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Coupled nano-plasmons

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Abstract

A simple model of coupled plasmons arising in two neighbouring nano-particles is presented. The coupled oscillations and the corresponding eigenfrequencies are computed. It is shown that the plasmons may be periodically trasferred between the two particles. For larger separation distances between the two particles the retardation is included. The oscillation eigenmodes are called polaritons in this case. There are distances for which the particles do not couple to each other in this case, *i.e.* the polaritonic coupling gets damped. The van der Waals-London-Casimir force is estimated for the two particles; it is shown that for large distances the force is repulsive.

Plasmons is an old and fundamental concept in condensed matter physics:[1] they are long-wavelength longitudinal oscillations of the charge density in matter. In a simple model, which is usually called the Drude-Lorentz model, matter can be represented as a plasma, consisting of identical mobile charges q of mass m and concentration (density) n (*e.g.*, electrons) moving uniformly and collectively against a quasi-rigid background of neutralizing charges $-q$ (*e.g.*, ions). The practical realization of long wavelength limit implies finite-size polarizable bodies, which entail, in turn, boundary conditions. Consequently, we may have many branches of plasmons: for instance, in a homogeneous conducting sphere the plasmon spectrum is given by $\Omega_l = \omega_p \sqrt{l/(2l+1)}$, where $\omega_p = \sqrt{4\pi nq^2/m}$ is called the plasma frequency and $l = 1, 2...$ is the azymuthal quantum number. A much more convenient representation simplifies the things to point particles, which may be a reasonably useful model for the nowadays nano-plasmonics.

As it is well known, the Maxwell equations in matter imply four unknowns: $\mathbf{E}$ (electric field), $\mathbf{D}$ (electric displacement), $\mathbf{H}$ (magnetic field) and $\mathbf{B}$ (magnetic induction); and only two independent equations (Faraday and Maxwell-Ampere equations, which contain the *curl* and the time derivatives). In order to solve them, we introduce constitutive relations between these unknowns through the semi-phenomenological and quasi-empirical dielectric function ε and magnetic permeability μ. A large class of matter is quasi-non-magnetic, such that we may equal $\mathbf{H}$ and $\mathbf{B}$ and put $\mu = 1$; still, we have three unknowns ($\mathbf{E}$, $\mathbf{D}$ and $\mathbf{H}$) and two equations.

On the other hand, the motion of the mobile charges in polarizable matter can be described by a displacement field $\mathbf{u}(t, \mathbf{r})$, which is a function of the time t and position $\mathbf{r}$. In the classical limit of small and slow variations (corresponding to classical electromagnetism), this displacement field generates a polarization charge density $\rho = -nq div\mathbf{u}$ and a corresponding current density

[1] There is an enormous and ubiquituos plasmon literature, which makes a formal list of references both impossible and useless.

$\mathbf{j} = nq\dot{\mathbf{u}}$. These charge and current densities generate in matter an electric field $\mathbf{E}$ and a magnetic field $\mathbf{H}$; but we still have two independent equations and three unknowns: $\mathbf{E}$, $\mathbf{H}$ and $\mathbf{u}$. However, the displacement field obeys an equation of motion, which, in this classical limit, is the Newton equation of motion

$$m\ddot{\mathbf{u}} = q(\mathbf{E} + \mathbf{E}_0) - m\omega_c^2\mathbf{u} - m\gamma\dot{\mathbf{u}} \; ; \tag{1}$$

$\mathbf{E}$ is the internal (polarization) electric field, $\mathbf{E}_0$ is an external electric field, ω_c is a characteristic frequency and γ is a damping coefficient (much smaller than any relevant frequency). The magnetic part of the Lorentz force is absent in equation (1) because the velocities of the charges in matter are much smaller than the speed of light; the internal magnetic field is also absent, in accordance with our assumption of small $\mathbf{u}$ and non-magnetic matter. Equation (1) is the missing equation (the third equation), which helps solving the Maxwell equations.

Obviously, $\mathbf{P} = nq\mathbf{u}$ is the polarization (density of dipole moments); equation (1) leads immediately to the well-known Drude-Lorentz (plasma) dielectric function $\varepsilon(\omega) = (\omega^2 - \omega_c^2 - \omega_p^2)/(\omega^2 - \omega_c^2 + i\omega\gamma)$, where only the optical dispersion is included (through the dependence on the frequency ω). As it is well known, $\omega_c = 0$ corresponds to conductors, while $\omega_c \neq 0$ describes dielectrics. The model and equation (1) can be generalized in multiple ways. We limit ourselves here to use equation (1) in conjunction with Maxwell equations, in order to describe a simple situation regarding coupled nano-plasmons.

The longitudinal internal (polarization) electric field in Gauss equation $div\mathbf{E} = -4\pi nq div\mathbf{u}$ is given by $\mathbf{E} = -4\pi nq\mathbf{u}$ (*i.e.*, $\mathbf{E} = -4\pi\mathbf{P}$). In the long-wavelength limit, the finite size of the body is usually taken into account by a (de-) polarizing factor f, such as the field is given by $\mathbf{E} = -4\pi nqf\mathbf{u}$; for instance, for a sphere $f = 1/3$. Introducing this polarization field in equation (1), taking the Fourier transform and leaving aside the coefficient γ, we get

$$(\omega^2 - \omega_c^2 - f\omega_p^2)\mathbf{u} = -\frac{q}{m}\mathbf{E}_0 \; ; \tag{2}$$

we can see that we have a plasmon resonance at frequency $\sqrt{\omega_c^2 + f\omega_p^2}$; for a conducting sphere with $\omega_c = 0$ and $f = 1/3$, we get the plasmon frequency $\omega_p/\sqrt{3}$, in accordance with the frequencies Ω_l given above for $l = 1$.

We consider two point particles, denoted by 1 and 2, each with its own plasmon frequency $\omega_{1,2}$, separated by the position vector $\mathbf{d}$. We describe the motion of the mobile charges in each particle by a displacement vector $\mathbf{u}_{1,2}$; equation (2) becomes

$$(\omega^2 - \omega_{1,2}^2)\mathbf{u}_{1,2} = -\frac{q}{m}\mathbf{E}_{02,1} \; , \tag{3}$$

where $\mathbf{E}_{01,2}$ is the electric field generated by particle 1 (2) at the position of the particle 2 (1). In the long wavelength limit this is the field generated by a point dipole

$$\mathbf{E}_{01,2} = v_{1,2}n_{1,2}q\frac{3(\mathbf{u}_{1,2}\mathbf{d})\mathbf{d} - \mathbf{u}_{1,2}d^2}{d^5} \; , \tag{4}$$

where $v_{1,2}$ are the volumes of the two particles and $n_{1,2}$ are the concentration of the mobile charges in the particles; equation (4) is valid in the near-field region $c/\omega \gg d$, where c is the speed of light. Since the particles are considered point-like, we have also $v_{1,2}^{1/3} \ll d$. Introducing this field in equations (3) we get two coupled equations for the displacement vectors. It is convenient to use the projection of the displacement vectors on the vector $\mathbf{d}$ and on a direction perpendicular to the vector $\mathbf{d}$; we call the former the longitudinal displacements and denote them by $u_{l1,2}$, while the latter, denoted by $\mathbf{u}_{t1,2}$, are called transverse displacements. The equations for the longitudinal displacements are decoupled from those corresponding to the transverse displacements; both sets of equations have the same structure. We limit ourselves here to the longitudinal displacements

$$(\omega^2-\omega_{1,2}^2)u_{l1,2}=-\frac{\omega_{p2,1}^2 v_{2,1}}{2\pi d^3}u_{l2,1}\ . \tag{5}$$

The solution of these coupled-oscillators equations is straightforward. The factor $v_{1,2}/d^3$ plays the role of a weak-coupling constant. The eigenfrequencies of equations (5) are close to the plasmon frequencies $\omega_{1,2}$, which should satisfy the condition $c/\omega_{1,2}\gg d$; $c/\omega_{1,2}$ is usually called the plasma wavelength. For typical values $\omega_{1,2}\simeq 10^{15}s^{-1}$ we get a critical distance of the order $d\simeq 0.1\mu m$; the treament given here holds for smaller distances, while for larger distances we need to take into account the retardation in estimating the polarization field.

An interesting situation occurs for two identical conducting particles $\omega_{c1}=\omega_{c2}=0$, $\omega_{p1}=\omega_{p2}=\omega_p$ and $v_1=v_2=v$. In this case the eigenfrequencies are given by

$$\Omega_{1,2}=\omega_p\left(1\pm\frac{v}{2\pi d^3}\right)^{1/2}\simeq\omega_p\left(1\pm\frac{v}{4\pi d^3}\right)\ . \tag{6}$$

The displacement vectors for the initial condition $u_{l2}(t=0)=0$ is given by

$$\begin{aligned} u_{l1}(t) &= 2Ae^{i\omega_p t}\cos\tfrac{v}{4\pi d^3}t\ ,\\ u_{l2}(t) &= -2iAe^{i\omega_p t}\sin\tfrac{v}{4\pi d^3}t\ ; \end{aligned} \tag{7}$$

we can see that the two coupled oscillations exhibit "beats", and the plasmons can be transferred periodically between the two particles, as expected. A similar situation holds for the transverse oscillations, with the factor 2π replaced by 4π in the above formulae.

A polarizable point-like particle can be approximated by a dipole, with the current density $\mathbf{j}=vnq\dot{\mathbf{u}}\delta(\mathbf{r})$ and charge density $\rho=-vnq(\mathbf{u}grad)\delta(\mathbf{r})$, where v is the volume of the particle placed at the origin. For these charge and current distributions we can compute easily the electromagnetic potentials (Fourier transforms):

$$\mathbf{A}=-i\lambda vnq\mathbf{u}\frac{e^{i\lambda r}}{r}\ ,\ \Phi=-vnq\frac{\mathbf{ur}}{r}\frac{\partial}{\partial r}\frac{e^{i\lambda r}}{r}\ \ , \tag{8}$$

where $\lambda=\omega/c$. The polarization electric field is given by $\mathbf{E}=-(1/c)\partial\mathbf{A}/\partial t-grad\Phi$, so that we can include the retardation in the equation of motion (1). For the longitudinal oscillations of two identical conducting particles we get

$$(\omega^2-\omega_p^2)u_{l1,2}=-\frac{\omega_p^2 v}{2\pi d^3}(1-i\lambda d)e^{i\lambda d}u_{l2,1} \tag{9}$$

(and a similar set of equations for the transverse oscillations). We can see that in the non-retarded limit $\lambda d\ll 1$ equations (9) go into equations (5) derived above. We are interested now in the wave-zone limit $\lambda d\gg 1$. The eigenfrequencies of equations (9) are given by

$$(\omega^2-\omega_p^2)^2=\frac{\omega_p^4 v^2}{(2\pi d^3)^2}(1-i\lambda d)^2e^{2i\lambda d}\ \ , \tag{10}$$

or

$$\tan\lambda d=\lambda d\ ,\ (\omega^2-\omega_p^2)^2=\frac{\omega_p^4 v^2}{(2\pi d^3)^2}(1+\lambda^2 d^2)\ . \tag{11}$$

It is convenient to introduce the notations $g = v/2\pi d^3 \ll 1$ and $\omega_p d/c = A$; the solution can be found as a series of powers of g:

$$\Omega = \omega_p \left[1 \pm \frac{1}{2} g(1+A^2)^{1/2} + \frac{1}{8} g^2 (A^2 - 1) + ...\right] \; ; \tag{12}$$

it should satisfy the equation $\tan(\Omega d/c) = \Omega d/c$, which, in the limit $g \ll 1$ becomes $\tan A \simeq A$ ($\Omega \simeq \omega_p$); for large values of A we get $A = \omega_p d/c \simeq n\pi$. We can see that there are real solutions for the eigenfrequencies only for certain values of the distance $d_n \simeq n\pi c/\omega_p$, which are approximate multiples of the plasma wavelength (in this limit). The corresponding oscillations are usually called polaritons. For intermediate values of d the eigenfrequencies are complex, *i.e.* the coupling between the two particles is damped (the damping parameter γ in the equation of motion (1) should be retained in this case). We can equally well say that the two particles are not coupled in this case.

The zero-point (vacuum fluctuations) energy can be estimated as $E = \sum \hbar\Omega/2$, where the summation extends over all the eigenfrequencies. The motion of the transverse degrees of freedom leads to the eigenfrequencies equation

$$\tan \lambda d = \frac{\lambda d}{1 - \lambda^2 d^2} \; , \; (\omega^2 - \omega_p^2)^2 = \frac{\omega_p^4 v^2}{(4\pi d^3)^2}(1 + 3\lambda^2 d^2 + \lambda^4 d^4) \; . \tag{13}$$

The solution is given by

$$\Omega = \omega_p \left[1 \pm \frac{1}{4} g(1 + 3A^2 + A^4)^{1/2} + \frac{1}{32} g^2 (3A^4 + 3A^2 - 1) + ...\right] \; . \tag{14}$$

Now we can compute the zero-point energy (the transverse degrees of fredom have a double multiplicity):

$$E = \hbar\omega_p \left[3 + \frac{g^2}{16}(3A^4 + 5A^2 - 3)\right] \tag{15}$$

and the corresponding force

$$F = \frac{\hbar\omega_p v^2}{32\pi^2}\left(\frac{3\omega_p^4}{c^4 d^3} + \frac{10\omega_p^2}{c^2 d^5} - \frac{9}{d^7}\right) \; . \tag{16}$$

We can see that in the non-retarded limit ($\omega_p d/c \ll 1$) the force is attractive and goes like $-1/d^7$; this is the van der Waals-London force; it comes from the longitudinal degrees of freedom. In the opposite, retarded limit $\omega_p d/c \gg 1$ the force is repulsive and goes like $1/d^3$; this is the limit of the Casimir force, coming entirely from the transverse oscillations. The force changes sign around $\omega_p d/c \preceq 1$ and has a maximum for $\omega_d d/c \succeq 1$. For intermediate distances the numerical coefficients in equation (16) are not reliable, since the transverse oscillations do not occur at the same distances d_n as the longitudinal ones; increasing the distance, the longitudinal and transverse oscillations contribute alternately to the repulsive force.

We have analyzed here the electromagnetic coupling between two polarizable point-like particles, modelled as point dipoles. This may be a reasonably useful model of coupled nano-plasmons and nano-polaritons. For small separation distances between the two particles (smaller than the plasma wavelength), where the non-retarded coupling regime dominates, the two particles exhibit coupled plasmons, which can be transferred from one particle to other. The zero-point fluctuations give the attractive van der Waals-London force in this case, acting between particles and behaving like $-1/d^7$, where d is the separation distance. For distances larger than the plasma wavelength the retardation comes into play and the coupling is realized through polaritons. This may happens only for certain discrete sets of separation distances (different for longitudinal and transverse oscillations of the charge density, with respect to the separation vector), in between the coupling being damped (non-coupling); it is realized either by longitudinal or transverse oscillations of the charge density, in turn. Immediateley after distances of the order of the plasma wavelength, the zero-point energy force acting between particles becomes repulsive, arising from transverse oscillations (polaritons) and going at infinity like $1/d^3$.

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