Semiconductor quantum plasmonics

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Abstract

We investigate the frontier between classical and quantum plasmonics in highly doped semiconductor layers. The choice of a semiconductor platform instead of metals for our study permits an accurate description of the quantum nature of the electrons constituting the plasmonic response, which is a crucial requirement for quantum plasmonics. In particular we show that our quantum model permits to calculate the collective plasmonic resonances from the electronic states determined by an arbitrary one-dimensional potential. Furthermore, it allows us to analytically demonstrate the Lindhard formula in the limit of a vanishing confinement. Our approach is corroborated with experimental spectra in which higher order longitudinal plasmonic modes are present. This work opens the way towards the applicability of quantum engineering techniques for semiconductor plasmonics.
The properties of plasmons in nanostructures can be profoundly modified whenever their elementary constituents, electrons and photons, enter the quantum regime [1–5]. One of the pioneering works in quantum plasmonics is the demonstration that the optical resonances of localized surface plasmons in metallic nanoparticles can be strongly affected by electronic confinement for nanoparticle sizes of the order of 10 nm [6]. In this case, the Drude model fails in describing the optical response of the electron gas, and a quantum treatment taking into account size confinement must be considered. Recently, the ultimate confinement of plasmons has been provided by van der Waals heterostructures in a one-atom-thick layer [7]. Another fundamental quantum effect, tunneling, has been shown to strongly impact the properties of the plasmons beyond the classical treatment [8, 9] and it can be controlled by using molecular junctions [10].

In this work we investigate the frontier between classical (i.e. the electron response is described by Drude model) and quantum plasmonics by studying volume plasmons, which are collective excitations of a three dimensional electron gas. A sketch of the system is presented in fig. 1(a). It is composed of a highly doped semiconductor layer, with thickness smaller than the plasma wavelength in the material. The oscillation of the free electrons in the doped layer under an incident electromagnetic field results in an optically active collective mode of the system, a plasmon confined in the doped layer with a dipole moment along the growth direction $z$. The absorption spectrum, also sketched in fig. 1(a), presents a single Lorentzian resonance, centered at the plasma energy, with a quality factor in the order of 10-20 [11]. This resonance is called a Ferrel – Berreman or Berreman mode [11–15]. This sharp feature can be simulated by using the Drude model to describe the isotropic permittivity of the doped semiconductor and by considering the finite thickness of the layer. This system is thus an example of classical plasmonics. Berreman modes have first been observed in thin metallic films, and their observation was considered as experimental evidence of the existence of the plasmon in the early days of plasmonics [12, 13, 16, 17]. More recently, Berreman modes have raised considerable attention because at the plasma frequency the real part of the dielectric permittivity is zero. For this reason, this resonance is also referred to as epsilon-near-zero (ENZ) mode [18, 19]. Hyperbolic metamaterials obtained by alternating ENZ and dielectric layers have also been demonstrated [20].

In the classical description, the plasma frequency is independent from the plasmon wavevector $\vec{k}$, as sketched in panel (b). However, this is a strong approximation, as it
has been experimentally demonstrated through optical excitation of plasmons in metal foils \[16, 17, 21\]. These experiments have shown that plasmon longitudinal dispersion is described by the Lindhard formula:

\[ \Omega_{k_z}^2 = \Omega_p^2 + \frac{3}{5} v_F^2 k_z^2 \]  

where \( \Omega_p \) is the bulk plasma frequency, \( v_F \) the Fermi velocity and \( k_z \) the \( z \) component of the plasmon wavevector. This dispersion relation, sketched in panel (c) of fig. 1, requires a quantum treatment of electrons in metals and a semiclassical treatment of light-matter interaction \[22\].

Quantum effects also appear in the properties of plasmons if the doped semiconductor layer has a thickness smaller than the de Broglie wavelength of electrons, typically in the order of few tens of nanometers in semiconductors. In this case size confinement gives rise to quantized energy levels for the electron movement along the growth direction. Panel (d) of fig. 1 sketches a doped semiconductor quantum well, with three occupied confined states. In this case there are three main optically active transitions, represented as three different sets of harmonic oscillators along the growth direction. Dipole - dipole interaction between these optically active transitions gives rise to a collective mode of the system, a confined plasmon, at an energy which is higher than the plasma energy of the electron gas \[11, 23\], and higher than the energy of the individual electronic transitions. The Drude model thus fails to describe the collective optical properties of confined electrons.

The quantum nature of the electrons building the plasmons can be more easily taken into account in a semiconductor platform than in a metal nanoparticle. We exploit envelope function approximation to account for size confinement and tunneling, with the aim of extending quantum engineering to the realm of plasmonics. This work is organized as follows. We first present our theoretical model, which derives the plasmonic resonances from the basis of the electronic states in a semiconductor quantum well \[19, 23–25\]. Our approach is validated by demonstrating that the plasmon longitudinal dispersion follows the Lindhard formula in the limit where the energy contribution resulting from confinement vanishes. We present the first experimental evidence of the existence of higher order longitudinal plasmon modes in highly doped semiconductors. These collective excitations are known as Tonks-Dattner modes \[17, 26, 27\] and follow the Lindhard formula. The effect of size confinement and tunneling is then discussed, demonstrating that our theoretical model can be applied
to semiconductor potentials with complex shapes and can be used to engineer the energies and oscillator strengths of plasmons.

FIG. 1. Frontier between classical and quantum plasmonics. (a) Sketch of a doped semiconductor layer embedded between two undoped layers. If the thickness $L$ is smaller than the plasma wavelength in the material, $\lambda_P = \frac{2\pi c}{\Omega_P}$, it is possible to optically excite a collective mode of the system, a confined plasmon, displaying a Lorentzian resonance centered at the plasma energy. (b) Dispersion of a volume plasmon of energy $E_P = \hbar \Omega_P$ in a classical model. (c) Longitudinal dispersion of a volume plasmon taking into account the material bandstructure and the Fermi distribution of electrons. (d) Effect of size confinement in the optical response of a semiconductor quantum well with three occupied electronic states. Due to dipole – dipole coupling the optical response of the system is a collective resonance, centered at a greater energy than the plasma one, due to the size confinement contribution.

In our formalism, the optical properties of the confined plasmons are described in the Coulomb gauge through the dipole representation of the light-matter interaction [24, 25]. By introducing the excitation between confined electronic states, the Hamiltonian of the
electron gas in the quantum well can be written as:

\[ H = \sum \alpha \hbar \omega_\alpha b_\alpha^\dagger b_\alpha + \frac{e^2}{2\epsilon_0 \epsilon_s} \sum_{\alpha,\beta} S_{\alpha,\beta} \sqrt{\Delta N_\alpha \Delta N_\beta} (b_\alpha^\dagger + b_\alpha)(b_\beta^\dagger + b_\beta) \]  

(2)

In this expression, the indices \( \alpha, \beta \) run over all transitions between confined states in the quantum well; the operators \( b_\alpha^\dagger, b_\alpha \) are the creation and annihilation operators of the transition \( \alpha \) of energy \( \hbar \omega_\alpha \) and \( \Delta N_\alpha \) the associated population difference; \( \epsilon_s \) is the background dielectric constant. The coupling coefficients \( S_{\alpha,\beta} \) are expressed as:

\[ S_{\alpha,\beta} = \frac{1}{\hbar \omega_\alpha} \frac{1}{\hbar \omega_\beta} \left( \frac{\hbar^2}{2m^*} \right)^2 \int_{-\infty}^{+\infty} dz \xi_\alpha(z) \xi_\beta(z) \]  

(3)

with \( m^* \) the electron effective mass. The coupling coefficients between the electronic transitions \( S_{\alpha,\beta} \) are proportional to the overlap between the a.c. microcurrent functions \( \xi_\alpha \) describing the electronic transitions \( \alpha = i \rightarrow i + j \) and defined as [25]:

\[ \xi_\alpha(z) \equiv \xi_{i\rightarrow i+j}(z) = \psi_i(z) \frac{\partial \psi_{i+j}(z)}{\partial z} - \psi_{i+j}(z) \frac{\partial \psi_i(z)}{\partial z}, \]  

(4)

where \( \psi_i \) is the envelope function of the confined electronic state of quantum number \( i \). The integral of each microcurrent function is proportional to the optical dipole of the corresponding transition [24, 25]. Note that in a square quantum well the transitions with a non-zero dipole are only those involving wavefunctions of different parity (i.e. with odd \( j \)).

In the limit of infinite barriers, for a quantum well with thickness \( L \), the microcurrent associated with the transition \( i \rightarrow i + j \) reduces to:

\[ \xi_{i\rightarrow i+j}(z) = \frac{\pi}{L^2} \left[ j \sin \left( \frac{2i + j}{L} \pi z \right) - (2i + j) \sin \left( \frac{j \pi z}{L} \right) \right] \]  

(5)

Examples of such functions for optically active transitions are plotted in fig. 2 (a) and (b). Panel (a) presents microcurrents describing transitions between consecutive confined states (i.e. corresponding to \( j = 1 \)) and for different values of \( i \): \( i = 2 \) (black line), \( i = 6 \) (red line), \( i = 12 \) (green line). For \( i \gg 1 \) the first sinus term in the square bracket in eq. 5 is negligible with respect to the second term and the microcurrent describing consecutive transitions is of the form \( \sin \left( \frac{2i}{L} \pi z \right) \) (blue line). Figure 2(b) shows the same microcurrents calculated for \( j = 3 \). From both panels one can see that, for a fixed \( j \), when increasing the value of the index \( i \), the shape of the functions representing the microcurrents approaches
FIG. 2. From single particle transitions to confined plasmon modes. (a), (b) Schematic representation of an infinite barrier quantum well of thickness $L$ with the calculated normalized microcurrents $\xi_{i\rightarrow i+j}(z)$ for $j = 1$ (a) and $j = 3$ (b) and $i = 2$ (black), $i = 6$ (red), $i = 12$ (blue). (c), (d) Charge density distribution (upper panels) and microcurrents (lower panels) calculated for confined plasmons with $j = 1$ (c) and $j = 3$ (d) in an infinite quantum well. The plasmon mode with index $j$ is issued from the ensemble of the electronic transitions $i \rightarrow i + j$.

As a consequence, in the limit of high values of $i$, i.e. when a large number of subbands are occupied, the microcurrents $\{\xi_{i\rightarrow i+j}\}_{i,j}$ are mutually orthogonal for different $j$. In this approximation the coupling coefficients between microcurrents are given by:

$$S_{i\rightarrow i+j,i\rightarrow i+j'} = \frac{L}{2\pi^2j^2j'^2}\delta_{j,j'}.$$  

The matrix of the coupling coefficients is block–diagonal: all the electronic transitions with the same value of $j$ contribute to the same plasmon mode. In other words, the Hamiltonian (2) can be independently diagonalized on the subspaces relative to transitions $i \rightarrow i + j$ for fixed $j$.

For each subspace of index $j$, light couples primarily with the plasmon corresponding to the highest frequency eigenmode. Its frequency $\Omega_j$ can be found, after Bogoliubov trans-
formation within the associated subspace, by calculating the zeros of the following determinant \[24, 25, 28\]:

\[
\Delta_j(\omega) = 1 - \frac{2e^2}{\hbar \epsilon_0 \epsilon_s} \sum_i S_{i \to i + j, i \to i + j} \frac{\Delta N_{i \to i + j} \omega_{i \to i + j}}{\omega^2 - \omega_{i \to i + j}^2}. \tag{6}
\]

Figures 2(c) and (d) present the microcurrents (lower panels) associated with the confined plasmon modes issued from the subspaces of index \(j = 1, 3\). These microcurrents have the same symmetry as the electronic transitions \(i \to i + j\), and are thus proportional to \(\sin \left( \frac{j\pi z}{L} \right) \).

The oscillator strength of the plasmon mode \(\Omega_j\) is proportional to the squared integral of the plasmon microcurrent \[24\]. Each plasmon mode \(\Omega_j\) concentrates the oscillator strength of the ensemble of the \(i \to i + j\) transitions \[23\]. Only plasmons with odd values of \(j\) will have a non-zero oscillator strength.

The top panels of fig. 2(c) and (d) present the charge distributions oscillating at frequency \(\Omega_j\), which are proportional to the derivative of the microcurrents \[24\]. We can clearly see that the fundamental mode \(j = 1\) corresponds to a dipole along the growth direction of the quantum well. This mode is thus the analogue of a Berreman mode in a metallic thin film. The modes with \(j > 1\) are higher order longitudinal confined plasmon modes, with smaller dipoles. They correspond to the so-called Tonks-Dattner modes that have been observed in rare gas \[26\], ultra-cold \[27\] plasmas and metallic thin films \[29\].

From eq. 6, we demonstrate that, up to the second order in \(j v_F/(L \Omega_p)\), we can derive the Lindhard formula describing the longitudinal dispersion of bulk plasmons in metals \[16, 17\]:

\[
\Omega_j^2 = \Omega_p^2 + \frac{3}{5} v_F^2 \left( \frac{\pi j}{L} \right)^2, \quad \Omega_p^2 = \frac{e^2 N_v}{m^* \epsilon_0 \epsilon_s} \tag{7}
\]

with \(j\) an odd integer number, \(\Omega_p\) the bulk plasma frequency, \(v_F\) the Fermi velocity, \(N_v\) the electronic density per unit volume of the doped semiconductor layer. It is important to note that the Lindhard formula has been demonstrated in our case by starting from the electronic wavefunctions in a square quantum well in the limit of thick layers (when the number of occupied states is large, \(i \gg 1\)).

In order to verify the validity of the approximations used above to derive the Lindhard formula, we report in fig. 8 the calculated absorptivity (colorscale) as a function of the thickness for InGaAs/AlInAs QWs ranging from 10 nm to 200 nm and for two different values of the electronic density per unit volume. The absorption is computed \[30\] by numerically diagonalizing eq. 2 by using envelope functions calculated within a \(\vec{k} \cdot \vec{p}\) model \[31\].
Nonparabolicity has been neglected in the numerical calculation of the electronic transition energies and microcurrents in order to obtain a quantitative comparison with equation (7). The energies of the first longitudinal plasmon modes obtained from equation (7) are indicated in the plot as dashed white lines. Lindhard dispersion provides a very good estimate of the plasmonic resonances in GaInAs layers. However equation (7) tends to underestimate the energy of large $j$ longitudinal modes, due to the influence of the finite barrier height and of the finite number of electronic levels. In the limit of narrow wells, in which quantum confinement plays a very significant role, remarkably equation (7) still provides a satisfactory estimation of the energy of the absorption maxima. However, additional lines appear, corresponding to individual intersubband transitions (particularly on fig. 3a, for lower electron density). The highest amplitude of these modes occurs when they are resonant with the Lindhard frequencies. From this figure we can also see that while the energy rise of the plasmon in thin quantum wells can be attributed to electron size confinement [11], the evolution of the oscillator strength is better understood from the viewpoint of the confinement of the collective electronic oscillation.

In the following we experimentally verify the Lindhard dispersion relation in a 100 nm highly doped GaInAs layer. Before presenting our experimental results, it is important to underline that the observation of Tonks Dattner modes through optical measurements in this system is not straightforward. Indeed, applying the sum rule derived in ref. [24], it is possible to show that, within the previous approximations, the effective oscillator strength [24] of the plasmon of frequency $\Omega_j$ is $\frac{4\epsilon_0 N_e}{m^* \pi^2 j^2}$. This expression clearly indicates that the efficiency of the coupling of the longitudinal plasmons with free space radiation decreases very rapidly with the index $j$. This is also apparent from the colormaps shown in fig. 3.

In order to observe higher order modes, we have exploited the fact that the main plasmon, due to its superradiant nature [32], radiatively broadens by increasing the internal angle of light propagation in the semiconductor [30]. This broadening is associated with a decrease of its amplitude. On the contrary, higher order modes, which are not radiatively broadened, increase their absorption amplitude at high angles. For this reason at high angle, the higher order collective modes become visible up to $j = 9$. In order to reach high angles, without dealing with saturated absorption for the main plasmon peak, we have performed angle resolved emission experiments under thermal excitation of the plasmon through the application of an in-plane current, as in ref. [33]. The experiment has been performed on a
FIG. 3. Colormap of the absorptivity simulated at low incidence angle for an InGaAs/AlInAs QW with electronic density (a) $N_v = 4 \times 10^{18} \text{ cm}^{-3}$ and (b) $N_v = 8 \times 10^{18} \text{ cm}^{-3}$. The absorptivity is computed from the full numerical diagonalization procedure and plotted using a non-linear colorscale to make the low oscillator strength plasmons more clearly visible. For each value of $L$, the maximum absorptivity has been normalized to 1. The dashed white lines indicate the energy of the confined longitudinal plasmons as obtained from Lindhard dispersion (7).

100 nm GaInAs/AlInAs layer with a doping density of $7.5 \times 10^{18} \text{ cm}^{-3}$. Figures 4(a)-(d) show normalized thermal emission spectra (in red) at different angles, measured at room temperature and extracted from the device through a polished facet. The detailed description of the device and of the experiment can be found in ref. 33.

Several peaks are clearly observed in the spectra. When increasing the angle, the main plasmon resonance, corresponding to $j = 1$, becomes broader and weaker due to the increase of the radiative decay rate of the plasmon and, simultaneously, the higher energy resonances, associated with bright plasmons with $j > 1$, become more and more visible. These two effects are very well reproduced by the calculated spectra, shown in black. These spectra have been obtained by solving quantum Langevin equations in the input-output formalism. The system consists of the bright plasmon modes coupled with the electromagnetic vacuum and with a bath of electronic oscillators describing the non-radiative decay of the collective modes [30].
FIG. 4. (a), (b), (c), (d) Measured (red lines) and simulated (black lines) emission spectra under thermal excitation of the plasmons for different values of the internal angle of light propagation. (e) Squared energy of the resonances (red dots) of the thermal emission spectra plotted as a function of the quantum number $j$ of the corresponding confined plasmon (blue line).

The plasmon eigenmodes have been calculated from a full numerical diagonalization accounting for the finite barrier height as well as for nonparabolicity effects. The computed microscopic current densities corresponding to the four brightest plasmon modes (not shown here) are very well approximated by $\sin\left(\frac{j\pi z}{L}\right)$ for $j = 1, 3, 5, 7$ and are very close to those sketched in fig. 2.

Figure 4(e) presents the squared plasmon energies (red symbols) extracted from the emission spectra measured at angles between $23^\circ$ and $83^\circ$ (15 spectra) plotted as a function of the index $j$ of the plasmon. The blue line shows a fit of the experimental dispersion following eq. 7. From the fit, we extract the Fermi velocity of the electrons $v_F = 1 \times 10^8$ cm/s and the plasma energy $\hbar\Omega_P = 114$ meV, in very good agreement with the calculated plasma energy, which is equal to 111 meV. Note that non-parabolicity is very important in the system. The effective mass that can be extracted from the plasma energy, by knowing the electronic density, is $m^* = 0.066 m_0$ (with $m_0$ the free electron mass), which is greater than the effective mass value in InGaAs at the bottom of the conduction band, 0.042$m_0$. The Fermi energy, extracted from the Fermi velocity by using $m^* = 0.066 m_0$, is $E_F = 216$ meV, in agreement with the Fermi energy calculated at 300K including the non-parabolic dispersion of the
FIG. 5. Upper panel: Calculated absorptivity for a square GaInAs highly doped semiconductor layer, with thickness 54 nm and electronic density $N_v = 2 \times 10^{19} \text{ cm}^{-3}$. Lower panel: Calculated absorptivity for a system of asymmetric tunnel coupled GaInAs/AlInAs quantum wells, with the same electronic density as the 54 nm structure. The conduction band profile and the square moduli of the wavefunctions for the two structures are presented as insets. The black horizontal lines indicate the Fermi energy.

Having confirmed our microscopic approach through the comparison between experimental and theoretical results, we discuss in the last part of this paper how electronic confinement and tunnel coupling can be used as a degree of freedom to engineer the plasmonic resonances. The starting point is a GaInAs layer of 54 nm thickness, sandwiched between two AlInAs barriers. The electronic density per unit volume in the GaInAs layer is $N_v = 2 \times 10^{19} \text{ cm}^{-3}$. Figure 5 presents the absorptivity spectrum simulated at 45° (black line), showing the Tonks - Dattner resonances as previously discussed. The inset of the figure (black panel) shows the corresponding band diagram, where we also plotted the square moduli of the electronic wavefunctions and the position of the Fermi energy (black horizontal line). We now insert in
the GaInAs layer 6 identical AlInAs barriers, of 1.5 nm thickness, such that the structure is now composed of tunnel coupled asymmetric quantum wells (of 7.5 nm and 5 nm thickness) as shown in the red panel of fig. The electronic structure is profoundly modified by the presence of the barriers, resulting in the formation of several minibands. In particular, as the tunnel coupled quantum wells have different sizes, we can now have optically active transitions between the states of the ground miniband and those of the second excited one (i.e. \( j = 2 \), which are forbidden in a single quantum well). The electronic density per unit volume is kept equal to that of the single GaInAs layer, \( N_v = 2 \times 10^{19} \text{ cm}^{-3} \). The absorptivity spectrum including the collective effects is presented in red in the main panel of the figure. This spectrum is completely different with respect to that obtained in the single layer case proving that also collective excitations can be engineered by a judicious size confinement of the single electronic states. Not only can many collective resonances be observed, but their oscillator strengths are distributed differently between them. In particular, the lowest energy collective mode is no longer the mode with the highest absorptivity.

This work clarifies the link between the single electron wavefunctions and their collective response to the light. In that sense it opens new degrees of freedom for engineering ad hoc plasmonic resonances in which the energy position and oscillator strength is determined by shaping single electron wavefunctions. Our theoretical approach is well suited for this goal, as it goes well beyond the Drude model, which is commonly used in semiconductor plasmonics. Our model thus allows quantum engineering techniques to enter the field of semiconductor plasmonics.

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