

Optics beyond the diffraction limit

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In this lecture, we introduce the basic concepts of near-field optics using two different points of view: the plane-wave angular spectrum and the electric-dipole radiation. The first approach leads naturally to the diffraction limit and to the concepts of evanescent waves and near field. The second approach shows that the basic laws of electromagnetic radiation also lead to the classical resolution limit, and allows to introduce the near-field concept based on the quasi-static approximation. In the last part, we discuss the problem of the fluorescence of a single atom close to a nano-object, based on a simple classical model. This is a key issue in nano-optics, and some important features are put forward, using the tools and ideas introduced in the section devoted to dipole radiation.

I. ANGULAR SPECTRUM OF PLANE WAVES

In this section, we describe the propagation of a light beam in a vacuum using the angular spectrum of plane waves (or plane-wave expansion) [1–3]. This tool allows to introduce in a natural way the concepts of diffraction, evanescent waves and optical near field.

A. Propagation of a light beam in a vacuum

Let us consider the propagation of a monochromatic light beam in a vacuum. The z -axis is chosen along the direction of propagation. We assume that the complex amplitude of the field is known in the plane $z = 0$. This situation allows to describe, e.g., the field transmitted by a slide of given transmissivity or the field in the plane of an opaque screen with one or several slits. For simplicity, we will consider a scalar two-dimensional wave. The result for a three-dimensional vector field will be given without derivation.

The monochromatic field of complex amplitude $E(x, z)$ and frequency ω (a temporal dependence $\exp(-i\omega t)$ is omitted throughout the text), obeys the Helmholtz equation:

$$\nabla^2 E(x, z) + \frac{\omega^2}{c^2} E(x, z) = 0 \quad (1)$$

where c is the light velocity in a vacuum. In the following, we will use the notation $k = \omega/c = 2\pi/\lambda$, λ being the wavelength. Two boundary conditions are necessary in order to have a well-defined problem: (1) we assume that the field $E(x, z = 0)$ is known, and (2) we assume that the beam propagates towards $z > 0$ (no sources in the half-space $z > 0$). We shall now show that it is possible to write the general solution of the problem in the form of an linear superposition of plane waves, known as plane-wave expansion or angular-spectrum representation.

In an arbitrary plane $z > 0$, let us introduce the Fourier transform of the field with respect to the variable x :

$$E(x, z) = \int_{-\infty}^{+\infty} \tilde{E}(\alpha, z) \exp(i\alpha x) \frac{d\alpha}{2\pi} . \quad (2)$$

Introducing (2) into (1), one obtains the equation satisfied by the Fourier transform of the field:

$$\frac{\partial^2 \tilde{E}(\alpha, z)}{\partial z^2} + \gamma^2 \tilde{E}(\alpha, z) = 0 , \text{ with } \gamma^2 = k^2 - \alpha^2 . \quad (3)$$

The general solution writes:

$$\begin{aligned} \tilde{E}(\alpha, z) &= A(\alpha) \exp(i\gamma z) + B(\alpha) \exp(-i\gamma z) \\ \text{with } \gamma &= \sqrt{k^2 - \alpha^2} \text{ if } |\alpha| \leq k \text{ and } \gamma = i\sqrt{\alpha^2 - k^2} \text{ if } |\alpha| > k. \end{aligned} \quad (4)$$

The two boundary conditions lead to $B(\alpha) = 0$ and $A(\alpha) = \tilde{E}(\alpha, z = 0)$. One finally obtains the angular-spectrum representation of the field:

$$E(x, z > 0) = \int_{-\infty}^{+\infty} \tilde{E}(\alpha, z = 0) \exp[i\alpha x + i\gamma(\alpha)z] \frac{d\alpha}{2\pi}. \quad (5)$$

In this equation, we recall that the z -component of the wavevector γ depends on α , its expression being given by Eq. (4).

The same result can be obtained for a three-dimensional vector field, which satisfies the vector form of Helmholtz equation $\nabla^2 \mathbf{E} + k^2 \mathbf{E} = 0$. One obtains for all $z > 0$:

$$\mathbf{E}(\mathbf{r}) = \int \tilde{\mathbf{E}}(\mathbf{K}, z = 0) \exp(i\mathbf{K} \cdot \mathbf{R} + i\gamma z) \frac{d^2 \mathbf{K}}{4\pi^2} \quad (6)$$

$$\text{with } \gamma(\mathbf{K}) = \sqrt{k^2 - \mathbf{K}^2} \text{ if } |\mathbf{K}| \leq k \text{ and } \gamma(\mathbf{K}) = i\sqrt{\mathbf{K}^2 - k^2} \text{ if } |\mathbf{K}| > k. \quad (7)$$

In this expression, we have used the notations $\mathbf{r} = (x, y, z)$, $\mathbf{R} = (x, y)$ and $\mathbf{K} = (\alpha, \beta)$ for the transverse wavevector. The summation is extended to the domain $0 < |\mathbf{K}| < +\infty$.

Equation (6) describes the field as a linear superposition of plane waves with wavevector $\mathbf{k} = (\mathbf{K}, \gamma)$, whose amplitude $\tilde{\mathbf{E}}(\mathbf{K}, z = 0)$ is the Fourier transform of the field in the plane $z = 0$. The variable \mathbf{K} can be understood as a spatial frequency associated to the field variations in the plane $z = 0$. If this spatial frequency satisfies $|\mathbf{K}| \leq k$ ($k = \omega/c = 2\pi/\lambda$), the corresponding plane wave is *propagating* (the z -component of the wavevector γ is real). High spatial frequencies such that $|\mathbf{K}| > k$ correspond to *evanescent* waves which decay exponentially along the z -direction (γ is purely imaginary). As a consequence, propagation acts as a low-pass filter for spatial frequencies. We will see that this spatial filtering is at the origin of the diffraction limit of classical optics.

B. Uncertainty relations and diffraction

Expression (6) of the field describes the propagation of a light beam in a vacuum. It is an exact expression, derived without any approximation. We will show that it contains diffraction effects, which appear as a natural consequence of wave propagation.

If the field $\mathbf{E}(\mathbf{R}, z = 0)$ is confined laterally over a distance $\Delta R = \Delta x \Delta y$, then the maximum spatial frequencies α_{max} and β_{max} for which its spectrum takes significant values obey:

$$\alpha_{max} \Delta x \simeq 2\pi \text{ et } \beta_{max} \Delta y \simeq 2\pi. \quad (8)$$

These relations are a consequence of the Fourier transform relationship between $\mathbf{E}(\mathbf{R}, z = 0)$ and $\tilde{\mathbf{E}}(\mathbf{K}, z = 0)$. We will refer to these relations as *uncertainty relations* in the following.

Let us consider a typical diffraction geometry, namely, a slit in an opaque screen corresponding to the plane $z = 0$, with width L along the x -direction, and infinitely long along the y -direction. The slit is illuminated from the half-space $z < 0$ by a monochromatic plane wave at normal incidence. Let us assume that the field is observed in the half-space $z > 0$, far away from the plane $z = 0$, so that only propagating waves contribute. The field in the plane $z = 0^+$ is confined along the x -direction, with a characteristic length scale L . The field in the half-space $z > 0$ is a superposition of plane waves with wavevectors (α, β, γ) . The wave with the largest angle of propagation, with respect to the z -axis, has a wavevector along the x -direction given by $\alpha_{max} \simeq 2\pi/L$ (note that the field is not confined along the y -direction, so that only the spatial frequency $\beta = 0$ contributes). This result shows that the transmitted field has an angular aperture in the $x - z$ plane. If we denote by θ the angle such that $\alpha_{max} = k \sin \theta$, which measures the angular aperture of the beam, one has $\theta \simeq \lambda/L$. We have obtained, *without any calculation of the transmitted field*, the order of magnitude of the angular aperture of a beam due to diffraction.

II. DIFFRACTION LIMIT

The preceding discussion allows to understand qualitatively the origin of the resolution limit of classical optical systems, i.e., systems using far-field imaging. Let us assume that an optical system detects only the

propagating waves coming from the object. For simplicity, we restrict the discussion to a two-dimensional system working in the $x - z$ plane. The highest spatial frequency that can be measured is $\alpha = k \sin \theta$, θ being the collection angle of the optical system ($\sin \theta$ is the numerical aperture). Using the uncertainty relationship, one can evaluate the smallest lateral variation of the field that can be detected:

$$\Delta x \simeq \frac{2\pi}{\alpha} = \frac{\lambda}{\sin \theta} \quad (9)$$

This result gives the order of magnitude of the lateral resolution of far-field optical systems. For a hemispherical detection ($\sin \theta = 1$), the resolution limit is on the order of the wavelength of the source. Its physical origin is the same as that leading to diffraction, and lies on the existence of the uncertainty relationship, which relates the lateral confinement of a beam to its angular aperture. For this reason, the resolution limit is often called *diffraction limit*.

It is possible to rewrite expressions (8) in a "quantum" manner, by identifying the plane wave with wavevector $\mathbf{k} = (\alpha, \beta, \gamma)$ to a photon with momentum $\mathbf{p} = \hbar \mathbf{k}$. This yields:

$$\Delta x \Delta p_x \simeq h \text{ et } \Delta y \Delta p_y \simeq h \quad (10)$$

where we have used the notations $\Delta p_x = \hbar \alpha_{max}$ and $\Delta p_y = \hbar \beta_{max}$. We recover the Heisenberg uncertainty relations. Note that in this context, they have been obtained as a consequence of a Fourier transform relationship. This way of writing the uncertainty relations explains why the diffraction limit in optics is often presented in textbooks as a consequence of the Heisenberg relations. We will see in the following that this may be misleading when one deals with resolution beyond the diffraction limit (this may lead to an apparent, but wrong, paradox).

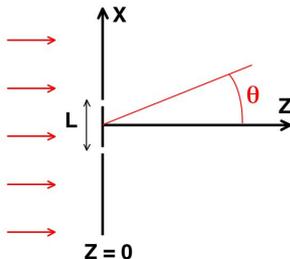


FIG. 1: *Geometry of a standard diffraction experiment with two slits in an opaque screen. The system is invariant along the y -direction, normal to the plane of the figure (two-dimensional geometry).*

The resolution limit can be obtained more precisely by calculating the diffraction pattern of two slits (of negligible width) in an opaque screen. The slits are separated by a distance L (see figure 1). For a two-dimensional geometry and a scalar field, the field in the plane $z = 0^+$ writes:

$$E(x, z = 0) = A\delta(x - L/2) + A\delta(x + L/2) \quad (11)$$

where A is a constant factor. The associated angular spectrum is readily obtained:

$$\tilde{E}(\alpha, z = 0) = A \exp(i\alpha L/2) + A \exp(-i\alpha L/2) \quad (12)$$

Let us assume that an optical system produces an image, in an arbitrary image plane, of the field in the object plane $z = 0$, and that the system only collects propagating waves, with a numerical aperture $\sin \theta$. The field in the image plane writes:

$$\begin{aligned} E_{det}(x) &= \int_{-k \sin \theta}^{+k \sin \theta} \tilde{E}(\alpha, z = 0) \exp(i\alpha x) \frac{d\alpha}{2\pi} \\ &= \left(2A \frac{\sin \theta}{\lambda}\right) \{\text{sinc}[k \sin \theta(x + L/2)] + \text{sinc}[k \sin \theta(x - L/2)]\} \end{aligned} \quad (13)$$

This field is composed of two $\text{sinc}(x) = \sin x/x$ functions, centered in $x = -L/2$ and $x = L/2$, respectively. Regarding lateral resolution, the key question is: under which condition can one separate the two slits

from a measurement of the field (or its intensity) in the image plane ? One can refer to the Rayleigh criterion, which states that the two slits can be separated if the maximum of one of the diffraction patterns coincides with the first minimum of the other one [4]. For two sinc functions, the condition can be obtained by requiring coincidence between the maximum of one function and the first zero of the other one. This leads to $k \sin \theta L = \pi$, which finally yields:

$$L = \frac{\lambda}{2 \sin \theta} . \quad (14)$$

This distance L is the smallest one which allows to distinguish the two slits in the image, according to the Rayleigh criterion. It gives the resolution limit of classical (far-field) imaging systems. For a numerical aperture $\sin \theta = 1$, one obtains $L = \lambda/2$. The order of magnitude of the resolution limit is 200 nm for visible wavelengths.

III. BEYOND THE DIFFRACTION LIMIT

A. Evanescent waves and length scales

The analysis in the preceding section shows that high spatial frequencies of the field (corresponding to sub-wavelength lateral variations of $\mathbf{E}(\mathbf{R}, z = 0)$) are exponentially attenuated on propagation away from the plane $z = 0$. Detecting sub-wavelength spatial variations of the field amounts to measuring evanescent waves. This is one of the goals of near-field optics [3, 5–7].

Let us consider a field with spatial variations in the plane $z = 0$ at a scale much smaller than the wavelength: $\Delta x \ll \lambda$ (we assume variations along the x -direction only for simplicity). What is the typical distance at which the field has to be measured in order to detect these spatial variations ? The involved spatial frequencies are on the order of $\alpha = 2\pi/\Delta x$. The associated plane waves have an exponential decay of the form:

$$\exp[-\sqrt{\alpha^2 - k^2} z] \simeq \exp(-|\alpha|z) = \exp(-z/\delta) \text{ with } \delta = \frac{\Delta x}{2\pi} . \quad (15)$$

This simple results allows to get an idea of the typical length scales of near-field optics. In order to detect a spatial variation on the order of $\Delta x = 50$ nm, one needs to detect the field at a distance $z \simeq 10$ nm.

The angular-spectrum approach allows to give a first definition of the optical near field: *the near-field is the zone in which the evanescent components of the field have a significant contribution*. The near-field zone spreads over a distance on the order of $\delta \simeq \Delta x/2\pi$, where Δx is the lateral length scale of the field (assumed to be much smaller than the wavelength λ). It is important to note that the distance δ is independent on λ . At scales much smaller than λ , the wavelength itself is not a relevant length scale. We will come back to this point in the next section, when we deal with the quasi-static limit.

B. Sub-wavelength resolution and uncertainty relations

In the preceding section, we have shown that the diffraction limit can be understood as a consequence of the uncertainty relations (8). We have also shown using the same tool (the angular spectrum of plane waves) that one can overcome the diffraction limit. Is this paradoxical ? There is actually no contradiction (see ref. [8] for a detailed discussion). If we rewrite relations (8) in the following manner:

$$\alpha_{max} \Delta x \simeq 2\pi , \quad \beta_{max} \Delta y \simeq 2\pi \quad (16)$$

$$\alpha^2 + \beta^2 + \gamma^2 = k^2 , \quad (17)$$

we see that if we allow γ^2 to take negative values (which amounts to allow the detection of evanescent waves), then the spatial frequencies α and β can be arbitrarily high, and still satisfy the dispersion relation (17). Arbitrarily high spatial frequencies correspond to arbitrarily small spatial variations Δx and Δy , as shown by relations (16). In fact, the origin of the diffraction limit lies more in the restriction to propagating waves in the detection process, than in the existence of uncertainty relations. In particular, it may be misleading to understand the diffraction limit as a consequence of the Heisenberg uncertainty

relations (10), which are no more than relations (16) written in a different manner. As we have just shown, we can obtain a lateral resolution beyond the diffraction limit by measuring evanescent components of the field, and still satisfy the Heisenberg uncertainty relations (10) and the dispersion relation (17).

IV. ELECTRIC-DIPOLE RADIATION

In this section we will introduce the optical near field with a different approach, without invoking evanescent waves. Instead, we will consider the basic properties of electromagnetic radiation. The simplest approach consists in dealing with an elementary source, namely, an electric dipole. Such a source deserves to be studied for at least two reasons: (1) many sources can be assimilated to a single electric dipole when their size is much smaller than the wavelength of radiation, and (2) electric-dipole radiation is a key point to understand spontaneous emission by a single atom (or molecule), which is a very important topic in nano-optics [9].

A. Electric field radiated by an electric dipole

Let us consider a monochromatic electric dipole with frequency ω , dipole moment \mathbf{p} , placed at the origin of the system of coordinates. The electric field radiated at point \mathbf{r} writes [4, 10]:

$$\mathbf{E}(\mathbf{r}) = \frac{k^2}{4\pi\epsilon_0} \frac{\exp(ikr)}{r} \left\{ \mathbf{p} - (\mathbf{p} \cdot \mathbf{u})\mathbf{u} - \left(\frac{1}{ikr} + \frac{1}{k^2r^2} \right) [\mathbf{p} - 3(\mathbf{p} \cdot \mathbf{u})\mathbf{u}] \right\} \quad (18)$$

where $\mathbf{u} = \mathbf{r}/r$ is the unit vector in the direction of observation.

This expression of the field contains three terms, proportional to r^{-1} , r^{-2} and r^{-3} , respectively. The first term is the far-field term, which is the only one contributing to the radiated energy flux. The last term is the near-field term, which dominates at short distance. Their properties will be discussed in the following. Note the presence of a mid-field term (proportional to r^{-2}), which shows that the concept of near field (as used in nano-optics) cannot be simply associated to the "non far-field" term. Also note that the spatial structure of the field is strongly anisotropic, especially as far as the near-field term is concerned. This anisotropy is responsible for many so-called polarization effects in near-field optics.

The transition from the far-field to the near-field regime in dipole radiation is illustrated in figure (2). In particular, we see that one wavelength away from the source (figure c), the far-field regime is already reached (no spatial variation at a scale smaller than the wavelength λ in the intensity pattern). Conversely, at sub-wavelength distance (figure a), the field exhibits sub-wavelength lateral variations.

B. Far-field radiation and diffraction limit

We will now show that by restricting the discussion to far-field radiation, we retrieve the resolution limit given by Eq. (14). To proceed, we consider two identical monochromatic point sources (electric dipoles with dipole moments \mathbf{p}) located in the plane $z = 0$, along the x -axis, and separated by a distance L . We assume that the dipoles are oriented along the y -direction, and we restrict the discussion to the $x - z$ plane for simplicity (see figure 3).

Under these assumptions, the electric field radiated in the far field, in the direction θ , writes:

$$\mathbf{E}(\mathbf{r}) = \frac{\mu_0 \omega^2}{2\pi} \frac{\exp(ikr)}{r} \cos\left(k \sin \theta \frac{L}{2}\right) \mathbf{p} . \quad (19)$$

The resulting far field, in the angular region between the two angles $-\theta_{max}$ and $+\theta_{max}$, exhibits an interference pattern: the intensity is modulated as $\cos^2(k \sin \theta L/2)$. It is possible to give a simple resolution criterion: one cannot distinguish the two sources if one cannot measure any fringe between $-\theta_{max}$ and $+\theta_{max}$. The resolution limit is reached when only one fringe remains visible. The condition writes:

$$k \sin \theta_{max} \frac{L}{2} = \frac{\pi}{2} \text{ which yields } L = \frac{\lambda}{2 \sin \theta_{max}} . \quad (20)$$

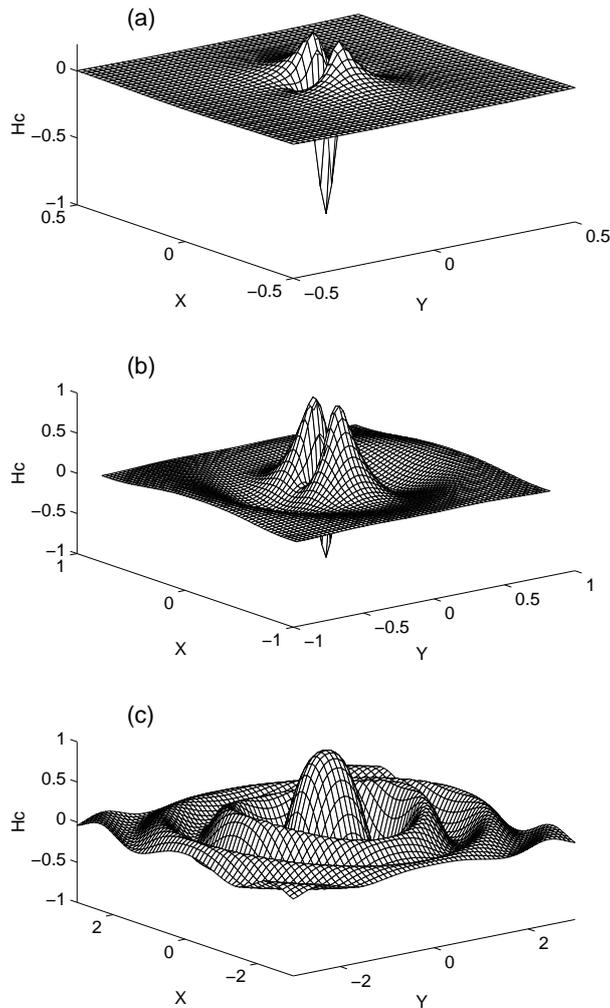


FIG. 2: Map of the near-field intensity above a glass dipole sphere deposited on a glass substrate. Illumination in total internal reflection. The z -axis is normal to the interface, which corresponds to $z = 0$. The x and y scales are normalized by the wavelength $\lambda = 633$ nm. The intensity is calculated in a plane at a constant height z . (a): $z = 0.05\lambda$, (b): $z = 0.1\lambda$, (c): $z = \lambda$.

This relation gives the minimum distance L between the two sources which allows to distinguish them in a far-field measurement. It coincides with relation (14) obtained in the previous section. The present approach shows that the resolution (or diffraction) limit is included in the basic laws of electromagnetic radiation.

C. Near-field radiation and quasi-static limit

When the observation distance r is much smaller than the wavelength λ , the electric field can be obtained from Eq. (18) in the limit $kr \rightarrow 0$. This yields:

$$\mathbf{E}(\mathbf{r}) = \frac{3(\mathbf{p} \cdot \mathbf{u})\mathbf{u} - \mathbf{p}}{4\pi\epsilon_0 r^3}. \quad (21)$$

In typical nano-optics experiments, the frequency ω is fixed, and the distance $r \rightarrow 0$. Mathematically, the limit $kr \rightarrow 0$ can also be understood as the limit $c \rightarrow \infty$, with both r and ω fixed. The limit $c \rightarrow \infty$,

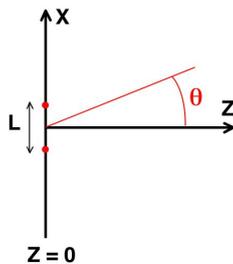


FIG. 3: Far-field radiation by two identical point sources.

in which retardation effects are neglected, is referred to as the *quasi-static* limit. In this limit, although the frequency ω is fixed to a value corresponding to visible or IR radiation, the electric field has the same spatial structure as the electrostatic field. In particular, Eq. (21) is identical to the expression of the electrostatic field of an electrostatic dipole. But in Eq. (21), both \mathbf{E} and \mathbf{p} oscillate at frequency ω (order of magnitude $10^{14} - 10^{15} Hz$!).

This point of view allows to give another definition of the optical near field: *the near-field zone is the region in which the quasi-static contributions dominate*. For point-dipole sources, this corresponds to the zone in which the terms proportional to r^{-3} in the expression of the electric field dominate.

This second definition gives an alternative point of view, compared to the approach used in section III A. In particular, it shows that in the context of the quasi-static limit, the wavelength λ is not a relevant parameter. The relevant quantities are the frequency (which determines the optical response of the objects) and the distance of observation. This second point of view explains why the wavelength does not enter relation (15), which defines the spatial scale of the near field region in the evanescent-waves approach.

V. SINGLE-ATOM EMISSION CLOSE TO NANOSTRUCTURES

In order to illustrate the concepts that have been introduced, we will discuss the problem of the spontaneous emission (or fluorescence) of a single atom (or molecule) in a nanostructured environment. We will use a simple classical model, and calculate both the modification of the decay rate and the frequency shift induced by the electromagnetic interaction with the environment. A detailed and tutorial approach of this problem can be found in ref. [11]. A survey of recent results is presented, e.g., in review articles [9, 12].

A simple model can be built by considering the atom-light interaction as the interaction between a bound electron (harmonic oscillator) and the classical electromagnetic field. Such a model predicts quantitatively the *modification* of the spontaneous decay rate due to the environment (a quantum approach is needed to calculate the absolute value of the decay rate). The calculations in this context are based on far-field and near-field electric-dipole radiation.

A. Dipole emission in free space

1. Radiated power and decay rate

Let us consider an atom (i.e. an elastically bound electron) in free space. The binding force is assumed to be proportional to the displacement \mathbf{R} of the electron from its mean trajectory, and writes $\mathbf{F}(t) = -m\omega_0^2\mathbf{R}(t)$, where m is the electron mass and ω_0 the resonance frequency of the bond. We will first consider that the electron has been excited, and oscillates around its mean trajectory at frequency ω_0 after the excitation has been removed. This accelerated electron radiates and loses energy. We will calculate the power lost by radiation, and the time evolution of the oscillator energy.

The dipole moment of the electron is $\mathbf{p}(t) = -e\mathbf{R}(t)$. For simplicity, we will use the notation $\mathbf{p}(t) = \mathbf{p}\exp(-i\omega_0 t)$ in the following and we will omit the factor $\exp(-i\omega_0 t)$. We will do the same for all

monochromatic quantities. Starting with the electric field given by Eq. (18), we can calculate the time-averaged Poynting vector $\mathbf{\Pi}(\mathbf{r}) = \epsilon_0 c/2 |\mathbf{E}(\mathbf{r})|^2$ (only the far-field term is needed in this calculation). The radiated power P is the flux of the Poynting vector across a sphere of radius r . It writes:

$$P = \frac{\mu_0 \omega_0^4}{12\pi c} |\mathbf{p}|^2 . \quad (22)$$

The dipole energy is the sum of the kinetic and potential energies, which are equal on average for a harmonic oscillator. It is given by:

$$U = \frac{m\omega_0^2}{2} |\mathbf{R}|^2 . \quad (23)$$

Expressions (22) and (23) show that the relationship $P = \Gamma_0 U$ is valid, with

$$\Gamma_0 = \frac{e^2 \omega_0^2}{6\pi m \epsilon_0 c^3} . \quad (24)$$

The quantity Γ_0 is the classical decay rate in free space. Indeed, assuming a slow decay (compared to $2\pi/\omega_0$), we can write $dU/dt = -P = -\Gamma_0 U$. Therefore, the energy decays exponentially: $U(t) = U_0 \exp(-\Gamma_0 t)$. The decay time $\tau_0 = 1/\Gamma_0$, is the classical radiative lifetime of the atom. This classical quantity is analogous to the radiative lifetime of an excited states in quantum physics. Although it is well-known that the classical model fails to explain spontaneous emission (in particular the radiative decay rate can be calculated quantitatively only in quantum mechanics), we will see that the modification of Γ_0 due to the environment can be predicted quantitatively by the classical model (see ref. [11] for a detailed discussion).

2. Dipole dynamics

The oscillating electron obeys a damped harmonic oscillator equation, which can be written:

$$\frac{d^2 \mathbf{p}}{dt^2} + \Gamma_0 \frac{d\mathbf{p}}{dt} + \omega_0^2 \mathbf{p} = 0 . \quad (25)$$

This equation accounts for both the elastic force (through the term involving the resonance frequency ω_0) and the radiative losses (through the term involving the free-space decay rate γ_0). We can search for a solution of the form $\mathbf{p}(t) = \mathbf{p} \exp(-i\Omega t)$, with Ω a complex frequency. Under the slow decay assumption $\Gamma_0 < 2\omega_0$, one obtains $\Omega = 1/2(\sqrt{4\omega_0^2 - \Gamma_0^2} - i\Gamma_0)$. This finally yields:

$$\mathbf{p} = \mathbf{p}_0 \exp(-\Gamma_0 t/2) \exp(-i\omega t) . \quad (26)$$

We retrieve the time decay of the dipole moment (a decay rate $\Gamma_0/2$ for the dipole moment corresponds to a decay rate Γ_0 for the energy). The dipole also oscillates at a frequency slightly shifted from the resonance frequency of the bond:

$$\omega = \omega_0 \sqrt{1 - \frac{\Gamma_0^2}{4\omega_0^2}} . \quad (27)$$

The frequency shift is a consequence of radiative damping.

At this stage, it is useful to give some orders of magnitude. Expression (24) gives $\Gamma_0 \simeq 10^9 \text{ Hz}$ for a frequency in the visible range ($\omega_0 \simeq 10^{15} \text{ Hz}$). The radiative lifetime is $\tau_0 \simeq 10^{-9} \text{ s}$. Note that the slow-decay condition $\Gamma_0 < 2\omega_0$ is satisfied. Also note that the frequency shift given by (27) is weak, so that we can assume $\omega \simeq \omega_0$.

B. Dipole emission in complex environments

If we now consider an atom located close to a nano-object (e.g., a nanoparticle, a tip of a near-field microscope, a microcavity, the surface of a photonic crystal), then its dynamics will be modified. Physically, the atom interacts with its own field “reflected” by the environment. We will see that both the decay rate and the emission frequency are modified with respect to their free-space value.

Let us denote by \mathbf{E}_{loc} the electric field radiated by the oscillating electron at its own position through the environment (the interaction of the electron with its self-field is described by the decay rate in vacuum Γ_0). The electron undergoes a new force $-e\mathbf{E}_{loc}$, and its equation of motion now writes:

$$\frac{d^2\mathbf{p}}{dt^2} + \Gamma_0 \frac{d\mathbf{p}}{dt} + \omega_0^2 \mathbf{p} = \frac{e^2}{m} \mathbf{E}_{loc}. \quad (28)$$

Note that the magnetic force has been neglected (it is always negligible compared to the electric force for non-relativistic electrons). Seeking a solution of the form $\mathbf{p}(t) = \mathbf{p} \exp(-i\Omega t)$, we obtain:

$$\mathbf{p} = \mathbf{p}_0 \exp(-\Gamma t/2) \exp(-i\omega t). \quad (29)$$

Under the slow-damping condition ($\Gamma_0 < 2\omega_0$), the decay rate (given by the imaginary part of the complex frequency Ω) is:

$$\Gamma = \Gamma_0 + \frac{e^2}{m\omega_0 |\mathbf{p}|^2} \text{Im}(\mathbf{p}^* \cdot \mathbf{E}_{loc}). \quad (30)$$

The frequency shift, defined by $\Delta\omega = \text{Re}(\Omega) - \omega_0$ is:

$$\Delta\omega = -\frac{\Gamma_0^2}{8\omega_0} - \frac{e^2}{2m\omega_0 |\mathbf{p}|^2} \text{Re}(\mathbf{p}^* \cdot \mathbf{E}_{loc}) \quad (31)$$

where Re and Im denote the real part and the imaginary part of a complex number, respectively, and $*$ the complex conjugate.

The decay rate and the frequency shift are different from their free-space value. The modification depends on the local field \mathbf{E}_{loc} , namely, on the radiation of the dipole on itself through the environment. Actually, the modification is independent on the dipole moment, as we will now show.

For a dipole (with dipole moment \mathbf{p}) located at point \mathbf{r}_0 , the electric field radiated at point \mathbf{r} writes:

$$\mathbf{E}_{loc}(\mathbf{r}) = \mathbf{S}(\mathbf{r}, \mathbf{r}_0, \omega_0) \mathbf{p}. \quad (32)$$

This relation simply states that the relationship is linear, and that the field and the dipole moment are, in general, not parallel. The dyadic \mathbf{S} is called the Green dyadic (or the field susceptibility). Introducing this expression of \mathbf{E}_{loc} into Eqs. (30) and (31) leads to:

$$\frac{\Gamma}{\Gamma_0} = 1 + \frac{6\pi\epsilon_0 c^3}{\omega_0^3} \text{Im}[\mathbf{u} \cdot \mathbf{S}(\mathbf{r}_0, \mathbf{r}_0, \omega_0) \mathbf{u}] \quad (33)$$

$$\frac{\Delta\omega}{\Gamma_0} = -\frac{\Gamma_0}{8\omega_0} - \frac{3\pi\epsilon_0 c^3}{\omega_0^3} \text{Re}[\mathbf{u} \cdot \mathbf{S}(\mathbf{r}_0, \mathbf{r}_0, \omega_0) \mathbf{u}] \quad (34)$$

where \mathbf{u} the unit vector along the direction of the dipole ($\mathbf{p} = p\mathbf{u}$).

These expressions give the modification of the spontaneous decay rate and the classical frequency shift, with respect to the free-space values, due to the interaction between a single atom and a nano-object. The expressions depend only of the Green dyadic $\mathbf{S}(\mathbf{r}_0, \mathbf{r}_0, \omega_0)$, which describes the electromagnetic response of the environment to a point electric-dipole excitation (more precisely, \mathbf{S} is the modification of the free-space Green dyadic due to the presence of the object). Let us point out that calculating \mathbf{S} amounts to solve a classical antenna radiation problem. Results are available in textbooks for simple geometries [13, 14]. In more complicated situations, the Green dyadic can be calculated numerically [15].

C. Example : spontaneous decay rate close to a metallic nanoparticle

In order to illustrate the preceding result, let us consider a single-atom emitting close to a spherical nanoparticle with radius $a = 5$ nm. If the distance between the atom and the surface of the particle is greater than a , then the particle can itself be described in the electric-dipole approximation (in particular, its optical properties are described by a polarizability). In this case, the Green dyadic \mathbf{S} can be calculated analytically. We show in figure 4 the decay rate versus the observation distance, for a silver particle and an emission frequency $\lambda = 354$ nm, which corresponds to the plasmon resonance of the particle. We represent the decay rate Γ , the radiative decay rate Γ_R (which corresponds to real radiation in the far field, or the emission of a photon in the quantum picture) and the non-radiative decay rate Γ_{NR} (which corresponds to absorption inside the particle). We see that at short distance, both Γ_R and Γ_{NR} increases,

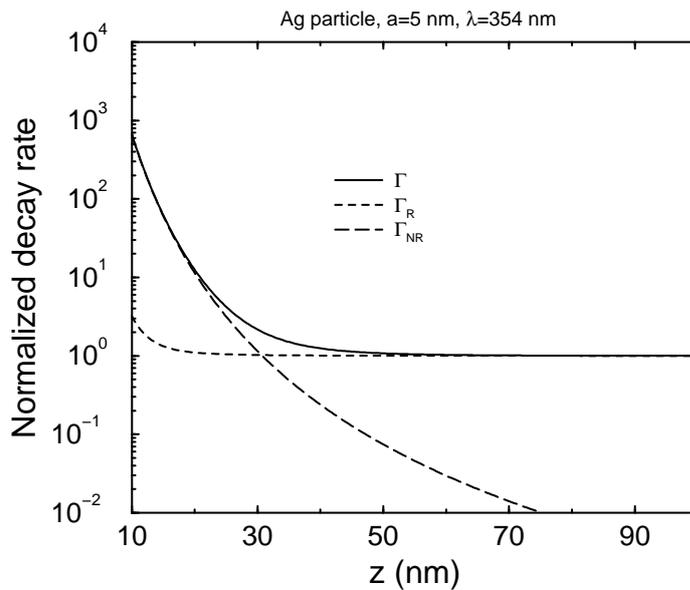


FIG. 4: Single-atom fluorescence close to a silver nanoparticle. Particle radius: 5 nm. The figure shows the decay rate Γ , the radiative decay rate Γ_R and the non-radiative decay rate Γ_{NR} . All rates are normalized by the spontaneous emission rate in free space Γ_0 . The distance z is the distance between the atom and the center of the nanoparticle. The emission wavelength is $\lambda = 354$ nm, corresponding to the plasmon resonance of the nanoparticle.

due to the near-field interaction with the nanoparticle. We also see that after a critical distance, the non-radiative rate dominates. This means that the atom decays spontaneously without emitting light, its energy being transferred to the particle and dissipated in the metal. This simple example illustrates the competition between radiative and non-radiative coupling in the interaction between an atom and a nanostructure placed at short distance. This is a key issue in nano-optics [16, 17].

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