons along the high-symmetry axes \(\Gamma K\) and \(\Gamma M\). We also plot for comparison the dispersion relations with nearest-neighbors coupling only. In the latter case, the intrasublattice sums \(f_{\mathbf{n}}^{\tau |\mathbf{G}}(\mathbf{q})\) vanish, so that the \(8 \times 8\) system given by Eq. \((24)\) simplifies and can be straightforwardly solved analytically. The resulting four dispersion relations read

\[\omega_{\mathbf{n}, \tau}^{\tau |\mathbf{q}}(\mathbf{k}) = \begin{cases} 
\omega_0 \left[1 + \sqrt{2} \frac{\Omega}{\omega_0} \sqrt{G_1(\mathbf{q})} \pm \sqrt{G_2(\mathbf{q})} \right]^{1/2}, \\
\omega_0 \left[1 - \sqrt{2} \frac{\Omega}{\omega_0} \sqrt{G_1(\mathbf{q})} \pm \sqrt{G_2(\mathbf{q})} \right]^{1/2},
\end{cases}\]

with

\[G_1(\mathbf{q}) = |G_{\mathbf{n}, 12}(\mathbf{q})|^2 + |G_{\mathbf{n}, 12}(\mathbf{q})|^2 + 2|G_{\mathbf{n}, 12}(\mathbf{q})|^2\]

\[G_2(\mathbf{q}) = |G_{\mathbf{n}, 12}(\mathbf{q})|^2 + |G_{\mathbf{n}, 12}(\mathbf{q})|^2 + 2|G_{\mathbf{n}, 12}(\mathbf{q})|^2\]
and
\[
\mathcal{G}_2(q) = \left[|f_{nn,12}(q)|^2 - |f_{nn,12}^y(q)|^2\right]^2 \\
+ 4|f_{nn,12}^x(q)|^2 \left(|f_{nn,12}^y(q)|^2 + |f_{nn,12}^y(q)|^2\right) \\
+ 8 \text{Re} \left\{ f_{nn,12}(q)f_{nn,12}^y(q) \left[f_{nn,12}^x(q)\right]^2 \right\}, \tag{44}
\]

In the expressions above, the intersublattice sums within the nearest-neighbor approximation read as \( f_{nn,12}(q) = e^{i\mathbf{q} \cdot \mathbf{e}_1 - \frac{\Omega}{\omega_0}}(e^{i\mathbf{q} \cdot \mathbf{e}_2 + e^{i\mathbf{q} \cdot \mathbf{e}_3}), f_{nn,12}^y(q) = -2e^{i\mathbf{q} \cdot \mathbf{e}_1 + \frac{\Omega}{\omega_0}}(e^{i\mathbf{q} \cdot \mathbf{e}_2 + e^{i\mathbf{q} \cdot \mathbf{e}_3}), \) and \( f_{nn,12}^x(q) = \frac{3\omega_0^2}{4}(e^{i\mathbf{q} \cdot \mathbf{e}_3} - e^{i\mathbf{q} \cdot \mathbf{e}_2}). \) As can be seen from Fig. 3(d), the nearest-neighbor approximation is sufficient to qualitatively describe the plasmonic dispersion relations, apart from the second less energetic band for wavenumbers close to the \( \Gamma \) point, where a cusp appears in the full quasistatic band structure. Importantly, we note the presence of two inequivalent conical intersections (where the band degeneracy point occurs at the frequency \( \omega_0 \)) at the \( K \) and \( K' \) points of the 1BZ for IP polarized modes. In the vicinity of the \( K \) point, we find for the second and third bands \( \omega_{\text{nn}}(k) \approx \omega_0 \pm v_{\text{nn}}^z|k|, \) with \( v_{\text{nn}}^z = 9\Omega/4. \) The presence of conical intersections for IP polarized modes has been reported by Han et al. \cite{95} using a numerical solution to Maxwell’s equations. Our transparent method allows us to analytically describe such a complex band structure hosting Dirac-like bosonic modes.

2. Tripartite lattices

For tripartite lattices (\( S = 3 \)), the \( 6 \times 6 \) system obtained from Eq. (24) for \( \sigma = z \) can still be solved analytically, yielding the three plasmonic bands

\[
\omega_{\pm}^z(q) = \sqrt{\omega_0^2 \left[ 1 + 2 \frac{\Omega}{\omega_0} \left[ f_{11}^z(q) + s_+(q) + s_-(q) \right] \right]^2}, \tag{45a}
\]

\[
\omega_{\pm}^z(q) = \omega_0 \left[ 1 + \frac{\Omega}{\omega_0} \left[ 2f_{11}^z(q) - s_+(q) - s_-(q) \right] \pm i\sqrt{3}(s_+(q) - s_-(q)) \right]^{1/2}, \tag{45b}
\]

where

\[
s_\pm(q) = \frac{\Pi_{-}^z(q) \pm i \sqrt{\frac{\left[ \Sigma_{z}(q) \right]^3}{3} - [\Pi_{+}^z(q)]^2}}{1/3}, \tag{46}
\]

with

\[
\Sigma_{z}(q) = |f_{12}^z(q)|^2 + |f_{13}^z(q)|^2 + |f_{23}^z(q)|^2 \tag{47}
\]

and

\[
\Pi_{+}^z(q) = \text{Re} \left\{ f_{12}^z(q)f_{13}^z(q)f_{23}^z(q) \right\}. \tag{48}
\]
persions in tight-binding models with nearest-neighbor interactions (see, e.g., Refs. [106–110]). It is therefore of interest to study if these features survive in the case of plasmonic metasurfaces, where the long-range nature of the dipolar interaction might qualitatively change the band structure.

We plot in Fig. 4(c) the three dispersion relations (45) for the Lieb lattice and for OP polarization as solid lines. For comparison, we also show by dashed lines the plasmonic band structure within the nearest-neighbor approximation, for which we have
\[ f_{nn,11}(q) = f_{nn,22}(q) = f_{nn,33}(q) = f_{nn,23}(q) = 0, \]
\[ f_{nn,12}(q) = 2 \cos(q_yd), \]
\[ f_{nn,13}(q) = 2 \cos(q_yd). \]

Within this approximation, the resulting band structure is characterized by the presence of a flat band \( \omega_n^{zz}(q) = \omega_0 \) and two dispersive bands \( \omega_n^{z_1}(q) = \omega_0 [1 + 4n(d) \sqrt{\cos^2(q_xd) + \cos^2(q_yd)}]^{1/2} \).

As shown in Fig. 4(c), the long-range dipolar interactions affect differently the topological features of the plasmonic band structure described above (compare solid and dashed lines). Indeed, while the presence of a conical dispersion in the vicinity of the \( M \) point is not ruled out by long-range interactions (as is the case for the honeycomb array, see Sec. IV B 1), the band which is flat in the whole 1BZ within the nearest-neighbor approximation becomes dispersive, and is only locally flat close to the \( M \) point. One can understand these features by expanding the band structure (45) in the vicinity of the \( M \) point. With \( q = M + k \), to linear order in \( |k| \ll |M| \) we find
\[ f_{zz}^{zz}(q) \approx f_{zz}^{zz}(M) \approx -0.331, \]
\[ f_{zz}^{zz}(q) \approx -1.65k_xd, \]
\[ f_{13}^{zz}(q) \approx -1.65k_yd, \]
\[ f_{23}^{zz}(q) \approx 0, \]
resulting in a band \( \omega_0(k) \approx \omega_0 - \Omega f_{11}(M) \) which is only locally flat, and two conical dispersions \( \omega_{1}^{z_1}(k) \approx \omega_0 - \Omega f_{11}(M) \pm v^*|k| \) with renormalized group velocity \( v^* = 1.65\Omega d \).

Interestingly, the plasmonic Hamiltonian within the RWA for modes polarized in the \( z \) direction takes the form
\[ H_{zz}^{\text{eff}} = \sum_k \hat{\Psi}_k^\dagger \hat{H}_{k}^{\text{eff}} \hat{\Psi}_k, \]
where \( \hat{H}_k^\text{eff} = [\hbar \omega_0 - \hbar \Omega |f_{11}^{zz}(M)|] 33 - \hbar v^* \mathbf{S} \cdot \mathbf{k}, \)
and
\[ \mathbf{S} = (S_x, S_y, S_z). \]

Here, the pseudospin-1 matrices (corresponding to the three inequivalent sublattices of the Lieb lattice) are defined as
\[ S_x = \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad S_y = \begin{pmatrix} 0 & 0 \ & 1 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix}, \quad S_z = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & -i \\ 0 & i & 0 \end{pmatrix}, \]
and fulfill the angular momentum algebra \( [S_i, S_j] = i \epsilon_{ijk} S_k \), with \( \epsilon_{ijk} \) the Levi-Civita symbol. The matrices \( S_x, S_y, \) and \( S_z \) (which correspond, respectively, to the Gell-Mann matrices \( \lambda_1, \lambda_4, \) and \( \lambda_7 \)) therefore correspond to a three-dimensional representation of the special unitary group \( SU(2) \). However, contrary to the Pauli matrices, they do not form a Clifford algebra (i.e., \( \{ S_i, S_j \} \neq 2 \delta_{ij} 1_3 \)), so that Eq. (49) does not correspond to a massless Dirac Hamiltonian [112], despite presenting a conical spectrum.

We show in Fig. 4(d) the plasmonic dispersion relations calculated numerically for \( \sigma = x, y \) as solid lines. We further plot, for comparison, the numerical results with nearest-neighbor couplings only. We observe from the figure that for IP polarization, the long-range dipolar interactions completely reconstruct the plasmonic band structure. Notably, the topological features (flat bands and conical dispersions) occurring in the nearest-neighbors coupling approximation are not preserved when long-range interactions are included.

V. RADIATIVE FREQUENCY SHIFTS AND RADIATIVE LINEWIDTHS OF THE PLASMONIC BAND STRUCTURE

We now consider the effects of the photonic environment alone [encapsulated in the Hamiltonian (8)] onto the collective plasmonic excitations supported by our generic metasurface of ordered metallic nanoparticles. The plasmon–photon interaction [cf. Eq. (9)] leads to two effects crucially affecting the quasistatic band structure discussed above: (i) the photon-induced frequency shifts resulting from retardation effects in the dipole–dipole interaction renormalize the quasistatic plasmonic dispersion (see Sec. VA); (ii) moreover, the spontaneous decay of plasmons into free photons leads to a finite radiative lifetime of the collective excitations, and consequently to a broadening of the plasmon lines (see Sec. VB).

A. Photonic-induced frequency shifts

1. Perturbative calculation for arbitrary polarization

We start our analysis of the effects of the photonic environment onto the quasistatic plasmonic dispersion by considering the radiative frequency shifts induced by the light–matter coupling. Along the lines of Refs. [38, 53, 65], we treat the plasmon–photon coupling Hamiltonian (9) to second order in standard nondegenerate perturbation theory, yielding for the collective plasmon energy levels the result
\[ E_{n_{zz}}(q) = \hbar \omega^*_{zz}(q) + E_{n_{zz}}^{(1)}(q) + E_{n_{zz}}^{(2)}(q). \]

The first term in the right-hand side of the equation above corresponds to the energy levels of the unperturbed Hamiltonian \( H_{zz}^{(2)}(q) \) [cf. Eq. (20)]. The first-order (in \( \hbar \omega_{\text{pl-ph}} \)) correction stems from the diamagnetic term in Eq. (9) (proportional to the vector potential squared) and reads
\[ E_{n_{zz}}^{(1)}(q) = 2\pi SNm_0^2 \alpha^3 \sum_k \frac{1}{\omega_k}. \]

Since the latter expression does not depend on the quantum number \( n_{zz}(q) \), it does not participate to the
renormalization of the collective plasmon frequency, and merely represents an irrelevant global energy shift. The second-order correction to \( n_\|^2(q) \) arises from the first (paramagnetic) term on the right-hand side of Eq. (9) and reads

\[
E_{n_\|}^{(2)} = \frac{\pi \hbar \omega_0^3}{V} \sum_{k, \lambda k} \frac{1}{\omega_k} \left\{ \frac{n_\|^2(q)}{\omega_\|^2(q) - \omega_k} \right\} \times \left[ F_{k,q}^- \sum_{\sigma} (\hat{\sigma} \cdot \hat{\lambda}_k) e^{-i(q-k) \cdot \mathbf{d}_s} P_{\tau s}^{\sigma\sigma}(q) \right]^2
\]

\[
- \frac{n_\|^2(q) + 1}{\omega_\|^2(q) + \omega_k} \times \left[ F_{k,q}^+ \sum_{\sigma} (\hat{\sigma} \cdot \hat{\lambda}_k) e^{-i(q+k) \cdot \mathbf{d}_s} P_{\tau s}^{\sigma\sigma}(q) \right]^2 \right\}.
\]

In Eq. (52), we have defined the array factor

\[
F_{k,q}^\pm = \frac{1}{\sqrt{N}} \sum_{\mathbf{R}} e^{i(q \pm k) \cdot \mathbf{R}}
\]

and

\[
P_{\tau s}^{\sigma\sigma}(q) = u_{\tau s}^{\sigma\sigma}(q) + v_{\tau s}^{\sigma\sigma}(q).
\]

In the large-metasurface limit (\( N \gg 1 \)), the modulus squared of the array factor above entering Eq. (52) takes the simpler form

\[
|F_{k,q}^\pm|^2 \simeq \frac{(2\pi)^2}{|t_1 \times t_2|} \delta(q_x \pm k_x) \delta(q_y \pm k_y),
\]

so that the radiative frequency shift, defined through the relation \( \delta^\tau(q) = \left[ E_{n_\|}(q) + 1 - E_{n_\|}(q) / h - \omega_\|^2(q) \right] \), is given by

\[
\delta^\tau(q) = \pi \omega_0^3 \frac{a^3}{\Omega} \sum_{k, \lambda k} \left\{ \frac{\sum_{\sigma} (\hat{\sigma} \cdot \hat{\lambda}_k) P_{\tau s}^{\sigma \sigma}(q)}{\omega_k} \right\}^2
\]

\[
\times \left[ \frac{|F_{k,q}^\pm|^2}{\omega_\|^2(q) - \omega_k} - \frac{|F_{k,q}^\pm|^2}{\omega_\|^2(q) + \omega_k} \right].
\]

Notably, the calculation leading to the above result does not need the introduction of an ultraviolet frequency cut-off for the photonic degrees of freedom [which prevent to take into account photons with a wavelength smaller than the nanoparticle size, cf. the dipolar approximation in Eq. (9)], as is the case for nanoparticle dimers [38] and linear chains [53, 65]. We have checked that the inclusion of such a cutoff, of the order of \( k_c \simeq 1/a \), does not significantly modify the result (59). Notice also that Eq. (59) is not periodic in reciprocal space since we consider only the interaction between the collective plasmons and photons for which the associated light cone belongs to the 1BZ.

We plot in panels (a)–(c) of Fig. 5 the plasmonic band structure for OP-polarized modes \( \omega_\|^2(q) + \delta^\tau(q) \) including Eq. (59) for (a) the square, (b) the honeycomb, and (c) the Lieb lattices along the high symmetry lines of their respective 1BZ. As shown in Fig. 5(a) for the square lattice, the radiative frequency shift \( \delta^\tau(q) \) induces an important renormalization of the quasistatic band structure, which is of purely transverse nature [see Fig. 2(c)]. A similar effect is also observed for other simple Bravais lattices, as exemplified for the rectangular and hexagonal ones in

2. Radiative frequency shifts for out-of-plane polarized plasmonic modes

In order to proceed with an explicit evaluation of the radiative frequency shifts (56), we first concentrate on the case of plasmonic modes polarized perpendicular to the plane of the metasurface, for which we have \( \sigma = \varepsilon = z \). The “selection rule” \( P_{\tau s}^{\sigma\sigma}(q) = 0 \) for \( \sigma = x, y \) allows us to simplify Eq. (56), yielding

\[
\delta^\tau(q) = \frac{\pi \omega_0^3 a^3}{\Omega} |t_1 \times t_2| \frac{\sum_{\sigma} P_{\tau s}^{\sigma\sigma}(q)}{|\omega_\|^2(q) - \omega_k|} \times \left[ \delta_{\sigma\sigma'} - \frac{k_\sigma k_{\sigma'}}{|k|^2} \right].
\]

The summation over photon polarizations above can then be performed using the general result [99]

\[
\sum_{\lambda_k} \frac{\delta^\tau(q)}{\omega_\|^2(q) - \omega_k} = 0 \quad \text{for} \quad \sigma = \varepsilon = z.
\]
FIG. 5. The colored lines display the plasmonic band structure $\omega_\varepsilon(q) + \delta_\varepsilon(q)$ (scaled by $\omega_0$) including retardation effects and the corresponding radiative frequency shift (56) for (a),(d) the square, (b),(e) the honeycomb, and (c),(f) the Lieb lattices. Panels (a)–(c) [(d)–(f)] show the OP (IP) modes. For comparison purposes, the thin grey lines reproduce the corresponding quasistatic dispersion relations $\omega_\varepsilon(q)$, while the dashed grey lines depict the light cone within each 1BZ. In the figure, $d = 3a$ and $k_0a = 0.15$.

Appendix B. Comparing the fully-retarded dispersion relation (red line) to the quasistatic one (thin grey line), we first observe that the retarded one diverges at the intersection of the quasistatic band structure with the light cone (dashed lines). Such a divergence was already reported in the literature for the transverse modes in one-dimensional plasmonic chains by means of involved numerical calculations based on the fully-retarded solutions to Maxwell’s equations [41–47, 52]. Recently, studies [53, 65] using the open quantum system approach which we use in this work have shown a similar divergence occurring in the dispersion relations of transverse plasmonic modes in chains. Importantly, the results presented in Ref. [53] show an excellent quantitative agreement with numerical electromagnetic calculations for regular nanoparticle chains. Therefore, we expect that the open quantum system approach is justified for studying the retardation effects in two-dimensional plasmonic metamaterials. We here point out that the singularities observed in Fig. 5 arise from calculations based on a second-order perturbative treatment of the light–matter coupling. Consequently, important variations from the
LSP resonance frequency $\omega_0$ should be treated carefully. Notice also that the renormalized dispersion relation diverges as the inverse of a square root [see Eq. (59)] instead of logarithmically as is the case for one-dimensional arrays [53, 65].

As can be seen from Fig. 5(a), taking into account the radiative shift implies that the cusp appearing at the $\Gamma$ point of the IBZ within the quasistatic approximation disappears (compare the red and solid lines in the figure). In the vicinity of the $\Gamma$ point ($|q|d \ll 1$), Eq. (59) applied to the square lattice reduces (considering $\Omega/\omega_0 \ll 1$) to $\delta^z(\mathbf{q}) \approx 2\pi\Omega|q|d + O(|q|d^3)$. Such a linear $|q|$-dependence cancels out exactly the one of $\omega^z(\mathbf{q})$ in this regime of parameters [cf. Eq. (34)]. We hence find $\omega^z(\mathbf{q}) + \delta^z(\mathbf{q}) \approx \omega_0 + \Omega(4 + \sqrt{2}\pi + (4|q|d)^2(\pi/\sqrt{2} - 1))$ close to the $\Gamma$ point, leading to a quadratic dependence of the dispersion relation. The results presented in Fig. 5(a) thus demonstrate that considering retardation effects is crucial for the study of the collective plasmonic modes in metasurfaces of near-field coupled nanoparticles, since the dispersion relations are qualitatively affected by the interactions with free photons. Such renormalization effects are not that prominent in one-dimensional plasmonic systems, where, apart from the divergence of the dispersion relations at the crossing of the light cone, the dispersion relations are qualitatively unaffected by retardation effects [41–47, 52, 53, 65].

We show in Fig. 5(b) the band structure including retardation effects (colored lines) of the OP plasmonic modes for the honeycomb lattice. For comparison, we also reproduce as grey solid lines the transverse-polarized quasistatic dispersion (grey solid lines) and the light cone (dashed lines) as well as the absence of a cusp at the $\Gamma$ point. Conversely, the two low-energy bands correspond to dark modes.

### 3. Radiative frequency shifts for in-plane polarized plasmonic modes

In the case of IP polarized plasmonic modes ($\varepsilon = \varepsilon_{\parallel}$), we have $P_{\tau s}^{\varepsilon_{\parallel}z}(\mathbf{q}) = 0$, so that Eq. (56) takes the form

$$\delta^z(\mathbf{q}) = \pi\omega_0^3 a^3 \sum_{k,\lambda_k} \frac{1}{\omega_k} \times \left[ \sum_s \left( \hat{x} \cdot \hat{\lambda}_k P_{\tau s}^{\varepsilon_{\parallel}x}(\mathbf{q}) + \hat{y} \cdot \hat{\lambda}_k P_{\tau s}^{\varepsilon_{\parallel}y}(\mathbf{q}) \right) \right]^2 \times \left[ \frac{|F_{\tau k,\mathbf{q}}^-|^2}{\omega_{\tau s}^z(\mathbf{q}) - \omega_k} - \frac{|F_{\tau k,\mathbf{q}}^+|^2}{\omega_{\tau s}^z(\mathbf{q}) + \omega_k} \right].$$

Using Eq. (58) to perform the summation over photon polarizations and going to the continuum limit, we obtain with Eq. (55) after a lengthy, but straightforward calculation [113], the result

$$\delta^z(\mathbf{q}) = -\frac{\pi\omega_0^3 a^3 |\mathbf{q}|}{|\mathbf{t}_1 \times \mathbf{t}_2| |\omega_{\tau s}^z(\mathbf{q})|^2} \times \left[ \sum_s \left( \mu \frac{P_{\tau s}^{\varepsilon_{\parallel}x}(\mathbf{q})}{|\mathbf{q}|} + q \frac{P_{\tau s}^{\varepsilon_{\parallel}y}(\mathbf{q})}{|\mathbf{q}|} \right) \right]^2 \times \left[ 1 - \frac{c|\mathbf{q}|}{\sqrt{(c|\mathbf{q}|)^2 - [\omega_{\tau s}^z(\mathbf{q})]^2}} \Theta(c|\mathbf{q}| - \omega_{\tau s}^z(\mathbf{q})) \right]$$

$$+ \sum_s P_{\tau s}^{\varepsilon_{\parallel}x}(\mathbf{q})^2 + \sum_s P_{\tau s}^{\varepsilon_{\parallel}y}(\mathbf{q})^2 \right] \times \frac{[\omega_{\tau s}^z(\mathbf{q})]^2}{c|\mathbf{q}| \sqrt{(c|\mathbf{q}|)^2 - [\omega_{\tau s}^z(\mathbf{q})]^2}} \Theta(c|\mathbf{q}| - \omega_{\tau s}^z(\mathbf{q})) \right].$$

We display in Fig. 5(d) the fully-retarded dispersion relation of the IP modes in the square lattice (see red and orange lines). The quasistatic band structure from...
Fig. 2(d) is reproduced here by grey lines for comparison. Three important features appear from the comparison of these two results. The low-energy band (cf. red line), which corresponds essentially to transverse modes, present a singularity at the crossing of the quasistatic dispersion relation with the light cone. Conversely, the high-energy band (cf. orange line), which corresponds essentially to longitudinal modes, does not present such a singularity at the crossing, as is the case for plasmonic chains [41–47, 52, 53, 65]. In addition, the cusp that presents the quasistatic high-energy band in the vicinity of the Γ point is washed away by retardation effects [see Eq. (58)] and going to the continuum limit. Performing the remaining integrals in Cartesian coordinates using Eq. (55) yields [113]

\[
\gamma_\tau^c(q) = \frac{2\pi \omega_0^3 a^3}{V} \sum_s \frac{1}{\omega_k} \delta(\omega_k^c(q) - \omega_k) \times \left| P_{\tau\tau}^\sigma(q) \right|^2 \times \Theta(\omega_k^c(q) - |c|)
\]

(65)

which is valid for both OP and IP polarizations.

2. Radiative linewidths for out-of-plane polarized plasmonic modes

We first consider the specific case where \( \sigma = C \approx z \), corresponding to OP plasmonic modes. In such a case, Eq. (63) reduces to

\[
\gamma_\tau^c(q) = 2\pi \omega_0^3 a^3 \sum_{s} \left| \sum_{k,\lambda_k} F_{k,q}^{-1} \right|^2 \left( \delta(\omega_k^c(q) - \omega_k) \right) \times \delta(\omega_k^c(q) - \omega_k),
\]

(63)

where \( F_{k,q}^{-1} \) and \( P_{\tau\tau}^\sigma(q) \) are defined in Eqs. (53) and (54), respectively. In the large metasurface limit (\( N \gg 1 \)), using Eq. (55) yields the result

\[
\gamma_\tau^c(q) = 2\pi \omega_0^3 a^3 \sum_{s} \left| \sum_{k,\lambda_k} F_{k,q}^{-1} \right|^2 \left( \delta(\omega_k^c(q) - \omega_k) \right) \times \delta(\omega_k^c(q) - \omega_k),
\]

(63)

which is valid for both OP and IP polarizations.

B. Radiative linewidths

1. Fermi’s golden rule for arbitrary polarization

We now concentrate on the evaluation of the radiative decay rate of the collective plasmons. To this end, we treat the plasmon–photon coupling Hamiltonian (9) as a weak perturbation to the plasmonic subsystem. In such a regime, the radiative decay rate of the plasmonic eigenmode \( |\Gamma^c_\tau(q)\rangle \) with band index \( \tau \), polarization \( \varepsilon \) and wavevector \( q \) is given by the Fermi golden rule expression

\[
\gamma_\tau^c(q) = 2\pi \omega_0^3 a^3 \sum_{k,\lambda_k} \frac{1}{\omega_k} \delta(\omega_k^c(q) - \omega_k) \times \left| P_{\tau\tau}^\sigma(q) \right|^2,
\]

(62)

where \( F_{k,q}^{-1} \) and \( P_{\tau\tau}^\sigma(q) \) are defined in Eqs. (53) and (54), respectively. In the large metasurface limit (\( N \gg 1 \)), using Eq. (55) yields the result

\[
\gamma_\tau^c(q) = 2\pi \omega_0^3 a^3 \sum_{k,\lambda_k} \frac{1}{\omega_k} \delta(\omega_k^c(q) - \omega_k) \times \delta(\omega_k^c(q) - \omega_k),
\]

(63)

since \( P_{\tau\tau}^\sigma(q) = 0 \) for \( \sigma = x, y \). Equation (64) can be easily evaluated by summing over photon polarizations [cf. Eq. (58)] and going to the continuum limit. Performing the remaining integrals in Cartesian coordinates using Eq. (55) yields [113]

\[
\gamma_\tau^c(q) = \frac{2\pi \omega_0^3 a^3 \delta(\varepsilon |q|^2)}{|t_1 \times t_2| |\omega_k^c(q)|^2} \sum_s P_{\tau\tau}^\sigma(q)^2 \times \Theta(\omega_k^c(q) - |c|) \times \Theta(\omega_k^c(q) - |c|)
\]

(65)

We plot in Fig. 6 the radiative damping rate (65) for OP plasmonic modes for the square [panel (a)], the honeycomb [panel (b)], and the Lieb lattices [panel (c)] along the high symmetry lines connecting the Γ point of their respective 1BZ. Note that we do not show in the figure the results along the entire 1BZ since the damping rate vanishes for wavevectors outside of the light cone, as can be easily inferred from the Heaviside step function in Eq. (65). In Fig. 6, the displayed results are scaled by the radiative decay rate of a single isolated nanoparticle, \( \gamma_0 = 2\omega_0^3 a^3/3c^3 \). For the square lattice [Fig. 6(a)], the OP plasmonic modes present a highly superradiant profile (\( \gamma_\tau(q) \gg \gamma_0 \)) inside of the light cone for wavevectors not too close to the Γ point, while displaying a vanishing rate outside of the light cone. From the figure, we observe that the radiative decay rate \( \gamma_\tau(q) \) increases rapidly as \( q \) moves away from the center of the 1BZ and diverges at wavevectors corresponding to the intersection of the quasistatic band structure with the light cone. Such singularities are related to those observed in the fully-retarded dispersion relation [cf. Fig. 5(a)]. We here point out again that these divergencies arise from our perturbative treatment of the light–matter coupling and, consequently, should be renormalized by an exact treatment of the latter interaction. The superradiant behavior of the radiative damping rate observed in Fig. 6(a) is reminiscent to the one reported for one-dimensional plasmonic chains [37, 41, 42, 44–46, 52, 53, 62, 65]. Such a behavior is nevertheless much more prominent in two-dimensional metamaterials, due to the enhanced constructive interferences between the dipolar electric fields produced by each LSP.

We plot in panels (b) and (c) of Fig. 6 the radiative decay rates of the OP plasmonic modes for the honeycomb and Lieb lattices, respectively. In both cases, one of the plasmonic band shows a similar profile as that in Fig. 6(a). Indeed, in both Figs. 6(b) and 6(c), the red lines display a superradiant behavior which diverges at the intersection of the quasistatic band structure and the
FIG. 6. The colored lines display the radiative damping rate $\gamma^\varepsilon(q)$ from Eq. (63) (scaled by $\gamma_0$) for (a),(d) the square, (b),(e) the honeycomb, and (c),(f) the Lieb lattices. Panels (a)–(c) [(d)–(f)] show the damping for the OP [IP] modes. Same parameters as in Fig. 5.

light cone. Conversely, the blue and cyan lines show the existence of subradiant modes (i.e., $\gamma^\varepsilon(q) \ll \gamma_0$) corresponding to dark, nonradiative modes, for which retardation effects on the plasmonic band structure are essentially negligible [cf. blue and cyan lines in Figs. 5(b) and 5(c)].

3. Radiative linewidths for in-plane polarized plasmonic modes

In the case of IP polarized plasmonic modes ($\varepsilon = \varepsilon_\parallel$), since we have $P_{\tau s}^{\varepsilon \parallel}(q) = 0$, Eq. (63) takes the form

$$\gamma^\varepsilon_\parallel(q) = 2\pi^2 \omega_0^3 \frac{q^3}{\beta}\sum_{k,\lambda_k} \frac{|F_{k,\varepsilon}^{-}|^2}{\omega_k} \delta \left(\omega^\varepsilon_\parallel(q) - \omega_k\right)$$

$$\times \left| \sum_s \left[ (\hat{\varepsilon} \cdot \hat{\lambda}_k) P_{\tau s}^{\varepsilon \parallel x}(q) + (\hat{y} \cdot \hat{\lambda}_k) P_{\tau s}^{\varepsilon \parallel y}(q) \right] \right|^2.$$

(66)
Going to the continuum limit and using Eq. (58), we then obtain [113]

\[
\gamma_{\tau}^{\varepsilon}(q) = -\frac{2\pi\omega_{0}^{2}a^{3}c|q|^{2}}{|t_{1}\times t_{2}|(|\omega_{\tau}^{\varepsilon}(q)|)^{2}} \Theta(\omega_{\tau}^{\varepsilon}(q) - c|q|) \times \left\{ \sum_{s} \frac{q_{x}}{|q|} P_{\tau s}^{x}(q) - \frac{q_{y}}{|q|} P_{\tau s}^{y}(q) \right\}^{2} - \left\{ \sum_{s} P_{\tau s}^{x}(q)^{2} + \sum_{s} P_{\tau s}^{y}(q)^{2} \right\}^{2} \times \frac{[\omega_{\tau}^{\varepsilon}(q)]^{2}}{(c|q|)^{2}}. \tag{67}
\]

We show in Fig. 6(d) the radiative damping rate (67) for IP polarized modes in the square lattice. In the figure, the red line corresponds to the lower transverse plasmonic band in Fig. 5(d) [see also Fig. 2(d)] and presents singularities coinciding with those in Fig. 6(a). Conversely, the orange line in Fig. 6(d) which corresponds to the upper longitudinal band [see Fig. 5(d)] displays an opposite trend, as the radiative decay rate decreases for wavevectors moving away from the center of the 1BZ. This is reminiscent to what has been previously reported for longitudinal plasmonic modes in one-dimensional chains [37, 41, 42, 44–46, 52, 53, 62, 65]. We draw similar conclusions for the honeycomb and Lieb lattices, see Figs. 6(e) and 6(f), respectively. Additionally, some of the bands [cyan lines in Figs. 6(e) and 6(f)] display an almost vanishing radiative decay rate as they correspond to dark, out-of-phase modes.

VI. EFFECTS OF THE ELECTRONIC ENVIRONMENT ON THE COLLECTIVE PLASMON EXCITATIONS

In this section, we now focus on the effects induced by the second environment the collective plasmons are subject to [cf. Eq. (1)], i.e., the electronic environment, which is represented by the Hamiltonian (12). Similarly to the photonic bath which we considered in the preceding Sec. V, the coupling between plasmonic and single-particle electronic degrees of freedom, encapsulated in the Hamiltonian (13), leads to two distinct effects. First, the collective plasmons dissipate their energy by producing electron-hole pairs inside each nanoparticles composing the metasurface, corresponding to the well-known Landau damping (Sec. VI A), and yielding a second (non-radiative) decay channel which adds up to the radiative one. Second, the electronic environment induces an additional renormalization of the quasistatic dispersion relation, which comes on top of the one induced by free photons (Sec. VI B).

A. Landau damping

We start this section by first evaluating the Landau damping of the collective plasmonic modes. To this extent, we treat the coupling (13) between plasmonic and electronic degrees of freedom perturbatively. Within this regime, the zero-temperature Landau damping linewidth \( \Gamma_{\tau}^{\varepsilon}(q) \) of the plasmonic eigenmode \( |\Gamma_{\tau}^{\varepsilon}(q)\rangle \) with band index \( \tau \), polarization \( \varepsilon \), and wavevector \( q \) is given by the Fermi golden rule

\[
\Gamma_{\tau}^{\varepsilon}(q) = \frac{2\pi}{\hbar^{2}}A^{2} \sum_{s} \sum_{e_{h}} \left| \sum_{\sigma} [M_{\tau s}^{\sigma}(q)]^{2} (e|\sigma|h) \right|^{2} \delta(\omega_{\tau}^{\varepsilon}(q) - \omega_{eh}), \tag{68}
\]

where

\[
M_{\tau s}^{\sigma}(q) = u_{\tau}^{\sigma}(q) - v_{\tau}^{\sigma}(q) \tag{69}
\]

is given in terms of the Bogoliubov coefficients entering Eq. (18). In Eq. (68), \( A \) is the coupling constant defined in Eq. (14) and \( \omega_{eh} = (e_{e} - e_{h})/\hbar \) corresponds to the frequency associated to an electron-hole pair, where \( e_{e} \) (\( e_{h} \)) is the energy of a single-particle electron (hole) state in the self-consistent hard-wall potential associated to each nanoparticle. The corresponding dipolar matrix elements \( (e|\sigma|h) \) are given in Appendix D.

Equation (68) can be evaluated by first expending the sum over the polarizations \( \sigma = x, y, z \) in the modulus squared. Then, since

\[
\sum_{m_{e}, m_{h}} \langle e|\sigma|h\rangle\langle h|\sigma'|e \rangle = 0 \tag{70}
\]

for \( \sigma \neq \sigma' \) (cf. Appendix D and Ref. [113] for details), Eq. (68) reduces to

\[
\Gamma_{\tau}^{\varepsilon}(q) = \sum_{s, \sigma} |M_{\tau s}^{\sigma}(q)|^{2} \Sigma(\omega_{\tau}^{\varepsilon}(q)), \tag{71}
\]

where

\[
\Sigma(\omega) = \frac{2\pi}{\hbar^{2}}A^{2} \sum_{e_{h}} \left| \langle e|\sigma|h \rangle \right|^{2} \delta(\omega - \omega_{eh}). \tag{72}
\]

Due to the spherical symmetry of the electronic wavefunctions, we have \( \Sigma^{x}(\omega) = \Sigma^{y}(\omega) = \Sigma^{z}(\omega) = \Sigma(\omega) \), where \( \Sigma(\omega) \) has been evaluated in Ref. [103] using semiclassical expansions. To leading order in \( \hbar \), it reads

\[
\Sigma(\omega) = \frac{3}{4d} \left( \frac{\omega_{0}}{\omega} \right)^{3} g \left( \frac{\hbar\omega}{E_{F}} \right) \tag{73}
\]

4 The typical Fermi temperature of ordinary metals is of the order of \( 10^{4} \) K, so that the zero-temperature limit which we employ here is a very good approximation.
with $v_F$ and $E_F$ the Fermi velocity and energy of the considered metal, respectively. In Eq. (73), $g$ is a monotonically decreasing function of the parameter $\nu = \hbar \omega_0 / E_F$ given by [114, 115]

$$g(\nu) = \frac{1}{3\nu} \left[ (1 + \nu)^{3/2} - (1 - \nu)^{3/2} \right] + \frac{\nu}{4} \left( \sqrt{1 + \nu} - \sqrt{1 - \nu} - \nu \ln \nu \right) + \frac{\nu}{2} \left[ (1 + \nu^2) \ln (1 + \nu) - (1 - \nu^2) \ln (1 - \nu) \right]$$

(74a)

for $\nu \leq 1$ and

$$g(\nu) = \frac{1}{3\nu} \left( 1 + \nu \right)^{3/2} + \frac{\nu}{4} \left( \sqrt{1 + \nu} - 1 - \frac{\nu}{2} \ln \sqrt{\nu} \right)$$

(74b)

for $\nu > 1$. For $\omega = \omega_0$, Eq. (73) corresponds [103] to the Landau decay rate $\Gamma_0$ of a single nanoparticle [114, 116, 117],

$$\Gamma_0 = \Sigma_\sigma (\omega_0) = \frac{3v_F}{4a} \left( \frac{\hbar \omega_0}{E_F} \right).$$

(75)

Using the system of equations (24) together with Eq. (22) enables us to show that [113]

$$\sum_{s\sigma} |M_{rs}^{\sigma}(q)|^2 = \frac{\omega_0}{\omega_F^2(q)},$$

(76)

so that substituting Eq. (73) into Eq. (71) yields for the Landau damping rate of the collective plasmons the analytical expression

$$\Gamma_\sigma^r(q) = \frac{3v_F}{4a} \left( \frac{\omega_0}{\omega_F^2(q)} \right)^4 \left( \frac{\hbar \omega_F(q)}{E_F} \right).$$

(77)

To leading order in the coupling constant (7), the Landau damping decay rate of the collective plasmonic modes thus scales with the nanoparticle size as $1/a$, as it is the case for the single-particle result (75). This is in stark contrast to the radiative linewidth, which increases with the nanoparticle radius as $a^3$ [cf. Eq. (63)].

In Fig. 7, we illustrate the result of Eq. (77) for the special case of the OP modes in the honeycomb lattice. In the figure, the displayed results are scaled by the single-particle Landau damping linewidth (75), and the blue (red) curve corresponds to the lower (higher) energy band in Fig. 3(c). We therefore conclude that the higher the energy of the mode, the lower its nonradiative Landau decay rate, as is the case for single nanoparticles [118]. Importantly, in contrast to the radiative decay rate analyzed in Sec. VB, the Landau damping is nonvanishing over the whole 1BZ, and is of the order of $\Gamma_0$. It is therefore crucial to take into account such a decay mechanism, especially for small nanoparticles where it dominates over radiative losses.

\section*{B. Electronic-induced frequency shift}

We now calculate analytically the frequency shift induced by the electronic environment on the plasmonic band structure. Treating the plasmon-electron coupling Hamiltonian (13) to second order in perturbative theory yields for the collective plasmon energy levels $\xi_N(q)\hbar \omega_F(q) + \mathcal{E}_{\sigma(n)}^1(q) + \mathcal{E}_{\sigma(n)}^2(q)$. While the first-order correction $\mathcal{E}_{\sigma(n)}^1(q)$ vanishes due to the selection rules contained in the coupling Hamiltonian (13), the zero-temperature second-order correction reads as

$$\mathcal{E}_{\sigma(n)}^2(q) = \frac{\Lambda^2}{\hbar^2} \sum_s \sum_{eh} \left[ \frac{\xi_N(q)}{\omega_F(q) - \omega_{eh}} \sum_\sigma |M_{rs}^{\sigma}(q)|^2 \langle e|\sigma|h \rangle \right]^2 - \frac{\xi_N(q) + 1}{\omega_F(q) + \omega_{eh}} \left| \sum_\sigma M_{rs}^{\sigma}(q) \langle e|\sigma|h \rangle \right|^2.$$

(78)

The electronic-induced frequency shift, defined as $\Delta^r_N(q) = [\mathcal{E}_{\sigma(n)}^2(q) + 1 - \mathcal{E}_{\sigma(n)}^1(q)]/\hbar$, is therefore given by

$$\Delta^r_N(q) = \frac{\Lambda^2}{\hbar^2} \sum_s \sum_{eh} \left[ \frac{1}{\omega_F(q) - \omega_{eh}} \sum_\sigma |M_{rs}^{\sigma}(q)|^2 \langle e|\sigma|h \rangle \right]^2 - \frac{1}{\omega_F(q) + \omega_{eh}} \left| \sum_\sigma M_{rs}^{\sigma}(q) \langle e|\sigma|h \rangle \right|^2.$$

(79)

Using Eqs. (70), (73), and (76), the expression (79) above can be rewritten along the lines of Ref. [103] as

$$\Delta^r_N(q) = \frac{\omega_0}{\omega_F(q)} \frac{1}{\pi} \int_{-\infty}^{+\infty} d\omega \frac{\omega \Sigma(\omega)}{|\omega_F(q)|^2 - \omega^2}.$$

(80)

To evaluate the principal value integral in Eq. (80), we must replace the lower boundary by a cutoff frequency.
Such a cutoff arises from the fact that the particle-hole pairs that contribute to the frequency shift $\Delta \epsilon(q)$ in Eq. (79) belong to the high-energy sector of the RPA Hilbert space, while the collective plasmonic excitation is a superposition of particle-hole pairs of the restricted low-energy subspace [103, 114, 118]. In analogy with the single nanoparticle case [103], we choose the cutoff frequency to be $\omega^c_0(q) = \eta \overline{\Gamma}_\tau(q)$, where $\eta$ is a constant of the order of one. Equation (80) thus reads

$$
\Delta \epsilon(q) = \frac{\omega_0}{\omega^c_0(q)} \frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{d\omega}{\omega^c_0(q) - \omega^c_0(q)} \omega \Sigma(\omega) |\omega^c_0(q) - \omega|^2.
$$

Evaluating the remaining integral within the semiclassical limit $k_F a \gg 1$, with $k_F$ the Fermi wavevector, yields for the electronic frequency shift the result [103, 113, 119]

$$
\Delta \epsilon(q) = \frac{\omega_{\omega}}{4 \pi} \mathcal{Z}(\omega^c_0(q))
$$

where

$$
\mathcal{Z}(\omega) = \frac{4}{15 \pi} \sqrt{\frac{E_F}{\hbar \omega}} \left( \frac{\omega_0}{\omega}\right)^4 
\times \left[ \ln \left( \frac{8 \hbar \omega k_F a}{3 \eta E_F g(\hbar \omega/E_F)} \left[ \frac{\omega}{\omega_0} \right]^4 - \frac{\pi}{2} - \frac{4}{3} \right) \right],
$$

Note that the frequency shift (82) scales with the size of the nanoparticles as $1/a$, up to a logarithmic correction. As is the case for the Landau damping linewidth (77), such a frequency renormalization is therefore of relevance only for the smallest nanoparticles. Moreover, the electronic shift of the collective plasmons involves only the plasmonic band structure $\omega^c_0(q)$ in contrast with the radiative frequency shifts [cf. Eqs. (59) and (61)] which depend on the eigenvectors as well. Notice that substituting $\omega_0$ with $\omega^c_0(q)$ in Eq. (82) allows to recover the electronic-induced frequency redshift of an isolated nanoparticle [103],

$$
\Delta_0 = \frac{3 \omega_{\omega}}{4 a} \mathcal{Z}(\omega_0).
$$

We plot in Fig. 8 the result of Eq. (82) applied to the special case of the OP collective plasmonic modes in the honeycomb lattice. As in Fig. 7, the blue (red) line in Fig. 8 corresponds to the lower (higher) energy band in Fig. 3(c). The displayed results are scaled by the absolute value of the frequency shift (84) corresponding to a single nanoparticle. Immediately noticeable from the figure is that the electronic shift (82) is negative in the entire 1BZ, thus corresponding to a redshift, and is of the order of $\Delta_0$. This is in contrast to the radiative frequency shift (56), whose sign is both depending on the wavevector and polarization of the collective mode (see Sec. V A). Finally, the higher the energy of the mode, the lower is its associated electronic shift. Such a conclusion is reminiscent of what occurs in isolated nanoparticles [118].

![Figure 8](image_url)

**VII. OBSERVABILITY OF THE COLLECTIVE PLASMONIC MODES**

Experimentally, the ability to observe the plasmonic dispersion relations presented, e.g., in Fig. 5, is governed by the resolution of the separation between the bands with respect to their respective linewidths. The spectral function $A(\omega, q)$, which characterizes the response of the system to an external perturbation at a given frequency $\omega$ and inplane wavevector $q$, is the key quantity which is usually determined in a spectroscopy experiment (using, e.g., photons or hot electrons). Assuming a Breit–Wigner form for $A(\omega, q)$, we have

$$
A_{\text{OP}}(\omega, q) \propto \frac{1}{[\omega - \omega_0^c(q)]^2 + [\tau_0^c(q)/\pi]^2},
$$

for the OP modes and

$$
A_{\text{IP}}(\omega, q) \propto \frac{1}{[\omega - \omega_0^c(q)]^2 + [\tau^c_0(q)/\pi]^2},
$$

for the IP modes, respectively. In Eq. (85), the renormalized resonance frequency

$$
\tilde{\omega}_n^c(q) = \omega_0^c(q) + \delta^c_\tau(q) + \Delta^c_\tau(q)
$$

takes into account both the radiative shift $\delta^c_\tau(q)$ [cf. Eq. (56)] and the electronic one $\Delta^c_\tau(q)$ [cf. Eq. (82)]. Additionally, the quantity

$$
\tau_n^c(q) = \gamma_\tau(q) + \Gamma^c_\tau(q) + \gamma_0
$$

is the total linewidth of the plasmonic modes and includes three distinct contributions: (i) the radiative losses $\gamma_\tau(q)$ [Eq. (63)], (ii) the Landau damping $\Gamma^c_\tau(q)$ [Eq. (68)], and (iii) the Ohmic losses, inherent to any (bulk) metal, characterized by the damping rate $\gamma_0$, and which we assume to be mode- and size-independent.
FIG. 9. Spectral function (85) for (a),(d) the square, (b),(e) the honeycomb, and (c),(f) the Lieb lattices. Panels (a)–(c) [(d)–(f)] show the results for the OP [IP] modes. In the figure, we choose the parameters $\hbar \omega_0/E_F = 0.47$ and $k_0/k_F = 1.1 \times 10^{-3}$, corresponding to silver nanoparticles. In addition, we take $d = 3a$ and $k_0a = 0.15$ (corresponding to $a = 11$ nm). The Ohmic losses $\gamma_O$ entering the total damping rate (87) are neglected.
FIG. 10. Spectral function (85) for (a),(d) the square, (b),(e) the honeycomb, and (c),(f) the Lieb lattices. Panels (a)–(c) [(d)–(f)] show the results for the OP [IP] modes. The Ohmic losses $\gamma_0 = 0.027\omega_0$ entering the total damping rate (87) and corresponding to Ag [120] are taken into account. The other parameters are the same as in Fig. 9.
We show in Figs. 9 and 10 the spectral function (85) for both the OP and IP modes and for the square, the honeycomb, and the Lieb lattices along the high-symmetry lines of their respective 1BZ as a function of $\omega$. The parameters used in both figures are $\omega_0 = 2.6\,\text{eV}$/nm and $E_F = 5.5\,\text{eV}$, corresponding to Ag nanoparticles [2].

The interparticle distance is $d = 3a$ and the (reduced) nanoparticle radius is $k_0 a = 0.15$ (corresponding to $a = 11\,\text{nm}$). In Fig. 9, the Ohmic losses entering Eq. (87) are neglected, while in Fig. 10 we consider the case where $\gamma_0 = 70\,\text{meV}$/nm, which we extracted from the experiments of Ref. [120] on silver clusters.

As can be seen in Fig. 9(a), which displays $A(\omega, q)$ (without Ohmic losses) for the OP modes in the square lattice, the spectral function is reminiscent of the fully-retarded dispersion relation shown in Fig. 5(a), since the electronic shift (82) only induces a finite $q$-dependent redshift of the band structure. In the figure, we clearly distinguish two different profiles corresponding to wavevectors inside ($|q| \lesssim k_0$) and outside ($|q| \gtrsim k_0$) of the light cone. Within the light cone, the total linewidth (87) is dominated by the radiative damping [see Fig. 6(a)], so that it is difficult to resolve the plasmonic dispersion relation. However, outside of the light cone, only the Landau damping (77) contributes to the total linewidth of the spectral function, allowing for a clear resolution of the plasmonic band. Note that, since the plasmonic modes outside of the light cone are essentially dark, nonoptical techniques are required to excite such modes. Dark modes in nanoparticle dimers and chains have been observed using electron energy loss spectroscopy experiments [27–29], and such a technique may be transposed to study plasmonic metasurfaces.

We plot in panels (b) and (c) of Fig. 9 the spectral function (85a) corresponding to the OP modes for the honeycomb and Lieb lattices, respectively. In both cases, we observe a similar trend as that in Fig. 9(a). Indeed, the entire band structure is clearly visible outside of the light cone in both Figs. 9(b) and 9(c), while the upper bands inside the light cone display a rather large linewidth. However, we see that the lower bands for both the honeycomb and the Lieb lattices are still well resolved inside of the light cone. These dispersion relations correspond to dark plasmonic modes, so that their radiative linewidths for wavevectors $|q| \lesssim k_0$ are nearly vanishing [see the blue line in Fig. 6(b) and the blue and cyan lines in Fig. 6(c)], and only the Landau damping contributes to the observed linewidth.

In panels (d)–(f) of Fig. 9, we display the spectral function (85b) for the IP plasmonic modes for (d) the square, (e) the honeycomb, and (f) the Lieb lattices, respectively. Similar conclusions as that drawn above for the OP modes can be put forward: while the nonradiative bands outside of the light cone are clearly visible, the bright ones inside of the light cone are essentially almost not resolvable.

For completeness, we show in Fig. 10 the spectral functions including in the total decay rate $\gamma_{\tau}(q)$ entering Eq. (85) a nonvanishing Ohmic damping rate $\gamma_0$ corresponding to silver and extracted from the experiments of Ref. [120]. Panels (a)–(c) [(d)–(f)] display the OP [IP] polarized modes, respectively. In the figure, the results correspond to (a), (d) the square, (b), (e) the honeycomb and (c), (f) the Lieb lattices, respectively. One immediately notice that the increased linewidth induced by Ohmic losses results in a global resolution of the spectral function which is significantly lower than the one shown in Fig. 9. As a consequence, several plasmonic bands cannot be distinguished properly as it is the case, e.g., for the OP modes in the honeycomb and Lieb lattices in the vicinity of the corners of their respective 1BZ. Nevertheless, within the regime of parameters used in Fig. 10, the linewidth of a majority of the plasmonic modes is still sufficiently small to allow for an experimental detection (except for the bright modes within the light cone). Embedding the metasurface in a gain media material [8, 44, 121–123] should help diminishing the effects induced by Ohmic losses on the spectral function and further improve the experimental observability of the collective plasmons.

**VIII. CONCLUSION**

We have considered the plasmonic properties of metasurfaces constituted by an ordered two-dimensional arbitrary array of spherical metallic nanoparticles. We have focused on the case where the interparticle distance is much smaller than the wavelength associated with the dipolar localized surface plasmon resonance frequency of single nanoparticles, where the near-field, quasistatic dipole–dipole interaction dominates. We have developed a comprehensive open quantum system framework to analyze in full analytical detail the dispersion relations and the lifetimes of the resulting collective plasmonic modes supported by the various metasurfaces which we have studied, including, e.g., the honeycomb, Lieb, and kagome lattices. Such metasurfaces present appealing topological features in their band structures, such as massless Dirac-like conical dispersion, as well as nearly flat bands, and may be a possible experimental platform to explore new states of hybrid light–matter waves.

Our model enabled us to unveil analytical expressions for the fully-retarded dispersion relations of the plasmonic collective modes, including also the effects of the particle-hole environment to which such modes are coupled to, and which are of particular relevance for nanoparticles of only a few nanometers in size. Our theory further allowed us to provide analytical expressions for the radiative and nonradiative (Landau) damping rates of the plasmonic modes, which enabled us to critically examine their experimental observability. While Ohmic losses, inherent to the metallic nature of plasmonic particles, may make the detection of the collective modes elusive, the use of gain materials should give scope to their experimental observation.
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Appendix A: Perturbative treatment of the plasmonic Hamiltonian (16)

In this appendix, we present an approximate treatment of the plasmonic Hamiltonian (16), where the nonresonant terms are treated perturbatively, while the resonant ones are diagonalized exactly. Such an approach is justified by the fact that the coupling constant \( \Omega \ll \omega_0 \) in Eq. (16). We thus split the Hamiltonian (16) into two parts,

\[
H_{\text{pl}} = H_{\text{pl}}^{(0)} + H_{\text{pl}}^{(1)},
\]

(A1)

where the rotating-wave, resonant Hamiltonian \( H_{\text{pl}}^{(0)} \) reads as

\[
H_{\text{pl}}^{(0)} = \hbar \omega_0 \sum_q \sum_s \sum_\sigma b_\sigma^\dagger(q) b_\sigma^\text{(0)}(q)
+ \frac{\hbar \Omega}{2} \sum_q \sum_{ss'} \sum_{\sigma\sigma'} \left[ f_{ss'}^{\sigma\sigma'}(q) b_\sigma^\dagger(q) b_{\sigma'}^\dagger(q) + \text{h.c.} \right],
\]

(A2)

while the nonresonant part is

\[
H_{\text{pl}}^{(1)} = \frac{\hbar \Omega}{2} \sum_q \sum_{ss'} \sum_{\sigma\sigma'} \left[ f_{ss'}^{\sigma\sigma'}(q) b_\sigma^\dagger(q) b_{\sigma'}^\dagger(-q) + \text{h.c.} \right].
\]

(A3)

In order to treat the nonresonant Hamiltonian (A3) perturbatively, we decompose the eigenfrequencies \( \omega_\tau^\varepsilon(q) \) and the Bogoliubov operators \( \beta_\tau^\varepsilon(q) \) entering the diagonal form (20) into

\[
\omega_\tau^\varepsilon(q) = \omega_\tau^{(0)}(q) + \omega_\tau^{(1)}(q)
\]

(A4)

and

\[
\beta_\tau^\varepsilon(q) = \beta_\tau^{(0)}(q) + \beta_\tau^{(1)}(q).
\]

(A5)

Here, the frequencies \( \omega_\tau^{(0)}(q) \) and the operators

\[
\beta_\tau^{(0)}(q) = \sum_{ss} \tilde{\nu}_{\tau ss}^\varepsilon(q) b_\sigma^\text{(0)}(q)
\]

(A6)
diagonalize the resonant part of the Hamiltonian (A1), i.e.,

\[
H_{\text{pl}}^{(0)} = \sum_q \sum_{\tau\varepsilon} \hbar \omega_\tau^{(0)}(q) \beta_\tau^{(0)\dagger}(q) \beta_\tau^{(0)}(q),
\]

with the normalization condition

\[
\sum_{ss} \tilde{\nu}_{\tau ss}^\varepsilon(q) \tilde{\nu}_{\tau ss'}^\varepsilon(q) = \delta_{\tau\tau'} \delta_{\varepsilon\varepsilon'}.
\]

(A8)

Treating the terms \( \omega_\tau^{(1)}(q) \) and

\[
\beta_\tau^{(1)}(q) = \sum_{ss} \tilde{\nu}_{\tau ss}^{\varepsilon'}(q) b_\sigma^\dagger(-q)
\]

(A9)

perturbatively in the Heisenberg equation (23), and solving order by order gives the equations of motion

\[
\begin{align*}
[H_{\text{pl}}^{(0)}, H_{\text{pl}}^{(0)}] &= \hbar \omega_\tau^{(0)}(q) \beta_\tau^{(0)}(q),
\text{and}

[H_{\text{pl}}^{(1)}, H_{\text{pl}}^{(1)}] &= \hbar \omega_\tau^{(1)}(q) \beta_\tau^{(0)}(q) \\
&+ \hbar \omega_\tau^{(0)}(q) \beta_\tau^{(1)}(q).
\end{align*}
\]

(A10)

Equation (A10) then yields

\[
\left[ \omega_0 - \omega_\tau^{(0)}(q) \right] \tilde{\nu}_{\tau ss}^{\varepsilon}(q) + \Omega \sum_{s's''} \tilde{\nu}_{\tau ss'}^{\varepsilon}(q) f_{s's''}^{\varepsilon}(q) = 0.
\]

(A12)

The latter expression represents a \( 3S \times 3S \) block-diagonal system which, once solved, gives access to the unperturbed eigenfrequencies \( \omega_\tau^{(0)}(q) \) and Bogoliubov coefficients \( \tilde{\nu}_{\tau ss}^{\varepsilon}(q) \). Equation (A11) then dictates that the first-order correction to the plasmonic eigenfrequencies vanishes, i.e., \( \omega_\tau^{(1)}(q) = 0 \), while the coefficients \( \tilde{\nu}_{\tau ss}^{\varepsilon}(q) \) are obtained by solving the \( 3S \times 3S \) system of equations

\[
\begin{align*}
- \left[ \omega_0 + \omega_\tau^{(0)}(q) \right] \tilde{\nu}_{\tau ss}^{\varepsilon}(q) + \Omega \sum_{s's''} \left[ \tilde{\nu}_{\tau ss'}^{\varepsilon}(q) - \tilde{\nu}_{\tau ss'}^{\varepsilon}(q) \right] f_{s's''}^{\varepsilon}(q) &= 0.
\end{align*}
\]

(A13)

Appendix B: Plasmonic band structures of the rectangular and hexagonal lattices

In order to showcase the versatility of our Hamiltonian approach, we present in this appendix results for the plasmonic band structure of two additional Bravais lattices, the rectangular and hexagonal ones shown in Figs. 11(a) and 11(c), respectively. The corresponding 1BZs are displayed in Figs. 11(b) and 11(d). The solid lines in panels (e) and (g) of Fig. 11 represent the quasistatic plasmonic band structure of two additional Bravais lattices, played in Figs. 11(b) and 11(d). The solid lines in panels (e) and (g) of Fig. 11 represent the quasistatic plasmonic band structure of two additional Bravais lattices, respectively. We show by dashed lines the corresponding band structure considering nearest-neighbor couplings only. As can be seen from Figs. 11(e) and 11(g), these band structures are qualitatively similar to the case of the square lattice shown in Fig. 2(c). In Figs. 11(f) and 11(h), we show by solid lines the band structure from Eq. (27) of the IP.
FIG. 11. (a),(c) Sketch of (a) a rectangular lattice with primitive lattice vectors $t_1 = d (1, 0)$ and $t_2 = d (0, \frac{1}{2})$ and (c) a hexagonal one with $t_1 = d (1, 0)$ and $t_2 = d (\frac{1}{2}, \frac{\sqrt{3}}{2})$. (b),(d) Corresponding first Brillouin zones, with (b) primitive reciprocal vectors $b_1 = \frac{2\pi}{d} (1, 0)$ and $b_2 = \frac{4\pi}{\sqrt{3}d} (0, 1)$ and (d) $b_1 = \frac{2\pi}{\sqrt{3}d} (\sqrt{3}, -1)$ and $b_2 = \frac{4\pi}{\sqrt{3}d} (0, 1)$. (e)–(h) Quasistatic plasmonic band structure of (e),(f) the rectangular and (g),(h) the hexagonal lattices for (e),(g) out-of-plane (OP) and (f),(h) in-plane (IP) polarizations. The dashed and solid lines correspond, respectively, to nearest-neighbor and long-range couplings, and the color code represents the polarization angle (25). Same parameters as in Fig. 2 presented in the main text.

For completeness, we display in Fig. 12 the fully-retarded dispersion relation including the radiative shift for both the rectangular and hexagonal lattices, as well as for both the OP and IP modes. When comparing the results of Figs. 11 and 12, similar conclusions as in the main text can be drawn: when retardation effects are taken into account, transverse modes acquire a polaritonic singularity at the crossing of the quasistatic band structure and the light cone, while longitudinal modes do not present such a singularity. The results of Fig. 12 confirm that retardation effects are essential in polarized modes for the rectangular [panel (f)] and hexagonal [panel (h)] lattices (the dashed lines correspond to considering the nearest neighbors alone). There, qualitative differences arise in the dispersion relations (as well as in the polarization angles, cf. color coding) as compared to the one of the square lattice [Fig. 2(d)], due to the different symmetries of the corresponding lattices.
accurately describing the plasmonic dispersion relations in two-dimensional lattices of near-field coupled nanoparticles.

Appendix C: The kagome lattice

As another example of a tripartite lattice known to exhibit in electronic tight-binding models a flat band and conical dispersion, we here consider the kagome lattice sketched in Fig. 13(a). Figure 13(b) shows the corresponding 1BZ. The plasmonic band structure is plotted for $\sigma = z$ in Fig. 13(c) as solid lines [cf. Eq. (45)]. We also show by dashed lines the dispersion relations within the nearest-neighbor approximation, for which the lattice sums simplify to $f_{nn,12}^{zz}(q) = 2\cos(q_xd)$, $f_{nn,13}^{zz}(q) = 2\cos((q_x + \sqrt{3}q_y)d/2)$, and $f_{nn,23}^{zz}(q) = 2\cos((q_x - \sqrt{3}q_y)d/2)$, yielding a flat band at the frequency $\omega_{nn,\tau}^{zz}(q) = \omega_0(1 - 4\Omega/\omega_0)^{1/2}$, and two dispersive bands $\omega_{nn,\tau}^{zz}(q) = \omega_0\{1 + 2(\Omega/\omega_0)[1 \pm \sqrt{1 + \Pi^{zz}(q)}]\}^{1/2}$, where we used $\Sigma^{zz}(q) - \Pi^{zz}(q) = 4$ [cf. Eqs. (47) and (48)]. As can be seen from Fig. 13(c), the dispersive bands above correspond to a conical dispersion in the vicinity of the $K$ point of the 1BZ. In particular, for wavevectors $k$ close to $K = \frac{2\pi}{3d}(1,0)$, we have $\omega_{nn,\tau}^{zz}(k) \simeq \omega_0 + \Omega \pm v_{nn}^z|k|$, with $v_{nn}^z = \sqrt{3}\Omega d$. Comparing the solid and dashed lines in Fig. 13(c), we see that the conical dispersion is robust against long-range interactions, while the flat band gets slightly dispersive, although much less than for the case of the Lieb lattice (compare with Fig. 4). This can be understood by expanding the band structure (45) for $|k| \ll |K|$, with $q = K + k$, yielding $\omega_{\tau}^{z}(k) \simeq \omega_0 - \Omega(2 - |f_{11}^{zz}(K)|)$ and $\omega_{\tau}^{z}(k) \simeq \omega_0 + \Omega(1 - 2|f_{11}^{zz}(K)|) \pm v^z|k|$, with $v^z = 1.49\Omega d$. Hence, the long-range dipolar interactions here also renormalize the group velocity as compared to the nearest-neighbor approximation, and slightly shift down in energy the band degeneracy point.

The band structure obtained by solving numerically the system of equations (24) for $\sigma = x,y$ is plotted in Fig. 13(d) as solid lines. We also plot the numerical results considering only nearest-neighbor couplings (dashed lines) for comparison. As can be seen from the figure, despite the fact that the long-range interactions significantly modify the quasistatic plasmonic band structure as compared to the nearest-neighbor result, the conical dispersion in the vicinity of the $K$ and $K'$ points of the 1BZ remain qualitatively unaffected. Taking into account the retardation effects does not
FIG. 13. (a) Sketch of a kagome lattice with primitive lattice vectors \( t_1 = d (2, 0) \) and \( t_2 = d (1, \sqrt{3}) \), and basis vectors \( d_2 = t_1 / 2 \) and \( d_3 = t_2 / 2 \). (b) Corresponding first Brillouin zone, with primitive reciprocal vectors \( b_1 = \pi \sqrt{3} d (\sqrt{3}, -1) \) and \( b_2 = 2 \pi \sqrt{3} (0, 1) \). (c),(d) Quasistatic plasmonic band structure for (c) out-of-plane (OP) and (d) in-plane (IP) polarizations. The dashed and solid lines correspond, respectively, to nearest-neighbor and long-range couplings. Color code of the solid lines: polarization angle (25). Same parameters as in Fig. 2.

modify such a conclusion, as can be seen from Fig. 14.

Appendix D: Dipolar matrix elements in a spherical hard-wall potential

In this appendix, we provide explicit expressions of the matrix elements \( \langle e | \sigma | h \rangle (\sigma = x, y, z) \) that we use in Sec. VI for the evaluation of the effects induced by the electronic Hamiltonian (12) onto the plasmonic subsystem. Assuming that the self-consistent potential experienced by electron (e) and hole (h) states within each nanoparticle is given by a spherical hard-wall potential, we have [36, 115]

\[
\langle e | r | h \rangle = R(\epsilon_e, \epsilon_h) \left( \sum_{\mathcal{N}=\pm 1} A_{l_e, l_h, \mathcal{N}}^{m_e, m_h} \frac{x - iN\hat{y}}{\sqrt{2}} + A_{l_e, l_h, 0}^{m_e, m_h} \hat{z} \right). 
\]

(D1)

In the equation above, the radial part of the dipolar matrix element is given by [114]

\[
R(\epsilon_e, \epsilon_h) = \frac{2\hbar^2}{m_e a} \sqrt{\epsilon_e \epsilon_h} (\epsilon_e - \epsilon_h)^2, 
\]

(D2)

while the angular part is expressed in terms of Wigner-3\( j \) symbols as

\[
A_{l_e, l_h, \mathcal{N}}^{m_e, m_h} = (-1)^{m_e+N} \sqrt{(2l_e+1)(2l_h+1)} \begin{pmatrix} l_e & l_h & 1 \\ 0 & 0 & 0 \end{pmatrix} \times \begin{pmatrix} l_e & l_h & 1 \\ -m_e & m_h & \mathcal{N} \end{pmatrix}. 
\]

(D3)

The latter expression encapsulates the angular momentum selection rules \( l_e = l_h \pm 1 \) and \( m_e = m_h \) (for \( \mathcal{N} = 0 \)) and \( m_e = m_h \pm 1 \) (for \( \mathcal{N} = \pm 1 \)).

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FIG. 14. Colored lines: plasmonic band structure $\omega\epsilon\tau(q) + \delta\epsilon\tau(q)$ including retardation effects for the kagome lattice and for (a) OP and (b) IP modes. Thin grey lines: quasistatic dispersion relations $\omega\epsilon\tau(q)$. Parameters of the figure: $d = 3a$ and $k_0a = 0.15$.

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