Multiple Reflections–Cascaded Upshifting of Laser Pulses by Semiconductors

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Abstract

By studying the detailed dynamics of an ultrashort pulse at the semiconductor–vacuum interfaces, we have shown that the interaction of these pulses with nonstationary semiconductor plasmas can, under appropriate conditions, lead to a variety of interesting phenomena: controlled upshifting of the laser frequency, a possible cascading of upshifting, a trapped pulse bouncing back and forth in the sample, and a "machine–gun" of increasingly blueshifted pulses.

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Recent new developments in the generation of femtosecond pulses have brought a wide variety of new phenomena within the realm of investigation. It has become possible, for instance, to systematically study the expected [1] frequency changes in a pulse as it passes through a medium whose dielectric properties vary as a function of time. As an intense femtosecond laser pulse propagates through a gaseous medium, it may encounter a rapid increase in the electron density (and hence a decreasing refractive index) due to an optical–field induced ionization (multiphoton or tunneling) due to external sources or due to the pulses itself. Traversing a region of time varying refractive index, the incident laser pulse will experience a frequency upshift [2]. Recent experiments by Downer and his coworkers [3] have shown that a 100 fs, 620 nm pulse with peek intensity $I = 10^{16}\mathrm{W/cm}^2$ can be blueshifted by $10-50$ nm in the noble gases. This demonstration has stimulated considerable interest in the development of plasma radiation sources which can potentially provide coherent tunable radiation from the millimeter to the X–ray range of wavelengths. The gaseous media, however, may not be always the best or the most efficient vehicles for this purpose. It is generally difficult to have reproducible gaseous discharges; the observed blueshifts are also quite small and even these require comparatively large laser intensities ($I > 10^{15}\mathrm{W/cm}^2$). Only for these intensities, the electric field of the laser is comparable to the Coulomb field of the atom, and flash ionization through the tunneling effect, can produce the needed plasma.

It is natural to wonder if it is possible to find an alternative to the gaseous media in which much smaller laser intensities could induce fast time–variations in the refractive index. Fortunately semiconductors do provide such an alternative. In semiconductors, even moderately intense femtosecond laser pulses can cause a large and fast change in the refractive index by creating a dense electron–hole plasma. Free carrier pairs are created through multiphoton processes whose probability increases with the laser intensity. Various studies [4,5] have shown that carrier densities in excess of $10^{19}-10^{21}\mathrm{cm}^{-3}$ can be produced in a thin surface layer without damaging the material. An electron–hole plasma subsequently decays (radiatively, or nonradiatively by Auger recombination —) with a characteristic time measured in subnanoseconds; the decay time reaches picoseconds in the high density ($n >$

$10^{21}\mathrm{cm}^{-3}$) limit.

Thus, for the femtosecond processes (associated with the femtosecond laser pulses), these decay times are rather larger and the decay dynamics of the plasma can be safely neglected.

We must, of course, remember that the laser fluence must be kept bellow the damage threshold (typically $F_{th} = 1\,\mathrm{kJ/cm}^2$) for the material. Mazur and coworkers [5], in their extensive studies on the interactions of femtosecond laser pulses with GaAs samples, have shown: 1) that above this threshold the crystal is permanently damaged, and 2) near (and below) the threshold, one may observe, depending on the intensity, lattice heating, lattice disordering, or a semiconductor–to–metal transition. In the latter case the crystal reverts to its original state once the pulse is gone. Similar behavior is expected for other direct band gap III–V semiconducting compounds. One concludes, then, that to maintain the fidelity of these systems, the intensity of the laser source must be less than $10^{12}\mathrm{W/cm}^2$.

Barring this not too restrictive a constraint, the semiconductor plasmas, as experimental systems, have several advantages over the gaseous ones: automatic confinement, initial homogeneity, an easy density control over a large range, and the scope for being able to carry out relatively inexpensive and reproducible table–top experiments. The implications for physics as well as for the resulting technology are enormous.

In our recent studies we have explored a few scenarios in which the propagation of femtosecond laser pulses through a semiconductor waveguide could result in large blueshifts [6,7]. In the model experiment suggested in Ref. [6], the plane of an appropriate semiconductor sample (for instance GaAs) is illuminated by a reasonably intense source pulse (with its frequency tuned near the band gap) creating a sudden but almost uniform electron–hole plasma in the wave guide. A low intensity probe pulse, which had been launched prior to the source pulse, finds itself in a time varying medium, and responds by upshifting its frequency.

In the arrangement of Ref. [7], blueshifting of the pulse is entirely self–inflicted; the required electron–hole plasma is generated by the propagating intense laser pulse alone. In Ref. [7], we concentrated on the phenomena associated with the current originating from the free carriers, and neglected the energy loss due to the production of the said carriers. The current–pulse interaction could be justifiably the dominant effect if the energy taken from the pulse for the generation of new free carriers is less than the energy that these carriers gain during their acceleration in the field of the pulse.

We now extend the scope of these preliminary calculations to include several features of the pulse–semiconductor interaction which we had ignored earlier. Most important additional physics will be contained in: 1) the consideration of the detailed dynamics of the pulse at the semiconductor vacuum interfaces, and 2) the pulse energy loss as it creates the electron–hole plasma.

We find that, instead of filling the entire wave guide, the high density plasma created by the pulse resides only in a thin layer at the semiconductor surface. Although the density of plasma may reach above critical values, the complete reflection of the pulse does not take place and a considerable portion of the pulse energy penetrates the sample, leaving behind a high density electron–hole plasma wake. The reflected and the transmitted pulses are both blueshifted. However, since the transmitted pulse continuously loses energy due to the two (one) photon absorption, its intensity goes to zero inside the crystals with widths as short as hundreds of microns; there may be no final transmitted pulse. To obtain blueshifted transmitted pulses with a reasonable fraction of their initial intensity, one must use samples only a few microns wide. One must, of course, accept the fact that a considerable gain in frequency will necessarily cost a considerable loss in intensity.

Preceding considerations suggest the following optimum setting. Almost copropagating with the source pulse, we should launch a probe pulse with a frequency $\hbar\omega_o < E_g/2$ where E_g is the band gap energy . Since the probe is, now, detuned from the main resonances (one and two photon resonance), it will not undergo serious energy losses due to resonant absorption. The propagating probe pulse, sampling the plasma created by the source pulse, will then blueshift without suffering much attenuation over hundreds of microns. The attenuation due to the absorption by the intrinsic free carriers can be neglected for such distances. This process could be repeated if larger blueshifts are needed.

The probe pulse can be further upshifted by the use of a second source pulse. For instance, we could divide the initial intense source into two beams with the second beam illuminating the farther surface just when (by using an appropriate delay line) the probe pulse reaches it. In fact, one can literally cascade the pulse to a desirable high frequency.

To illustrate the proposed scenario, let us work out a one dimensional example. Assuming that all quantities vary along the propagation direction the wave equation for the optical field in an electron–hole plasma can be written as

$$\frac{\partial^2}{\partial t^2} - \frac{c^2}{\epsilon_0}\frac{\partial^2 \boldsymbol{E}}{\partial z^2} + \left(\frac{4\pi e^2}{m_* \epsilon_0}\right)\frac{N}{\gamma}\boldsymbol{E} + \frac{4\pi}{\epsilon_0}\frac{\partial \boldsymbol{J}_I}{\partial t} = 0 \tag{1}$$

where m_* is an affective mass of electrons at the bottom of conduction band, ϵ_0 is the optical constant of the medium, N is the density of electrons produced by the pulse in the conduction band by multiphoton absorption, and J_I is the ionization current related to the dipole moment that is built during the electron hole pair separation. In deriving Eq. (1), we assumed that due to their heavier mass, the hole contribution to the current can be neglected, and the free electrons are born in the conduction band with zero initial velocities, or they are distributed isotropically. The factor $\gamma = (1 + p^2/2m_* E_g)^{1/2}$, where p $(\sim E^2/\omega^2)$ is the quasimomentum of the carriers in the laser field, is a measure of the nonparabolicity in the energy–momentum relation for the conduction electrons. The effect of nonparabolicity in the dispersion can be important for the narrow gap semiconductors [8], while its role in the laser field dynamics for the wider gap semiconductors (i.e. $E_g > 1\,\mathrm{eV}$), is often quite small.

The density of the created plasma is determined by

$$\frac{\partial N}{\partial t} = \sum_{k=1} \frac{\alpha_k I^k}{\hbar \omega_o k} \tag{2}$$

where I is the pulse intensity, ω_0 is the carrier frequency, and α_k are the multiphoton absorption coefficients ($k = 1, 2, ...$). The duration T_L of laser pulse is assumed to be much less than the characteristic response times for recombination.

The ionization current takes a similar form

$$\boldsymbol{J}_I = \frac{c\epsilon_o^{1/2}}{4\pi}\boldsymbol{E}\sum_{k=1}\alpha_k I^{k-1} \tag{3}$$

obtained phenomenologically by averaging the energy conservation law (Pointing theorem), and can be attributed to the field attenuation caused by the energy expended on exciting an electron from the valence band. Indeed, for the monochromatic waves, Eqs. (1)–(3) can be reduced to the well known form

$$\frac{dI}{dz} = -\sum_{k=1}\alpha_k I^k \tag{4}$$

that describes the damping of light due to multiphoton absorption, and is widely used to experimentally determine various absorption coefficients [9].

In what follows we consider the propagation of femtosecond pulses whenever two or one photon processes are dominant. Higher order photon absorption processes are neglected. We must remind ourselves that higher order absorption rates can become comparable with the lower ones for high laser intensities. For instance, for the narrow–gap InSb, this will happen for an intensity $60\,\mathrm{GW/cm}^2$; the number, though, is considerably larger for the majority of semiconductors [9]. Higher order effects can enhance the plasma production rate, and at the same time reducing the absorption length of the pulse. For the intensities under consideration, we may safely neglect effects related with higher order absorption.

Let us introduce dimensionless variables: $t' = \omega_0 t$, $z' = (\epsilon_0^{1/2}\omega_0/c)z$, $N' = N/N_c$, $E' = (c\epsilon_0^{1/2}/4\pi I_m)^{1/2}E$, where $N_c = m_*\epsilon_0\omega_0^2/4\pi e^2$ is the critical plasma density. The electric field is so normalized that $E^{'2}$ represents its magnitude in units of the peak initial intensity $I_m(= |E_m|^2(c\epsilon^{1/2}/4\pi)$. Omitting primes, the dimensionless Eqs. (1)–(3), for a circularly polarized pulse, read as

$$\frac{\partial^2 E}{\partial t^2} - \frac{\partial^2 E}{\partial z^2} + B_1\frac{\partial E}{\partial t} + B_2\frac{\partial |E|^2 E}{\partial t} + \frac{N}{\gamma}E = 0 \tag{5}$$

and

$$\frac{\partial N}{\partial t} = D_1|E|^2 + D_2|E|^4. \tag{6}$$

Here $B_1 = \lambda_0\alpha_1/2\pi\epsilon_0^{1/2}$, $B_2 = (\lambda_0/2\pi\epsilon_0^{1/2})(\alpha_2 I_m)$, $D_1 = \alpha_1\lambda_0^2/4\pi c^2\hbar\omega_0^2 N_c$ and $D_2 = \lambda_0^2\alpha_2 I_m^2/8\pi^2 c^2\hbar\omega_0^2 N_c$, and λ_0 is the vacuum wavelength of the pulse.

We consider the case for normal incidence at the semiconductor surface $z = 0$. The problem will be eventually solved by numerical methods. But to gain some insight into the process of frequency upconversion of the pulse, we first use an analytic geometrical–optics approach. Representing the laser field as $E = A\exp[i\phi]$, and applying the standard geometrical–optics approximations: $\omega A \gg \partial A/\partial t$, $kA \gg \partial A/\partial z$, where $\omega = -\partial\phi/\partial t$, $k = \partial\phi/\partial z$, we obtain after some simple algebra

$$\frac{\partial\omega A^2}{\partial t} + \frac{\partial k A^2}{\partial z} + B_1\omega A^2 + B_2\omega A^4 = 0 \tag{7}$$

$$\frac{\partial\omega^2}{\partial t} + v_g\frac{\partial\omega^2}{\partial z} = D_1 A^2 + D_2 A^4 \tag{8}$$

where $v_g = (1-N/\omega^2)^{1/2}$ is the group velocity of the pulse. The effects of the nonparabolicity of the band are ignored for simplicity.

It follows from Eqs. (7) and (8) that the frequency upshifting of the pulse is accompanied by an attenuation of the field. The energy dissipation take place due to the energy spent in accelerating the newly created particles as well as due to the direct absorption of photons for the initial excitation of particles to the conduction band. For an underdense ($v_g \approx 1$ which can be justified at initial stages) plasma, with one photon absorption dominating ($B_2 = 0 = D_2$), Eqs. (7)–(8) allow the solution ($z' = z$, and $\tau = t - z/v_g$)

$$\omega^3 = \omega_0^3 + \frac{3}{2}\frac{D_1}{B_1}\omega_0 A_0^2(1 - \exp(-B_1 z)) \tag{9}$$

where $\omega_0(= 1)$ and $A_0(= 1)$ are, respectively, the initial frequency and the amplitude of the pulse. One can see that the inclusion of the absorption losses leads to a saturation of the frequency growth with z; the maximum possible frequency shift is $\Delta\omega_m \approx D_1/B_1$. Note that in gaseous plasmas, it is conventional to neglect absorption losses ($B_1 = 0$); the frequency shift, then, comes out (erroneously) to be a monotonically increasing function of z.

It is equally straightforward to solve for the case when the two photon absorption is dominant ($B_1 = 0 = D_1$). The maximum shift comes out to be $\Delta\omega_m \approx D_2/2B_2$, which has an explicit expression

$$\frac{\Delta\omega_m}{\omega_0} = 3.7\frac{m_e}{m_* \epsilon_0^{1/2}} \lambda_0^3 I_m 10^{-5} \tag{10}$$

where the wavelength is measured in microns and the field–intensity in GW/cm^2.

One can see that the maximum frequency shift of the pulse does not depend on the intrinsic rate of the two photon absorption ($\sim \alpha_2$), but it does increase with the applied intensity (the upper limit determined by the damage threshold of the semiconductor) and the wavelength. For GaAs, embedded in a field of intensity $I_m = 1.5 \times 10^{11}$W/cm^2 and wavelength $\lambda_0 = 1.24\mu$m, the maximum upshift is no more than 10% while for the narrow gap semiconductors like InSb, it can be 60% for $\lambda = 10\mu$m and $I_m = 1$ GW/cm^2.

The results contained in Eq. (10) have been derived under the geometrical–optics approximation, along with the assumption that the laser pulse propagates with a constant group velocity in an underdense plasma. Thus the condition (10) can give us just an upper cap for the frequency shift. Although the system of Eqs. (7), (8) can be solved numerically for more details, we do not do it here. Instead, we shall solve the full wave equations, since the geometric optics can not account for the fast variation of the pulse envelope; it also cannot deal with the reflection of the pulse from the boundary, a phenomenon of critical importance since it is the reflection that seems to be most important factor limiting the value of upshifting.

We have performed numerical simulations of the dynamics of a laser pulse interacting with an undoped sample of the narrow band–gap semiconductor InSb at room temperature ($E_g = 0.175$ eV). This semiconductor is established to have the largest two photon absorption coefficient, $\alpha_2 \approx 8$ cm/MW at the laser wavelength $\lambda_0 = 9.6\mu$m [10]; its conduction band electrons also have the smallest effective mass, $m_* = 0.0133 m_e$. According to the reasons given above, InSb should be a good candidate to induce considerable frequency blueshifting in the mid–infrared pulses.

The chosen laser pulse, with a wavelength $\lambda_0 = 9.6\mu\,$m, has a peak intensity $I_m = 0.5\,\mathrm{GW/cm}^2$, and its duration is $T_{\mathrm{FWHM}} = 130\,$fs, where T_{FWHM} is the full width at the half maximum. For this pulse, one photon absorption is energetically forbidden and just the two photon absorption is responsible for creating the electron–hole plasma. The laser absorption length is $l_{\mathrm{abs}} \approx 3\mu\,$m, and critical density of the plasma is $n_c = 2.6 \times 10^{18}\mathrm{cm}^{-3}$. The pump fluence is arranged to be $F \approx 10^{-4} J/\mathrm{cm}^2$ which is much below the damage threshold.

Results of numerical simulations of Eqs. (5) and (6), for the parameters presented above, are given in Fig. 1. The laser pulse is incident on the plane boundary of the sample, and has a profile: $E = \sin(t-z)\exp[-(t-z)^2/2T^2]$ with $T = 14$. The pulse, crossing the semiconductor boundary, creates a high density plasma and whenever the density becomes larger than the critical, the pulse suffers a strong reflection. However, since the above critical density is reached just in a very thin layer near the semiconductor surface ($\sim 2\mu\,$m), a complete reflection of the pulse energy does not take place; a considerable portion of the pulse energy penetrates inside the sample. The nonparabolicity factor has a value $\gamma = 2.4$ at the boundary, and like in the case of relativistic plasmas, reduces the effective plasma frequency by a factor γ thus helping the pulse to penetrate the sample. In Fig. 2 we plot the spectral composition of the reflected and the transmitted pulses. For both these parts, the spectrum is blueshifted. Note that for the reflected pulse, the blueshift is small ($\sim 1\%$), while for the transmitted pulse it is 10%. The transmitted pulse undergoes two photon absorption giving rise to the further creation of the electron–hole plasma in the bulk, but the rate of plasma creation decreases considerably. For crystal samples with widths of several hundreds of microns, the transmitted pulse will be attenuated before it reaches the end surface. Thus, strong laser pulses tuned to the photon absorption resonances can undergo blueshifting in a self created electron–hole plasma. However to get measurable outputs, the thickness of the semiconductor crystal must be limited to the tenths of a micron.

Next we simulate the dynamics of the probe pulse launched in the sample along with the source (pump) pulse. The almost copropagating pump and probe are incident on the crystal surface. The probe pulse is assumed to be outside the range of the photon absorption resonances ($\lambda_p > 14\mu$m); its intensity is also a few orders of magnitude smaller than the intensity of the pump. In this setting, the pump, which is tuned to a given photon resonance, creates the electron–hole plasma (Figs. 1), while the probe is free from this duty. In Fig. 3, we plot the evolution of a probe pulse which, in vacuum, copropagates with the intense pump (the pump pulse is not shown in the picture). The frequency of the probe is chosen to be $\omega = 0.9$ (i.e. $\lambda_0 \approx 10.6\mu$m), while its temporal duration is the same as that of the pump. When the pulses cross the left (front) surface of the sample, the pump begins the process of plasma creation. The probe, at the same time, encounters fast changes in the plasma density. The part of the pulse that confronts the critical (for the probe) plasma density $n_c^{\rm prob} = 0.8 n_c^{\rm pump}$, gets reflected. The transmitted pulse that is blueshifted by 11%, does not suffer losses due to resonant absorption, and can travel, without considerable attenuation, in the bulk of the sample. Free carrier absorption will eventually damp the pulse, but for the crystal widths (i.e. few hundreds of microns) we are concerned with, this effect should not be important. In Fig. 3, we also model a situation when the output surface can be used to enhance the frequency upconversion efficiency. A second pump pulse enters this surface of the sample, precisely when the probe pulse is just ready to leave it. One can see that the probe will be reflected again, and a part of it is transmitted out to the vacuum with an additional secondary blueshifting. The spectrum of the twice upshifted transmitted pulse ($2p$), in relation to the vacuum pulse, is presented in Fig. 4. The total frequency shift, now, has reached 30%. The shape of the spectrum indicates that self–phase modulation, and spectral broadening also takes place. This effect is due to the group velocity dispersion which modulates the pulse phase in the thin surface layer of the highly dense plasma. Note that the part of the probe pulse, reflected from the second boundary, appears to be trapped between plasma "mirrors." Since these mirrors may, subsequently, decay in subnanoseconds, the trapped pulse has enough time to conduct few oscillation between the surfaces of the sample. However, each time the trapped pulse reaches the boundaries, some of it will be transmitted, and only a part will be reflected back into the sample. Thus, the upshifted pulses will operate in some kind of a "machine–gun" mode till the pulse is either exhausted, or attenuated due to the free carrier absorption.

The largest blueshifting is expected for narrow–gap semiconductors like InSb for which the two photon absorption is dominant in the range $7.3\mu m < \lambda_0 < 14.6\mu$m. At these wavelengths, however, the generation of femtosecond laser pulses, though possible (see for instance Corkum in Ref. [4]), it still requires considerable effort. On the other hand, femtosecond lasers operating at $\sim 1\mu$m wavelengths are readily available. Such lasers can be used for wider–gap semiconductors such as GaAs. For wider–gap semiconductors, as remarked earlier, higher laser intensities will be needed to duplicate the processes described above; the physics is essentially the same.

We have investigated the problem of the propagation of ultrashort laser pulses entering a semiconductor from vacuum. We find that frequency upconversion results whenever the laser pulse meets an electron hole–plasma, whose density is changing in time. It was shown that a strong laser pulse, tuned to a band–gap resonance, blueshifts in the self–created highly dense plasma. However it may be difficult to observe the blueshifted (self–inflicted) transmitted pulse since the energy losses due to the plasma creation greatly reduce the pulse intensity (and hence the efficiency of upshifting as well) unless the width of the semiconductor sample is in tenths of microns. The situation is entirely different for a probe pulse which may be launched along with the intense pump pulse (the source of plasma creation), and which is detuned from the absorption resonance. An equally effective method for an observable blueshift is to choose the probe pulse intensity to be a few orders of magnitude smaller than the pump (at these intensities the absorption effects are negligible); the frequency upshifting is achieved due to the nonstationary plasma created by the source pulse. To further enhance the blueshifting, we suggest a cascading upshifting mechanism in which the probe pulse can be upshifted using several boundaries and appropriate sources. Every time the probe pulse meets a time varying plasma density (near the boundary) it undergoes reflection as well as transmission; both the transmitted and the reflected parts are blueshifted. Such a system creates two interesting entities: a pulse trapped inside the boundaries, and a transmitted (out of either boundary) "machine–gun" of upshifted pulses. We believe that the proposed phenomena can be be easily demonstrated in a variety of existing semiconductor optics experiments based on the pump–probe techniques.

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FIGURES

FIG. 1. The electric field intensity E (normalized to its initial value) versus z for several times. One can see the splitting into a reflected and a transmitted pulse due to the self–created plasma layer of above–critical density at the vacuum–semiconductor boundary. The density profile is also displayed.

FIG. 2. The spectral content of the reflected and the transmitted pulses displayed in Fig. 1. The incident pulse, peaking at $\omega = 1$, is shown for reference. The frequency is normalized to the initial frequency of the pump.

FIG. 3. The normalized electric field of the probe pulse versus z and t. A part of the pulse is trapped between the plasma mirrors at the boundaries. We can also see the first installment of the "machine–gun" of the transmitted pulses. V stands for vacuum and S for the semiconductor.

FIG. 4. The spectral contents of the once $(1p)$, and twice $(2p)$ upshifted (probe pulse. the incident probe pulse is peaked at $\omega = 0.9$. The frequency is still normalized to the initial frequency of the pump pulse.