Non-linear optics for dummies

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Most field of science are taught following historical developments. For instance, geometry started with cartesian coordinates, in which circles, ellipses hyperbolae are totally different and unrelated objects. Going from cartesian to slanted coordinates, one realizes that circles are just a particular case of ellipses. It took projective coordinates to realize that circles, ellipses and hyperbolae are just one object. Teaching analytical geometry in particularizing from the general projective coordinates towards the more narrow minded cartesian one gives a much richer and elegant understanding of geometry.

The same can be said of light-matter interaction, in particular nonlinear optics. We have taken the conventional approach in the preceding chapters, by describing as matter as an ensemble of electric dipoles, which respond linearly to the electric field. The next step, if we follow the ‘historical” approach, would be to expend the polarization in a power series of the applied field, to describe nonlinear optics. Another aspect of the same “weak field” nonlinearities is typically presented by a quantum mechanical perturbation treatment. In most nonlinear optics treatments, there is a chapter suggesting that it is the index of refraction that should be expanded in a power series of the intensity. Then it is suggested that the power expansion is sometimes invalid. Finally, there is generally a chapter on “short pulse nonlinear optics”.

It would be a daunting, but very useful task, to re-organize the field from the more general view of atom-field interaction, proceeding by successive approximations, down to the most particular case of classical nonlinear optics. A more modest approach will be used here, limited to semi-classical models of electron - atom - molecule near resonant interactions, to non-resonant
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nonlinear optics. Nearly all problems of linear and nonlinear optics are generally treated in a stationary approximation. Ultrashort pulses are bringing the awareness that not all situation can be treated as “steady-state”, the latter being an asymptotic limit of a transient behavior.

1.1 Laser-electron interaction

In the description of matter by an “index of refraction” of 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.

1.1.1 Free electrons after tunnel ionization

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

\[ \gamma = \sqrt{\frac{I_p}{2U_p}} \] (1.1)

where “\( I_p \)” is the ionization potential and \( U_p \) is the ponderomotive energy, average kinetic energy of a free electron oscillating with the laser field. 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
atom has a radius defined by atomic radius; electrons leave the atom from a so called 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 $F_c$:

$$F = -qE + F_c = ma,$$  \hspace{1cm} (1.2)

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:

$$E = \frac{\mathcal{E}_0(t, r)}{\sqrt{1 + \varepsilon}} \left[ \cos \omega(t - t_0)x + \varepsilon \sin \omega(t - t_0)y \right],$$  \hspace{1cm} (1.3)

where $\varepsilon$ 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:

$$v = \frac{q\mathcal{E}(t, r)}{m\omega}(\sin \omega t x - \varepsilon \cos \omega t y) + \frac{q\varepsilon}{m\omega}\mathcal{E}(t_0, r_0).$$  \hspace{1cm} (1.4)

In circular polarization ($\varepsilon = 1$), the electron acquires a drift velocity $v_d = \frac{q\mathcal{E}(t_0, r_0)}{(m\omega)}$ along $y$ 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. In circular polarization, with a corresponding field of $4.62 \times 10^8$ 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

$$r = \frac{qE_0}{m\omega^2}(-\cos \omega t x - y\varepsilon \sin \omega t) + \frac{q\varepsilon}{m\omega}t + r_0 + \frac{qE_0}{m\omega^2}x.$$  \hspace{1cm} (1.5)

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:

$$x = \frac{qE_0}{m\omega^2}(-\cos \omega t) + x_0 + \frac{qE_0}{m\omega^2}$$  \hspace{1cm} (1.6)

$$y = \frac{q\varepsilon}{m\omega}t(-\frac{\sin \omega t}{\omega t} + 1).$$  \hspace{1cm} (1.7)
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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 have moved $qE_0/(m\omega)t$ which is $1.23\times10^2$ nm or $6.52\times10^3$ atomic units in 100 fs.

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

$$\Delta E = \frac{q}{\epsilon_0c} \frac{\vec{n} \times (\vec{n} \times \beta)}{R} + \frac{qd}{\epsilon_0R^3}. \quad (1.8)$$

where $\beta = v/c$, $\epsilon_0$ is the vacuum permittivity, $\vec{n}$ is the unit vector of the observation point $\vec{R}/R$ and “$d$” is the displacement of the charge that can be calculated at time “$t$” from the position equations (1.7). 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 $2.8\times10^{14}$ 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 defines as $P = Nqd$, where $N$ is the density of electrons. This definition relates to the second term of Eq. (1.8) 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. 1.1.

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 $\Delta E$, calculated for each point in space as a function of time, which modifies the applied field: $E(z+\Delta z) = E(z) + \Delta z$. The radiated field $\Delta E$ 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:

$$\frac{\Delta E}{E} = -ik\Delta z = -i\frac{2\pi n}{\lambda} \Delta z \quad (1.9)$$

Note that this approach is not restricted to a particular motion. If the medium is excited by multiple laser frequencies or existing nuclear and elec-
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Figure 1.1: 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])

tromagnetic 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.1.4 and inserted in the dipole term of radiation equation Eq.1.8.

\[
\Delta E_p(t, r) = \frac{qd(t, r)}{R(t, r)^{3}\varepsilon_0} = \frac{q^2 \tilde{E}(t, r)}{2m\omega^2 R(t, r)^{3}\varepsilon_0},
\]

(1.10)
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 the next section:

\[
\Delta n = \frac{\omega_p^2}{\omega^2} = -\frac{Nq^2}{2\varepsilon_0 m\omega^2},
\]

(1.11)
if one considers a time dependent plasma frequency \(\omega_P\) 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, 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. 1.4) that is determined by the field value
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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

$$E + v \times \mu_0 H = 0$$

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 “$A$”.

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

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

and is the solution of

$$\nabla^2 A - \frac{1}{c^2} \frac{\nabla A^2}{\nabla t^2} = -\mu_0 J,$$  \(1.15\)

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

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

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. (1.4) and “$T = a/v - R/c$” where “$a$” is the radius of the sphere at a given time. The total response at each point in the beam cross section at a given time “$t'$” is calculated by adding all the responses of expanding spheres from ‘the initial ionization time “$t_i = -\infty$” to the final observation time “$t_f = t'$”, weighted
Figure 1.2: a) Radiation field of expanding spherical conductors b) Dipole radiation c) Difference between Dipole radiation and radiation due to the drift motion

by the probability of ionization at the moment of ionization \( t_i \).

\[
\Delta E_d(t_f, r) = -i \int_{t_i = -\infty}^{t_f = t'} 3J_0(c^2 W(t_i) \frac{E(t)v(t_i)^3}{(1 - v(t_i)/c)^2(1 + 2v(t_i)/c)} \left[ \frac{T}{Rc} + \frac{T^2}{2R^2} \right] dt.
\]

The refractive index due to the expansion of electron spheres [Fig. 1.2 (a)] is compared to the dipole radiation [Fig. 1.2 (b)] with circularly polarized light. The difference between the two is presented in Fig. 1.2 (c). The dipole index is stronger in the first half of the pulse. The index due to the drift is stronger in the central portion of the beam profile and in later times of the laser pulse. Note that the two responses have 90 degree phase difference; dipole index is a real index and the drift index is an imaginary (absorption index). The drift index (imaginary) of tunneled electrons with
linear polarization is neglected because of their low drift velocity and the index is mostly due to dipole radiation (real index).

Another point of view in connecting microscopic effects such as light polarization to macroscopic effects in filaments can be seen through conservation of energy. In the strong field ionization [11] conservation of energy enforces

\[ N_{\text{photon}} \hbar \omega = I_p + U_p + K, \]  

(1.18)

where “K” is the kinetic energy of the electrons and “\(N_{\text{photon}}\)” is the number of absorbed photons in the ionization process. \(I_p\) is the energy required to release an electron and \(U_p\) is the ponderomotive energy due to the oscillation of electrons with the applied field. For the filament intensities the electron kinetic energy “K” of linearly polarized light can be neglected with respect to the energy of electrons with circularly polarized light [12]. The plasma generated with circular polarization has higher energy (higher temperature).

1.2 Steady state limit: the Drude model for plasma and the bound electron

1.2.1 Electron gas

It is easy to associate a characteristic resonant frequency to an oscillator consisting in a dipole consisting in a positive and negative charge. Associating the resonant frequency of Eq. (1.11) 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:

\[ m_e \frac{dv}{dt} = -eE + e[v \times B] - m_e \nu_e v \]  

(1.19)

Note that in the equation of motion of the electron, the electric field can be Coulomb field from the surrounding electrons. Let us consider a perturbation \(\delta n_e\) 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 number of electrons per unit time in a
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An infinitesimal volume is equal to a source term, minus the current of particles out of that volume, leading to the conservation equation for the electrons:

\[
\left( \frac{\partial n}{\partial t} + \nabla n v = \text{Source terms} \right).
\]  

(1.20)

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 \( (\delta v = 0) \). From the conservation equation (without source term – the plasma is at equilibrium), neglecting the second order product \( \delta n \delta v \):

\[
\nabla \cdot \delta v = -\frac{1}{n_0} \frac{\partial n}{\partial t}.
\]  

(1.21)

Taking the divergence of Gauss law, and using the equation of motion (1.19):

\[
\nabla \cdot e E = \frac{ne^2}{\epsilon_0} = m_e \nabla \cdot \frac{dv}{dt} \approx m_e \frac{d}{dt} \nabla \cdot \delta v = m_e \frac{d}{dt} \left( -\frac{1}{n_0} \frac{\partial n}{\partial t} \right)
\]  

(1.22)

which leads to the differential equation for the plasma density:

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

(1.23)

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

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

\[
\nabla \times H = J + \frac{\partial D}{\partial t}
\]

\[
D = \epsilon E
\]

\[
J = -nev
\]

\[
\frac{\partial}{\partial t} \left( \epsilon \frac{\partial E}{\partial t} = -nev \right)
\]

\[
\frac{\partial^2 E}{\partial t^2} = -\frac{n_0 \epsilon}{\epsilon_0} \frac{\partial v}{\partial t}
\]

where we have set the magnetic field to zero. Since \( \partial v/\partial t = eE/m_e \) from the equation of motion,

\[
\frac{\partial^2 E}{\partial t^2} = -\left( \frac{n_0 e^2}{m_e \epsilon_0} \right) E
\]  

(1.24)
we see that the density fluctuation themselves give rise to the emission of a
field at the plasma frequency $\omega_p$.

The classical treatment of electron in plasma is not very different from
the bound electron: is it a stationary solution of a driven oscillator. It is
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.

1.2.2 The bound electron

The classical approach is to calculate the motion of the bound electron, model-
ed 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 oscil-
lator model. The Coulomb field produces a restoring force, which defines 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 in-
fluence of an optical oscillating field at $\omega$, 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:

$$\Delta E = \frac{q^2}{4\pi R^2} \left[ 1 - \frac{R^2}{(R + d)^2} \right] \approx \frac{2q^2 d}{4\pi R^3}$$  \hspace{1cm} (1.25)

Putting that in Maxwell’s propagation equation:

$$\frac{\partial^2 E}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 (E + \Delta E)}{\partial t^2} = 0$$  \hspace{1cm} (1.26)

or

$$\frac{\partial^2 E}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} = \frac{1}{c^2} \frac{\partial^2 \Delta E}{\partial t^2} = \frac{\omega^2}{c^2} \Delta E.$$  \hspace{1cm} (1.27)

Using:

$$E = \frac{1}{2} E e^{i(\omega t - kz)}$$

$$\Delta E = \frac{1}{2} \Delta E e^{i(\omega t - kz)}$$

we find:

$$-2ik \frac{\partial E}{\partial z} - 2\frac{\omega}{c^2} \frac{\partial E}{\partial t} = \frac{\omega^2}{c^2} \Delta E,$$  \hspace{1cm} (1.28)

and

$$\frac{\partial E}{\partial z} + \frac{1}{c} \frac{\partial E}{\partial t} = -i \frac{\omega}{2c} \Delta E,$$  \hspace{1cm} (1.29)
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Even though we started from a $\Delta E$ 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.

**Source polarization in Maxwell’s equations** For a propagating wave along $z$, the propagation of any function $f(z - ct)$ would be solution of a first order differential equation:

\[
\left( \frac{\partial}{\partial z} - \frac{1}{c} \frac{\partial}{\partial t} \right) f = 0. \tag{1.30}
\]

[which amount to $f'_z - (1/c)f'_t = 0$.] Maxwell’s wave equation is a bit different: it could be seen as the product of two wave equations along $+z$ and $-z$: (right propagation) $\times$ (left propagation)

\[
\left( \frac{\partial}{\partial z} - \frac{1}{c} \frac{\partial}{\partial t} \right) \times \left( \frac{\partial}{\partial z} + \frac{1}{c} \frac{\partial}{\partial t} \right) = \frac{\partial^2}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2}. \tag{1.31}
\]

The right hand side of Maxwell’s equations refers to local sources – not propagating ones, hence no $\partial/\partial z$ associated with them. Magnetic field effects are neglected in the derivation of plasma frequencies, polarization, approximation incompatible with propagation. Indeed, the magnetic field is an integrant part of Maxwell’s propagation equation.

**Recalling the derivation of Maxwell’s propagation equation** :

\[
\begin{align*}
\nabla \times E &= -\frac{\partial B}{\partial t} \\
\nabla \times B &= \mu_0 J + \mu_0 \frac{\partial (\epsilon_0 E + P)}{\partial t} \\
\nabla \times \nabla \times E &= -\frac{\partial (\nabla \times B)}{\partial t} = -\frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} - \frac{\partial^2 P}{\partial t^2} \\
\frac{\partial^2 E}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} &= \mu_0 \frac{\partial^2 P}{\partial t^2}. \tag{1.32}
\end{align*}
\]

Another way to look at it: instead of $P$, we are adding the local electric field $\Delta E$ from the radiation of the electron. Either of:

1. say $\Delta E = P/\epsilon_0$ hence

\[
\frac{\partial^2 E}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} = \frac{1}{c^2} \frac{\partial^2 \Delta E}{\partial t^2}
\]
2. The local electric field adds to the applied field only in the time derivative:

\[
\frac{\partial^2 E}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 (E + \Delta E)}{\partial t^2} = 0
\]

\(\Delta E\) may be in phase with the applied field, but, as a function of \(z\), and in the retarded frame of reference, \(\Delta \tilde{E}\) is \(\pi/2\) out of phase with \(\partial \tilde{E}/\partial z\). The standard propagation equation for the envelope

\[
\frac{\partial \tilde{E}}{\partial z} = -i \frac{\Omega}{c} \Delta \tilde{E}
\]  

(1.33)

comes from making the substitution:

\[
E = \frac{1}{2} \tilde{E} e^{i(\omega t - kz)}
\]

\[
\Delta \tilde{E} = \frac{1}{2} \Delta \tilde{E} e^{i(\omega t - kz)}
\]

The left side of Maxwell’s propagation equation becomes zero to first order by making \(\omega^2/c^2 = k^2\), while the right hand side becomes either \(\mu_0 \omega^2 P\) or \(\omega^2/c^2 \Delta \tilde{E}\).

\[
(-2ik \frac{\partial}{\partial z} - 2\frac{\omega}{c^2} i \frac{\partial}{\partial t}) \tilde{E} = -\frac{\omega^2}{c^2} \Delta \tilde{E}
\]

\[
\left( \frac{\partial}{\partial z} + \frac{\partial}{\partial t} \right) \tilde{E} = -i \frac{\omega}{2c} \Delta \tilde{E},
\]

or, going to the retarded frame \(t \leftarrow t - z/c\) and \(z \leftarrow z\):

\[
\frac{\partial \tilde{E}}{\partial z} = \frac{i \omega}{2c} \Delta \tilde{E}.
\]  

(1.34)

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


