Photon accelerator: Large blueshifting of femtosecond pulses in semiconductors

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The availability of relatively high intensity ($I>10^9$ W cm^{-2}) [but moderate ($\sim$nJ) total energy], femtosecond laser pulses with wavelengths ranging from the ultraviolet to the midinfrared has opened the doors for a serious investigation of the nonlinear optical properties of matter on ultrashort time scales in a new parameter regime. Even small intensity-dependent nonlinearities can begin to play a major role in the overall electrodynamics, and in determining the fate of the propagating pulse. It is shown that a femtosecond pulse propagating near a two-photon transition in a semiconductor waveguide can undergo a large blueshift. [S1050-2947(97)06112-X]

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Collisionless collective phenomena [1,2], induced by femtosecond laser pulses [3] in semiconductor plasmas, are emerging as a new and exciting area of research. These phenomena, taking place on time scales much shorter than collision times, reproduce in semiconductor plasmas a variety of physical processes normally associated with hot gaseous plasmas. These fast processes, therefore, are likely to become a meeting ground for the disciplines of semiconductor physics, nonlinear optics, and plasma physics. It is expected that rich scientific dividends may be in the offing.

In this paper we study the propagation of a short femtosecond laser pulse in a semiconductor when two-photon absorption (TPA) is dominant. TPA is a nonlinear resonant process in which an interband transition is induced by the simultaneous absorption of a pair of photons. For photon energies in excess of half the band-gap energy, TPA generates free carriers which contribute to nonlinear refraction. In conventional approaches, the TPA contribution is limited to the imaginary part of the third-order susceptibility $\hat{\chi}^{(3)}$ which determines the third-order induced polarization $P^{(3)}$ in the wave equation [4]

$$2ik\left(\frac{\partial}{\partial x}+\frac{1}{v_g}\frac{\partial}{\partial t}\right)E=-\omega^2 P^{(3)}(E), \tag{1}$$

where $k=n_0\omega/c$, n_0 is the background refractive index, v_g is the pulse group velocity in the medium, and E is the slowly varying envelope of the pulse electric field. Equation (1), derived by invoking the slowly-varying-envelope approximation (SVEA), is an adequate description only of those processes which happen at time scales much greater than the wave period. For instance, the nonlinear refractive and absorption properties described by $P^{(3)}$, and the effects induced by them, such as self-phase modulation (SPM) and nonlinear damping due to TPA, will manifest themselves after the pulse has traveled several pulse lengths through the medium. Equation (1) also neglects the group-velocity dispersion (GVD). These considerations limit the applicability of Eq. (1) to situations for which the TPA-generated excess carrier plasma density is far below the critical value at which the laser frequency is equal to the carrier plasma frequency.

New models, designed to simulate physics on time scales comparable to or shorter than the inverse of the pulse width, are thus needed for an adequate description of the fast interaction between semiconductors and high-intensity femtosecond laser pulses. An understanding of these processes will help in the exploitation of the large and extremely fast optical nonlinearities in semiconductors which are so attractive for optical devices. The TPA model developed in this paper also reveals several interesting and specific aspects of this interaction which are not found in the vast extant literature on the nonlinear optical properties of semiconductors [5,6].

Concentrating on TPA, we pick the incident laser photon energy $\hbar\omega$ to lie in the range $E_g/2<\hbar\omega<E_g$, where E_g is the band-gap energy. The laser pulse is tuned near the two-photon resonance well below the lowest one-photon transition threshold. Propagating through the semiconductor, the pulse, via the two-photon absorption, places a large number of mobile electrons in the conduction band. The density N of this newly created plasma is determined by

$$\frac{\partial N}{\partial t}=\alpha I^2, \tag{2}$$

where I is the laser intensity inside the semiconductor, $\alpha=\beta_2/2\hbar\omega$, and β_2 is the two-photon absorption coefficient. The laser pulse duration τ_L is assumed to be much less than the characteristic response times for recombination and diffusion; the latter processes are therefore neglected. It should be noted that the carrier recombination (both radiative and nonradiative) takes place typically on nanosecond or even longer time scales [4]. These times are much longer than the time scales of the duration of the processes described by our model. However, radiative recombination may be enhanced

in the presence of an intense radiation field. This complicated effect is left for a future investigation.

Traditionally, TPA is taken to be a source for generating free charge carriers, even a large number of them. The energy expended in this process results in an attenuation of the laser pulse. In our model, however, we stress a complementary aspect of the interaction, the acceleration of the newly created free carriers and the resulting currents (fastly varying) induced in the semiconductor. This current-pulse interaction could be the dominant effect if the energy taken from the pulse for the generation of a new free carrier (via TPA) is less than the energy which this new carrier gains by being accelerated in the field of the pulse. This is the principal point which distinguishes our approach from previous work. In fact, we will concentrate entirely on the phenomena associated with the current carried by the free carriers and neglect other processes including the energy loss due to the production of the said carriers.

Let us now examine if our model is relevant to any realistic physical situation. The following example clearly demonstrates that the answer is in the affirmative: In order to generate a free carrier via TPA in InSb we need 0.234 eV. This very carrier, when accelerated in the pulse field, gains from the pulse an energy $\mathcal{E}=(eE)^2/(2m_e^*\omega^2)$, where E is the peak amplitude of the laser electric field, m_e^* is the effective electron mass, ω is the laser frequency. The energy $\mathcal{E}$ is related to the peak intensity and the wavelength of a circularly polarized laser pulse by the formula $\mathcal{E}(\mathrm{eV})=7.02\times10^{-12}I(\mathrm{W/cm^2})\lambda^2(\mu\mathrm{m})$. With $I=3.3\times10^9$ W/cm^2 at $\lambda=10\,\mu$m, this formula gives $\mathcal{E}=2.32$ eV, which is almost ten times greater than the energy needed (by TPA) to generate one free carrier. It is also clear, that for a sufficiently large laser intensity, such a situation will pertain for a wide variety of semiconductors.

The preceding discussion suggests that for appropriately chosen systems, we can safely neglect the TPA contribution to the imaginary part of the effective third-order polarization, and concentrate only on the fast nonlinear current generated by TPA. Thus the wave equation for the optical field of the laser pulse (assuming that all quantities vary along the pulse propagation direction, i.e., along the waveguide) can be written as

$$\frac{\partial^2\mathbf{E}}{\partial t^2}-c_*^2\frac{\partial^2\mathbf{E}}{\partial x^2}=-\frac{4\pi}{\varepsilon}\frac{\partial\mathbf{j}}{\partial t}. \tag{3}$$

Here $c_*=c/\sqrt{\varepsilon}$, c is the speed of light in vacuum, ε is the optical dielectric constant of the medium, and $\mathbf{j}(x,t)$ is the pulse-induced current primarily carried by the electrons created by TPA. To complete the model, we need a ''constituent equation'' connecting this induced current with the electric field of the laser pulse.

Let $\Delta N(x,t')=[\partial N(x,t')/\partial t']dt'$ denote the small group of free carriers generated at time t'. After being accelerated in the field of the laser pulse, at time t, this group will have acquired a velocity (starting from rest)

$$\mathbf{v}(x,t)=-\frac{e}{m_e^*}\int_{t'}^{t}dt''\mathbf{E}(x,t''). \tag{4}$$

Integrating $\Delta\mathbf{j}=-e\Delta N(x,t')\mathbf{v}(x,t)$, the total induced current will be

$$\mathbf{j}(x,t)=\frac{e^2}{m_e^*}\int_{-\infty}^{t}dt'\,\frac{\partial N(x,t')}{\partial t'}\int_{t'}^{t}dt''\mathbf{E}(x,t''). \tag{5}$$

Equation (5), connecting the pulse driven current of free carriers with the pulse electric field, forms the essence of our model. This induced nonlinear current is of fifth order in the field amplitude. Substituting Eq. (5) in Eq. (3) we obtain the following wave equation governing the pulse propagation:

$$\frac{\partial^2\mathbf{E}}{\partial t^2}-c_*^2\frac{\partial^2\mathbf{E}}{\partial x^2}=-\frac{4\pi e^2}{m_e^*\varepsilon}N\mathbf{E}. \tag{6}$$

Equations (2) and (6) form a set of coupled nonlinear equations describing the dynamics of a short intense laser pulse in a semiconductor when the pulse first generates large densities of free charge carriers through the resonant TPA process and then accelerates them to produce a large nonlinear current. Needless to say, the characteristics of the pulse will be profoundly changed by this induced current. This current will strongly affect the dispersion properties of the medium, and will also cause pulse damping.

This damping can be physically understood by analyzing the response of the ''newly born'' free carriers to the pulse. Born at zero velocity, these carriers are accelerated in the pulse field, and thus gain considerable amount of energy from the pulse. Thus TPA, in our model, plays a twofold role: first it generates a plasma of excess carriers, which changes the refractive properties of the medium and, second, it contributes to the ''absorbing'' properties of the medium through the same term. This absorption (due to the energy expended in acceleration) is quite different from the conventional absorption (due to the energy expended in putting electrons in the conduction band) associated with TPA.

Going back to Eq. (2), and noting that $I\propto|\mathbf{E}|^2$, it seems that the TPA-induced free carrier nonlinearity (proportional to $N\mathbf{E}$) is equivalent to a nonlinearity described by a $\hat{\chi}^{(5)}$ optical susceptibility. Note that the free carrier absorption is neglected in our model. Let us make a digression to justify this assumption.

At high carrier densities ($\geqslant10^{17}$ cm^{-3}), scattering time of free carriers due to carrier-carrier interactions is of the order of $t_0=100$ fs and scattering times for phonon and ionized impurities are of the order of a few picoseconds. However, in a high-intensity radiation field, the interaction time is modified to $t=t_0(\mathcal{E}/\mathcal{E}_0)^{s/2}$, where $\mathcal{E}$ is the total average energy of the carriers (i.e., $\mathcal{E}=\mathcal{E}+\mathcal{E}_0$), and $\mathcal{E}_0$ is the average energy in the absence of an external field. The exponent $s=1$ if the optical phonon scattering is dominant, and $s=3$ for the momentum-loss scattering of the carriers on the ionized impurities or on other carriers [7]. For problems of interest to this work, the electrons, in the intense radiation field, acquire energy of the order of 1 eV. For these energies, the shortest relaxation time is of the order of several picoseconds. Thus our model could only be applied to fast processes which take place on shorter time scales. Fortunately, subpicosecond times are precisely the times of interest to us.

In order to gain some insight into the physical processes, let us consider first a model problem of a square-shaped circularly polarized wave packet

$$\mathbf{E}=\tfrac{1}{2}(\hat{\mathbf{y}}+i\hat{\mathbf{z}})e^{i\varphi}E_0+\text{c.c.} \tag{7}$$

traveling through the semiconductor. Here $E_0=E_0(x,t)$ is a real-valued amplitude, and $\varphi=\varphi(x,t)$ is the phase of the circularly polarized laser pulse.

Introducing the new variables $\xi=x-c_*t$, $\tau=t$, we can reduce Eqs. (2)–(6) to

$$\frac{\partial^2 E}{\partial\tau^2}-2c_*\frac{\partial^2 E}{\partial\xi\partial\tau}=-\frac{4\pi e^2}{m_e^*\varepsilon}NE, \tag{8}$$

$$\frac{\partial N}{\partial\tau}-c_*\frac{\partial N}{\partial\xi}=\alpha_I E_0^4, \tag{9}$$

where $E=E_0e^{i\varphi}$ is the complex amplitude.

Consider a right-going square-shaped laser pulse with amplitude E_0 and length $l=c_*\tau_L$,

$$E_0(\xi)=\begin{cases}E_0=\text{const} & \text{if } -l<\xi<0\\ 0 & \text{otherwise.}\end{cases} \tag{10}$$

For quasistationary propagation of the pulse ($c_*\partial/\partial\xi \gg \partial/\partial\tau$), Eqs. (8) and (9) become

$$-2c_*\frac{\partial^2}{\partial\xi\partial\tau}e^{i\varphi}=-\frac{4\pi e^2}{m_e^*\varepsilon}Ne^{i\varphi}, \tag{11}$$

$$-c_*\frac{\partial N}{\partial\xi}=\alpha_I E_0^4. \tag{12}$$

Integrating Eq. (12) we obtain the carrier density excited by the pulse,

$$N(\xi)=\begin{cases}N_0=\text{const}, & \xi>0\\ N_0-(\alpha_I/c_*)E_0^4\xi, & -l<\xi<0\\ N_0+(\alpha_I/c_*)E_0^4 l, & \xi>-l\end{cases} \tag{13}$$

which, coupled with Eq. (11), leads to the following equation governing the nonlinear evolution of the phase $\varphi(\xi,t)$ (inside the pulse):

$$i\frac{\partial^2\varphi}{\partial\xi\partial\tau}-\frac{\partial\varphi}{\partial\xi}\frac{\partial\varphi}{\partial\tau}=\frac{\omega_{p0}^2}{2c_*}\left(1-\frac{\alpha_I}{N_0c_*}E_0^4\xi\right), \tag{14}$$

where $\omega_{p0}=(4\pi e^2N_0/m_e^*\varepsilon)^{1/2}$ is the carrier plasma frequency corresponding to the carrier density in front of the pulse, i.e., the value of the ambient uniform density before the pulse enters the semiconductor. The ansatz

$$\varphi(\xi,\tau)=\frac{\omega(\tau)}{c_*}\xi+\psi(\tau) \tag{15}$$

leads to the following exact solution of the nonlinear Eq. (14):

$$\omega(\tau)=\omega_0\left(1+\frac{\omega_{p0}^2}{\omega_0^2}\frac{\alpha_I}{N_0}E_0^4\tau\right)^{1/2}, \tag{16}$$

$$\psi(\tau)=i\ln\frac{\omega}{\omega_0}-\frac{N_0}{\alpha_I E_0^4}(\omega-\omega_0)+\psi_0, \tag{17}$$

where ω_0 and ψ_0 are the initial frequency and phase of the pulse. The complex amplitude E, then, becomes

$$E=E_0\exp(i\varphi)=\frac{\omega_0}{\omega}E_0\exp\left(i\frac{\omega(\tau)}{c_*}\xi+i\,\mathrm{Re}[\psi(\tau)]\right). \tag{18}$$

Equations (16) and (18) display two important consequences of the nonlinear dynamics of the pulse propagation: (1) Equation (16) indicates that the pulse is upshifted in frequency, and this upshift is continuous in time, and (2) Eq. (18) clearly shows that the pulse amplitude damps in time.

Even this simplified model shows that the nonlinear self-consistent interaction of the pulse with the plasma of excess carriers may provide a tunable source of high-frequency radiation.

Note that our quasistationary approximation is valid when $\omega^2\gg\omega_p^2$, i.e., when the carrier plasma is transparent to the laser pulse. In this case, according to Eq. (16), large frequency shifts will require large interaction times or, equivalently, the pulse propagation over large distances inside the semiconductor waveguide. For such large traversal times, collisions and other complicating effects will render the simple analytical calculation quite useless. In order to deal with the physics of large frequency shifts which will require ω_p to be of the same order or even greater than ω, we must work directly with Eqs. (2) and (6).

In many real situations even when a pulse enters a semiconductor waveguide with initially underdense carrier plasmas, it can boost the carrier plasma density to near-critical or even over-critical values by generating excess carriers via the TPA process. In this case a much ''stronger and faster interaction'' between the pulse and the generated plasmas takes place. We carried out a numerical simulation of this process described by the nonlinear set of equations (2) and (6). The simulation reveals a number of new features of this interaction; it also provides reasonable arguments for explaining the nature of the spectrum observed in experiments.

Notice that we must solve three coupled nonlinear equations: Eq. (6) representing two full wave equations for the two components of the laser field, and Eq. (2) for the carrier density. We use a finite difference scheme, second order in both time and space. The results for the pulse propagation through a waveguide of the well-known III-V semiconductor InSb will be presented below.

The simulation employed an initially Gaussian (for $|\mathbf{E}|$) pulse with $\tau_{\mathrm{FWHM}}=200$ fs (full width at half maximum) and wavelength $\lambda=10.6\ \mu$m. It propagates from left to right in the InSb waveguide in which the free carrier density prior to the pulse is $N_0=0.01N_{\mathrm{cr}}$, where N_{cr} is the carrier density at $\omega=\omega_p$, the carrier plasma frequency. For the pulse tuned at $\lambda=10.6\ \mu$m, $N_{\mathrm{cr}}=2.37\times10^{18}\ \mathrm{cm}^{-3}$.

Although we solve the full wave equations for E_y and E_z components of the circularly polarized laser field, we display only the pulse envelope $|\mathbf{E}|$ versus x in Fig. 1. The envelope is normalized to the value corresponding to the initial peak

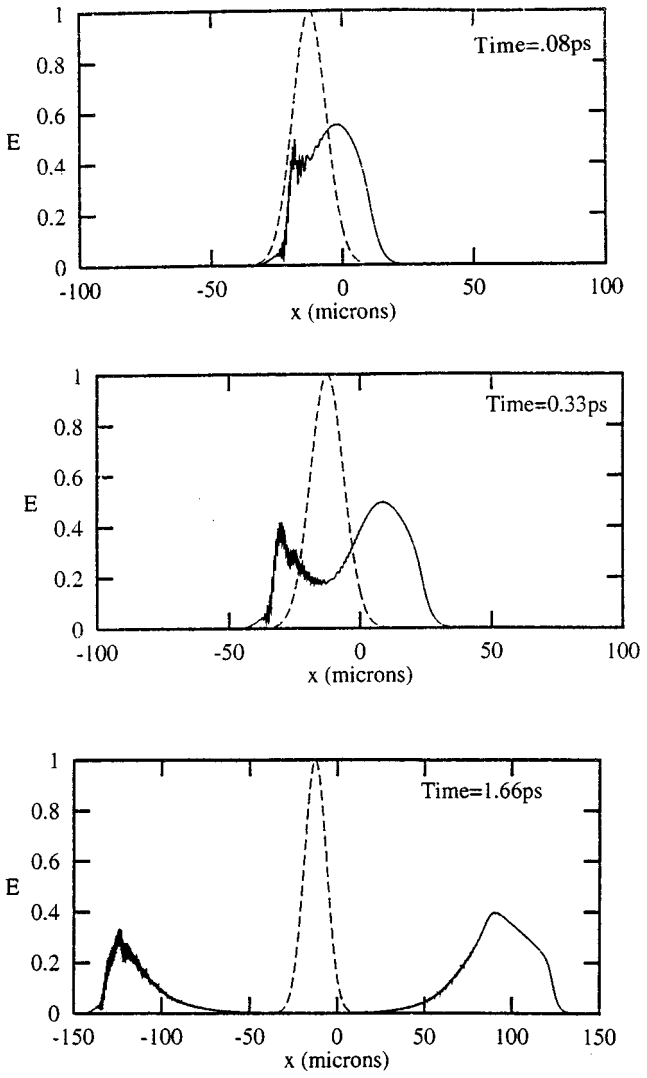

FIG. 1. Plots of $|E|=(E_x^2+E_y^2)^{1/2}$ vs x at different times for an initially (---) Gaussian pulse with wavelength $\lambda=10.6\,\mu$m, and width 200 fs.

intensity $I_0=10^9$ W/cm^2. In Fig. 2 we show the spectral content of the wave at the initial and final times of the simulation.

Figure 1(a) reveals that after a rather short period, $t=0.08$ ps, the pulse amplitude is significantly damped, and the envelope profile has developed strong short scale modulations. In this as well as later pictures, the dashed line shows the initial location of the pulse. In Fig. 1(b) (0.33 ps) we see that the pulse amplitude has damped to less than half its initial value. At the same time we observe that the pulse has developed a double-peaked structure; the initial right-going pulse is ''split'' into two: a transmitted and a reflected pulse. Later parts of the pulse are reflected from the region where the initial pulse, via TPA, created a dense enough carrier plasma. In Fig. 1(c), the trend shown in Fig. 1(b) continues, the reflected and the transmitted waves have moved further apart, and slow amplitude damping continues.

These figures show that one can distinguish at least two stages in the pulse dynamics. The first stage lasts up to 0.33 ps. At this stage the pulse evolves as a whole and the dominant process is its amplitude damping. This transient period

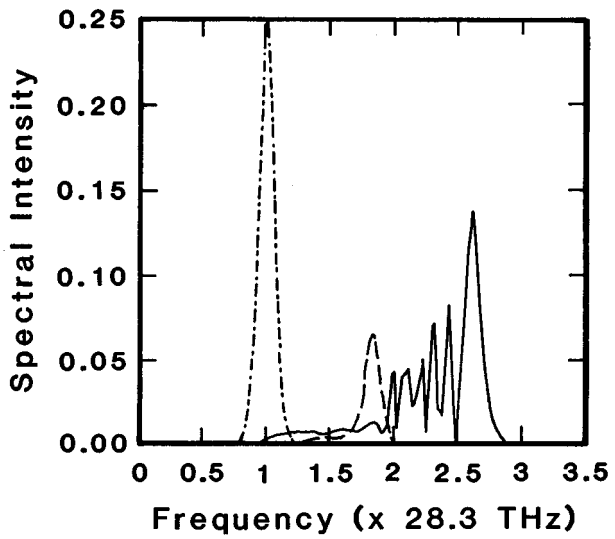

FIG. 2. Spectral content of the initial (-·--·), reflected (---), and transmitted (—) pulses. The central frequency of the original pulse is taken to be unity in the graph.

ends with the formation of the transmitted and reflected pulses. Both branches have clearly pronounced sharp leading edges and relatively stretched trailing edges. The dynamics of these pulses show the strong GVD effect which, in turn, is enhanced by nonlinear changes in the refractive index. These dynamics in the space-time domain cannot be completely understood without analysis in the frequency domain.

The spectral analysis, Fig. 2, shows that both the transmitted and the reflected pulses are significantly upshifted in frequency. The transmitted pulse is upshifted more than the reflected one. Its spectrum is represented by a set of almost evenly spaced peaks in the frequency interval $2\nu_i<\nu<2.6\nu_i$, where $\nu_i=28.3$ THz is the initial laser frequency. The biggest peak is centered at $\nu=2.6\nu_i$. The spectrum of the reflected pulse is shown by the dashed line. It consists of a single well-pronounced peak centered at $1.8\nu_i$.

Similar results are obtained for several other semiconductors; GaAs, for example.

The prediction of this spectacular frequency upshift (photon acceleration) caused by the strong and fast nonlinear interaction of the semiconductors with ultrashort laser pulses is the principal result of this paper.

Much of the blueshift exhibited in Fig. 2 seems to take place in the first stage of the pulse dynamics when the transmitted and the reflected pulses are not yet well separated and are still strongly influenced by the dramatic and rapid changes in the newly born excess carrier plasma. The sudden, almost uniform (throughout the width of the short propagating pulse), and large increase in plasma density induces a sudden and large temporal variation of the index of refraction of the background medium. The pulse frequency must upshift in order to satisfy the new dispersion relation defined by the new index of refraction. This simple and qualitative explanation constitutes the essential physics unraveled in our numerical modeling. The amount of blueshift is determined by the pulse intensity as well as the traversal time in the cavity; the system is tunable, i.e., an appropriate choice of parameters can lead to the desired blueshift. Such

frequency blueshifts are also observed in short intense pulses converting an initially neutral gas into a dense plasma via multiphoton ionization [8].

What does the frequency upshift do to the subsequent pulse dynamics? First of all, it means that the frequency is no longer tuned near the two-photon resonance, hence the TPA is automatically ''switched off.'' Thus the transmitted and the reflected pulses revert back to the linear mode of propagation. But unlike the initial pulse propagation at early stages when it enters the waveguide, the transmitted and the reflected pulses undergo enhanced influence of the positive GVD effect; this influences the final shapes of the pulses according to their spectral content.

We would like to emphasize that our numerical modeling employs the full wave equation instead of the reduced envelope equation. The simulation results clearly show that the envelope equations based on SVEA fail to adequately describe not only the fast processes that occur on short time scales of a wave period but also processes peculiar to a strongly inhomogeneous medium. For instance, one cannot describe the generation and further dynamics of the backward-going reflected waves within the framework of the SVEA, i.e., Eq. (1).

For narrow-gap semiconductors like InSb, the laser wavelength, for which TPA is dominant, should be in the range $7.3\ \mu\text{m}<\lambda<14.6\ \mu\text{m}$. At these wavelengths the generation of femtosecond laser pulses pushes the laser technology to its current limit (see Seifert *et al.* in Ref. [3]). On the other hand, femtosecond lasers operating at $\sim 1\ \mu$m wavelengths are widely accessible. Such lasers can be used for wider-gap semiconductors such as GaAs. For GaAs, however, the TPA coefficient is several orders of magnitude smaller than for InSb. Consequently higher laser intensities will be needed to duplicate the processes described above.

We have just demonstrated that in a semiconductor a sufficiently intense laser pulse tuned near a two-photon resonance first generates a large excess of free carriers (via TPA), and then accelerates them to create a large time-varying current. For a proper description of this process, we have introduced a new model which highlights the crucial role of the nonlinear current in determining the subsequent pulse behavior. The nonlinear interaction of the propagating pulse with this self-generated current induces (among other things) a spectacular blueshift in the pulse frequency; the semiconductor is turned into an efficient photon accelerator. The harnessing of this process for constructing a tunable radiation source is an obvious application. The emerging pulse, in addition, may serve as a useful diagnostic as it carries valuable information about the macroscopic characteristics of the medium, in interaction with which it was born.

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