1 Light-electron interaction

In the description of matter by an "index of refraction" or a "polarization", one tends to forget that the nature of light-matter interaction is simply re-radiation of electrons driven by the optical field. Electrons are accelerated by a combination of the applied electromagnetic field of the light and the field of other particles, and follow trajectories dependent on the light polarization. The moving electrons radiate a field that adds to that of the light, resulting in phase and amplitude changes of the optical field. This situation is traditionally described by an isotropic, polarization independent, polarizability, or index of refraction of a plasma. It is shown in the next subsection that this description does not match the response of free electrons created by tunnel ionization. It will be shown next how this case of free electrons connects to the conventional steady state response of a plasma.

1.1 Free electrons after tunnel ionization

Free electrons can be produced by ionization of a molecule under a high optical field. There are two channels of strong field ionization: multiphoton or tunneling. The two regimes are distinguished by the Keldysh parameter γ [1]:

$$\gamma = \sqrt{\frac{I_p}{2U_p}}, \tag{1}$$

where I_p is the ionization potential, and U_p is the ponderometive energy or the average kinetic energy of a free electron oscillating in the laser field. If e and m_e are the charge and mass of the electron; ω the (angular) frequency of the light field of amplitude $\mathcal{E}$:

$$U_p = \frac{e^2\mathcal{E}^2}{4m_e\omega^2}. \tag{2}$$

U_p expressed in eV as a function of the light intensity I_ℓ in W/cm^2 and the wavelength λ in microns is:

$$U_p = 9.33 \cdot 10^{-14} I_\ell \lambda^2 \tag{3}$$

In the "quasistatic limit" of $\gamma < 1$ the dressed Coulomb barrier is essentially static as seen by the electrons and the method of releasing the electrons is dominated by *tunneling*. For $\gamma > 1$ the electron release is most likely described by photon absorption, and *multiphoton* features are more dominant [2]. The difference between tunneling and multiphoton is easily recognized in measurements of velocity mapping imaging (VMI) where the electron momentum distribution following ionization is measured [3]. We consider here as an example the case of ionization by a fs pulse at 800 nm where a tunneled electron leaves its parent atom/molecule instantaneously along the direction of light polarization, at the moment of ionization, with zero velocity [4]. The electrons leave the atom from a Rydberg state that typically has an orbit radius one order of magnitude larger than the atomic radius. Formulae can be found in the literature for the tunneling rate and the ratio of electron production for various polarization [5, 6]. We are here just interested in following the motion of the electron, subjected to the force F due to a combination of the optical field E and a Coulomb field F_c:

$$F = -qE + F_c = ma, \tag{4}$$

where a is the acceleration of the electron of mass m and charge q. In this classical approach, we neglect the magnetic force on the electron. The tunneled electron is released at time t_0 in the optical field given by:

$$E = \frac{\mathcal{E}(t,r)}{\sqrt{1+\varepsilon}} \left[\cos\omega(t-t_0)\vec{x} + \varepsilon \sin\omega(t-t_0)\vec{y}\right], \tag{5}$$

where ε defines the light polarization ($\varepsilon = 0$ for linear polarization) and $\mathcal{E}(r,t)$ is the envelope of the field. At any time $t \geq t_0$, the velocity of the electron is given by:

$$v = \frac{q\mathcal{E}(t,r)}{m\omega}(\sin\omega t\ \vec{x} - \varepsilon\cos\omega t\ \vec{y}) + \vec{y}\varepsilon\frac{q\mathcal{E}(t_0,r_0)}{m\omega}. \tag{6}$$

In circular polarization ($\varepsilon = 1$), the electron acquires a drift velocity $v_d = q\mathcal{E}(t_0,r_0)/(m\omega)$ along $\vec{y}$, long after the laser pulse is gone. At the moment of ionization $t_0 = 0$, the electron velocity is zero, hence there must be a drift term to fulfill the initial condition. Let us consider a pulse of intensity of $2.8{\cdot}10^{14}$ W/cm^2 as is realized in a light filament in air (see Section **??**). To this circularly polarized pulse corresponds a field peak amplitude of 4.62×10^8 V/cm, the drift velocity of the electron ionized by this field is $3.45 \cdot 10^4$ cm/s or 1.6 atomic units.

The position of the electron is

$$r = \frac{qE_0}{m\omega^2}(-\cos\omega t\vec{x} - \vec{y}\epsilon\sin\omega t) + \vec{y}\epsilon\frac{qE_0}{m\omega}t + r_0 + \frac{qE_0}{m\omega^2}\vec{x}. \tag{7}$$

It means that the electron having the negative charge will oscillate in the same direction and phase of the laser field. Consistently with neglecting the magnetic forces, we ignore the motion out of the polarization plane "xy plane". The coordinates of the electron are:

$$x = \frac{qE_0}{m\omega^2}(-\cos\omega t) + x_0 + \frac{qE_0}{m\omega^2} \tag{8}$$

$$y = \epsilon\frac{qE_0}{m\omega}t(-\frac{\sin\omega t}{\omega t} + 1). \tag{9}$$

The initial position is taken to be 10 times the atomic radius of nitrogen which is 65 picometers or 0.65/0.52918 = 1.22 atomic units. The amplitude of the oscillation is $qE_0/(m\omega^2) = 1.4$ nm corresponding to 27.7 atomic units. Within the 200 femtosecond of a circularly circularly polarized pulse, the electron ionized at the peak has moved $qE_0/(m\omega)t$ which is $1.23{\cdot}10^2$ nm or $6.52\ {\cdot}10^3$ atomic units in 100 fs.

The radiation of a non-relativistic moving charge [7] is expressed as

$$\Delta E = \frac{q}{\epsilon_0 c} \frac{\vec{n} \times (\vec{n} \times \dot{\vec{\beta}})}{R} + \frac{qd}{\epsilon_0 R^3}. \tag{10}$$

where $\beta = v/c$, ϵ_0 is the vacuum permittivity, $\vec{n} = \vec{R}/R$ is the unit vector of the observation point $\vec{R}$ and "d" is the displacement of the charge that can be calculated at time t from the position equations (9). Note that there are two terms in the electron response: the first one is the "radiation term", and is only relevant at very high intensities. In our example of $2.8 \cdot 10^{14}$ W/cm^2 considered here, it is two orders of magnitude smaller than the second term. Since the latter involves the distance from the parent ion to the electron, it is called the "dipole term". The classical definition of the polarization relates to this dipole term, generally defined as $P = Nqd$, where N is the density of electrons. This definition relates to the second term of Eq. (10) in an homogeneous medium where $R^{-3} = N$, and the field of the electron cloud reacting to the applied field is $\Delta E = P/\epsilon_0$.

The electron trajectories in the first ps after ionization and their emission into the applied field is a deterministic problem that can only be solved by numerical calculations. Some calculations of the transient response of the electron cloud in linear polarization were reported by Romanov and Levis [8]. An example of the transient response is reproduced in Fig. 1.

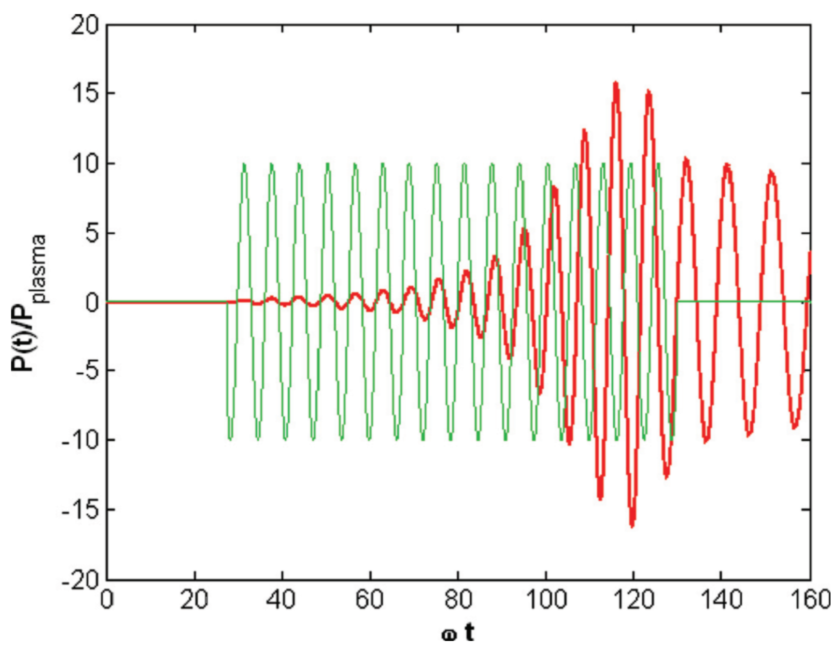

Figure 1: The cumulative polarization response of a medium that is being tenuously ionized by a laser pulse with rectangular envelope. The laser electric field oscillations are shown for comparison, not to scale. (from [8]) REQUEST PERMISSION

For mixed gases the contribution of each material (in the absence of interaction) can be calculated separately. The distance between electrons changes with time and position. The response of the electrons is a field ΔE, calculated for each point in space as a function of time, which modifies the applied field: $E(z + \Delta z) = E(z) + \Delta E$. The radiated field ΔE is related to the traditional notion of index of refraction $n(z,t)$ (no longer a constant) by the propagation equation written in the slowly varying envelope approximation and in retarded time:

$$\frac{\Delta E}{E} = -ik\Delta z = -i\frac{2\pi n}{\lambda}\Delta z \tag{11}$$

Note that this approach is not restricted to a particular motion. If the medium is excited by multiple laser frequencies or existing nuclear and electromagnetic fields, they all contribute in the motion of the electron and therefore its radiation.

The response due to the dipole radiation of the electrons at position r is calculated by time integration of Eq. (6) and inserted in the dipole term of radiation equation Eq. (10).

$$\Delta E_p(t,r) = \frac{qd(t,r)}{R(t,r)^3\epsilon_0} = \frac{q^2\mathcal{E}(t,r)}{2m\omega^2 R(t,r)^3\epsilon_0}, \tag{12}$$

in which $\tilde{E}(t,r)$ is the pulse envelope. Note that the dipole radiation exists only during the laser pulse. In this particular case the radiation of the moving electron agrees with the Drude model, which is detailed in Section 1.2 that follows. It leads to an index of refraction

$$\Delta n = -\frac{{\omega_p}^2}{\omega^2} = -\frac{Nq^2}{2\epsilon_0 m\omega^2}, \tag{13}$$

where ω_P is a time dependent plasma frequency that depends on the density of electrons N at each instant. Note that in a general case the motion of an electron is influenced by existing electromagnetic fields, collisions and Coulomb forces, and therefore the refractive index of electrons can not be defined solely by the density. Tunneled electrons with circularly polarized light withhold a drift velocity [Eq. (6)] that is determined by the field value at the moment of ionization. The spiral motion of the tunneled electrons results in generating an expanding sphere in time. The electromagnetic fields in the presence of moving matter are related through Maxwell's equations, suitably modified to include the effects of motion upon the electric and magnetic properties of matter [9]. We assume that the expanding electron sphere in time has the constituent parameters of free space ($\mu = \mu_0$ and $\epsilon = \epsilon_0$). Let us assume that the expanding electron sphere is a perfect conductor with the field zero for $r < b$, where b is the radius of the sphere. One relation is necessary to complete the set of basic equations, which is Ohm's law for a perfect moving conductor

$$E + v \times \mu_0 H = 0. \tag{14}$$

Here v is the velocity of a macroscopic element of volume of the moving conductor. The solutions of Maxwell's equations inside and outside the expanding sphere have to be matched across a moving surface. Due to the requirement of regularity at infinity, the problem is defined only by the magnetic vector potential ' A .

$$H = -\frac{1}{\mu_0}\nabla\times A \tag{15}$$

$$E = -\frac{\partial A}{\partial t} \tag{16}$$

and is the solution of

$$\nabla^2 A - \frac{1}{c^2}\frac{\nabla A^2}{\nabla t^2} = -\mu_0 J, \tag{17}$$

where J is the electric current density. Using the Green's function, the field at distance r from the center of a sphere moving with constant velocity [10] is

$$E = -\frac{\mu_0}{c^2}\frac{3Hv^3}{(1-v/c)^2(1+2v/c)}\left[\frac{T}{rc}+\frac{T^2}{2r^2}\right], \tag{18}$$

where $T = t - r/c$. In the case of tunneled electrons with circularly polarized light $r = R$ is the distance between the electrons , v is the expansion velocity of the sphere (the drift velocity $qE(t_0, r_0)/(m\omega)$ in Eq. (6) and $T = a/v - R/c$ where "a" is the radius of the sphere at a given time.

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1.2 Steady state limit: the Drude model

It is easy to associate a characteristic resonant frequency to an oscillator with a positive and negative charge. Associating the resonant frequency of Eq. (13) with a homogeneous electron plasma may seem less obvious. If an electron moves in the plasma from its equilibrium position, there will be a restoring force. The larger the number of surrounding electrons, the larger the restoring force, which explains the density dependence of the resonant frequency.

Let us consider a volume of electrons, of density n_0. The equation of motion of electrons under the influence of an electric field, neglecting collisions and magnetic forces, is:

$$m_e\frac{dv}{dt} = -eE + e[v\times B] - m_e\nu_c v \tag{19}$$

Note that in the equation of motion of the electron, the electric field can be the Coulomb field from the surrounding electrons. Let us consider a perturbation δn_e from the equilibrium density of the electrons n_0. We will for simplicity neglect collisions and the magnetic force in the following derivation. Expressing that the change in the number of electrons per unit time in a infinitesimal volume is equal to a source term, minus the current of particles out of that volume, leads to the conservation equation for the electrons:

$$\left(\frac{\partial n}{\partial t} + \nabla n v = \text{Source terms}\right). \tag{20}$$

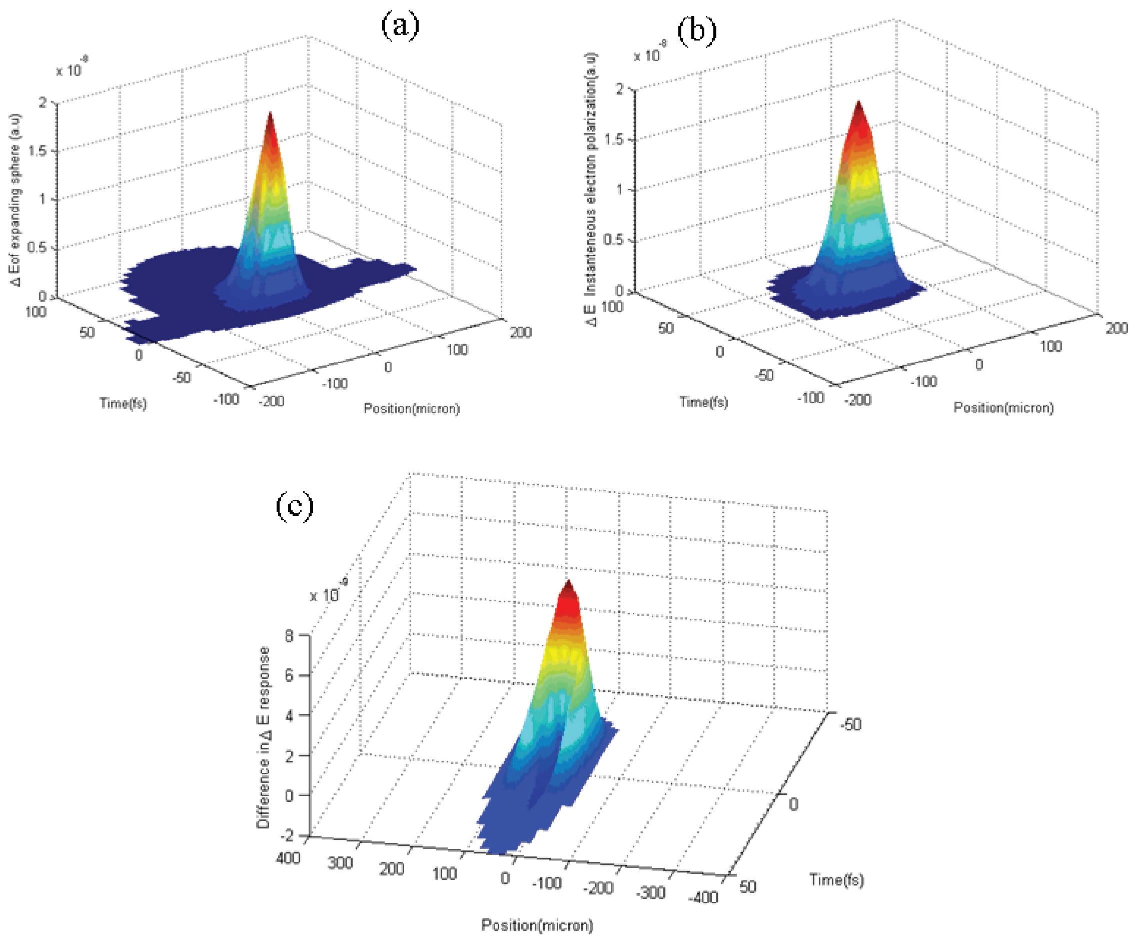

Figure 2: a) Radiation field of expanding spherical conductors b) Dipole radiation c) Difference between Dipole radiation and radiation due to the drift motion

with $n = n_0 + \delta n_e$ and $\delta n_e \ll n_0$. In the velocity $v = v_0 + \delta v$, we assume no drift velocity ($\delta v = 0$. The conservation equation (without source term – the plasma is at equilibrium), neglecting the second order product $\delta n \delta v$, leads to:

$$\nabla \cdot \delta v = \frac{-1}{n_0}\frac{\partial n}{\partial t}. \tag{21}$$

Taking the divergence of Gauss law, and using the equation of motion (19):

$$\nabla \cdot eE = \frac{ne^2}{\epsilon_0} = m_e \nabla \cdot \frac{dv}{dt} \approx m_e \frac{d}{dt} \nabla \cdot \delta v = m_e \frac{d}{dt}\left(\frac{-1}{n_0}\frac{\partial n}{\partial t}\right) \tag{22}$$

which leads to the differential equation for the plasma density:

$$\frac{\partial^2 n}{\partial t^2} = -\left(\frac{n_0 e^2}{m_e \epsilon_0}\right) n = -\omega_p^2 n, \tag{23}$$

which shows that indeed, density fluctuations in a plasma of electron have a resonant frequency.

The fluctuation of the density (position) of electrons gives rise to an electric field. Considering that there is no other electric field (no applied field), using Ampere law:

$$\begin{aligned}\nabla \times H &= J + \frac{\partial D}{\partial t} \\ D &= \epsilon E \\ J &= -nqv \\ & \frac{\partial}{\partial t}\left(\epsilon \frac{\partial E}{\partial t} = nqv\right) \\ \frac{\partial^2 E}{\partial t^2} &= \frac{n_0 q}{\epsilon_0}\frac{\partial v}{\partial t}\end{aligned}$$

where we have set the magnetic field to zero. Since $\partial v/\partial t = -qE/m_e$ from the equation of motion,

$$\frac{\partial^2 E}{\partial t^2} = -\left(\frac{n_0 q^2}{m\epsilon_0}\right) E \tag{24}$$

we see that the density fluctuation themselves give rise to the emission of a field at the plasma frequency ω_p.

The classical treatment of electron in plasma is not very different from the bound electron: it a is stationary solution of a driven oscillator, based on a fundamental assumptions that the medium response is isotropic and stationary. In particular, the density term that defines the plasma frequency is never a constant when dealing with fs pulses.

2 Transitions with bound electrons

2.1 Introduction: the classical oscillator and Maxwells equations

The classical approach is to calculate the motion of the bound electron, modeled as a dipole. The electron is at a (small) distance d from the positive ion. It oscillates with the applied electric field. This is the classical oscillator model. The Coulomb field produces a restoring force, which leads to a resonance frequency. One introduces a damping term. A similar model is used for the plasma. The result is that, away from resonance, under the influence of an optical oscillating field at ω, the motion of the electron follows the frequency of the applied field, in phase, and is thus $d = d_0 \cos\omega t$. At a point of observation at a distance R from the dipole, the field due to the dipole is:

$$\Delta E = \frac{q^2}{4\pi R^2}\left[1 - \frac{R^2}{(R+d)^2}\right] \approx \frac{2q^2 d}{4\pi R^3} \tag{25}$$

Putting that in Maxwell's propagation equation:

$$\frac{\partial^2 E}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2 (E + \Delta E)}{\partial t^2} = 0 \tag{26}$$

or

$$\frac{\partial^2 E}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = \frac{1}{c^2}\frac{\partial^2 \Delta E}{\partial t^2} = \frac{\omega^2}{c^2}\Delta E. \tag{27}$$

Using:

$$\begin{aligned} E &= \frac{1}{2}\mathcal{E} e^{i(\omega t - kz)} \\ \Delta E &= \frac{1}{2}\Delta\mathcal{E} e^{i(\omega t - kz)} \end{aligned}$$

we find:

$$-2ik\frac{\partial \mathcal{E}}{\partial z} - 2i\frac{\omega}{c^2}\frac{\partial \mathcal{E}}{\partial t} = \frac{\omega^2}{c^2}\Delta\mathcal{E}, \tag{28}$$

and

$$\frac{\partial \mathcal{E}}{\partial z} + \frac{1}{c}\frac{\partial \mathcal{E}}{\partial t} = -i\frac{\omega}{2c}\Delta\mathcal{E}, \tag{29}$$

Even though we started from a $\Delta\mathcal{E}$ in phase with the applied field, after insertion in the propagation equation it appears that its envelope is adding 90 degrees out of phase with the applied field, as is the case of an index of refraction.

It appears as if, by the time the electron re-radiates, the wave has already moved by a distance of $\lambda/4$.

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