Generalization of the Kerr effect for high intensity, ultrashort laser pulses

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Abstract. We have investigated the nonlinear susceptibility of atoms induced by high intensity, ultrashort laser pulses using a numerical solution of the time-dependent Schrödinger equation. We found that the instantaneous nonlinear susceptibility becomes saturated at high intensity. We also found that the saturation is closely linked to depletion of the ground state. Based on the numerical results, a simple model that generalizes the nonlinear susceptibility of atoms for high intensity, ultrashort laser pulses is proposed. We also investigated the ionization-induced dipole moment and found that the amplitude of the dipole moment induced by an ionized electron is, in general, smaller than that induced by a free-electron, and is attributable to a residual interaction between the ionized-electron and its parent ion.
1. Introduction

The development of high-intensity lasers has led to the possibility of investigating a wealth of nonlinear optical phenomena. A comprehensive understanding of the behavior of nonlinear susceptibility is important in the field of nonlinear optics; e.g. to quantitatively describe the dynamics of laser pulses propagating in nonlinear media and related phenomena [1]–[3], the propagation of high harmonic waves (see e.g. [4, 5]), the generation of sub-10 femtosecond pulses through spectral broadening of pulses [6, 7], as well as aerosol detection through white light filament analysis [8]. The nonlinear susceptibility of the medium enters into the dynamics of wave propagation via the total refractive index \( n \), given by \( n = \sqrt{1 + 4\pi \chi^{(T)}} \). Here, \( \chi^{(T)} \) is the total susceptibility comprised of the the instantaneous-, delayed-, and plasma-induced high-order nonlinear-susceptibility: \( \chi^{(T)} = \chi_{\text{Inst}}^{(T)} + \chi_{\text{Delay}}^{(T)} + \chi_{\text{Plasma}}^{(T)} \). We limit ourselves to a system with rotational symmetry, such that \( \chi_{\text{Delay}}^{(T)} = 0 \).

In the low-intensity regime, where the plasma has not yet been formed, the nonlinear responses are instantaneous and can be truncated to the third order: \( \chi^{(T)} = \chi_{\text{Inst}}^{(1)} + \chi_{\text{Inst}}^{(3)} I \), where \( I \) is the instantaneous intensity [9]. Recently, there have been several reports indicating the presence of high-order terms, hypothetically assumed to have negative signs, so that they saturate the nonlinear susceptibility (e.g. [3, 10]). Filamentation of the laser beam over distances much larger than its diffraction length, or self pulse compression are explicable if the nonlinearity of the medium is subject to saturation or there is a plasma-induced negative nonlinear refractive index [11]. Since both saturated instantaneous nonlinear susceptibility and plasma-induced higher order terms occur at high intensity, it is usually difficult to distinguish between them. However, one can distinguish between the effects resulting from the saturation of the nonlinear susceptibility and plasma-induced high-order nonlinear susceptibility, since in the latter case, the temporal and spectral profiles are found to be asymmetric [12]. It has been recently proposed that saturation of the instantaneous nonlinear susceptibility can be regarded qualitatively as the limiting value of the total nonlinear susceptibility when the ionization is close to zero [13]. The quantitative amount, however, is difficult to determine since this requires an extremely precise numerical procedure [14]. It is therefore important to identify the main mechanism that is responsible for saturation.

In this paper, we investigate the nonlinear susceptibility for high-intensity, ultrashort laser pulses using a numerical solution of the time-dependent Schrödinger equation (TDSE). By separating the numerical wavepacket into wavepackets in the ground state, bound-excited states and the continuum states, the associated dipole moments can be calculated. We found that the dipole moment induced by the electronic transition from the ground state to either
the excited or continuum state is instantaneous, but when depletion occurs, the associated susceptibility becomes time-dependent. We found that the saturation of the instantaneous nonlinear susceptibility is mainly caused by the depletion of the electronic ground state. Based on the simulation results, a simple model that can be used to predict saturation is proposed. We further analyze the dipole moments associated with ionization and compare these with those induced by free-electrons.

2. Method

The numerical method underlying the simulation is based on the TDSE that can be written as (we use the Hartree atomic units $\hbar = m = e = 1$) [15]:

$$i \frac{\partial}{\partial t} \psi(\mathbf{r}, t) = \left[ -\frac{\nabla^2}{2} + V(r) + \mathbf{E}(t) \cdot \mathbf{r} \right] \psi(\mathbf{r}, t),$$

where $\mathbf{E}(t) = \mathbf{k} \mathcal{E}(t) \sin(\omega t)$ is the electric field, given in the dipole approximation, and $\mathcal{E}(t)$ is the electric field envelope. We assume that the electric field is linearly polarized along the $z$-axis, such that only the magnetic quantum number $m = 0$ is required. Equation (1) is solved by expanding the wavefunction in a set of spherical basis functions $\psi(\mathbf{r}, t) = \sum_l R_l(r, t) Y_{l,m}(\theta, \varphi)$.

The resulting coupled set of partial differential equations is integrated numerically using the split operator method, described elsewhere [16].

Having obtained the solution at time $t$, the wavepacket $\psi(\mathbf{r}, t)$ is decomposed into terms belonging to the ground state, bound-exciton wavepackets and a continuum wavepacket, according to:

$$\psi_g(\mathbf{r}, t) = \langle \phi_{1s}(\mathbf{r}) | \psi(\mathbf{r}, t) \rangle \phi_{1s}(\mathbf{r}),$$

$$\psi_B(\mathbf{r}, t) = \sum_{n \neq 1s} \langle \phi_n(\mathbf{r}) | \psi(\mathbf{r}, t) \rangle \phi_n(\mathbf{r}),$$

$$\psi_C(\mathbf{r}, t) = \psi(\mathbf{r}, t) - \psi_B(\mathbf{r}, t) - \psi_g(\mathbf{r}, t).$$

The probability density of the electron in the ground state or the corresponding wavepacket can be easily obtained using $\rho_j = \langle \psi_j(\mathbf{r}) | \psi_j(\mathbf{r}) \rangle$, where the subscript $j$ refers to the ground, excited or continuum state.

In order to be able to track the origin of the saturation, we evaluate the time-dependent dipole oscillation, $d(t)$, and its components $d_{gb}(t), d_{gC}(t)$ and $d_{CC}(t)$ which have the following forms:

$$d(t) = \langle \psi(\mathbf{r}, t) | \mathbf{z} | \psi(\mathbf{r}, t) \rangle,$$

$$d_{gb}(t) = 2 \langle \psi_g(\mathbf{r}, t) | \mathbf{z} | \psi_B(\mathbf{r}, t) \rangle,$$

$$d_{gC}(t) = 2 \langle \psi_g(\mathbf{r}, t) | \mathbf{z} | \psi_C(\mathbf{r}, t) \rangle,$$

$$d_{CC}(t) = \langle \psi_C(\mathbf{r}, t) | \mathbf{z} | \psi_C(\mathbf{r}, t) \rangle.$$

Other components of the dipole oscillations are less relevant and therefore are not accounted for here. One therefore can see that $d(t) \approx d_{gb}(t) + d_{gC}(t) + d_{CC}(t)$.

The induced dipole polarization is related to the electric field by $d_j(\omega) = \chi_j(I, \omega) E(\omega)$, where $d_j(\omega)$ is the Fourier transform of the respective dipole moment, $E(\omega)$ is the Fourier transform of the electric field and $\chi_j(I, \omega)$ is the respective susceptibility. One can immediately see that the total susceptibility, $\chi^T(I, \omega)$, can be written as $\chi^T(I, \omega) \approx \chi_{gb}(I, \omega) + \chi_{gC}(I, \omega) + \chi_{CC}(I, \omega)$.

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The pulse envelope was chosen to have the form \( \sin^2(\pi t / T_{\text{max}}) \), with \( T_{\text{max}} \) being the total pulse duration. In the simulations, we employed a laser field with the central frequency of a Ti:sapphire laser system \( \omega_0 = 1.55 \text{ eV} \) (or laser wavelength of 800 nm). The total pulse duration \( T_{\text{max}} \) was taken to be 24 optical cycles, and each cycle was discretized into 512 temporal grids. In order to ensure the accuracy of the solution, we utilized a very fine radial grid \( \Delta r = 0.04 \text{ au} \), a large radial boundary \( r_{\text{max}} = 2500 \text{ au} \), and the maximum number of the angular momentum \( L_{\text{max}} = 60 \). The accuracy of the wavepacket separation was controlled using the identity \( |\rho_g + \rho_B + \rho_C - 1| = \epsilon \). We included 45 bound states and found that the error in the wavepacket separation, \( \epsilon \), was less than \( 10^{-6} \).

We controlled the numerical procedure by running the TDSE with a small intensity \( I = 9 \times 10^9 \text{ W cm}^{-2} \). With such a small intensity, the nonlinear and ionization effects are negligible, such that \( \chi^{(T)}(I, \omega) = \chi_B(\omega) + \chi_C(\omega) = \chi^{(1)}(\omega) \), i.e. only the linear term would exist. We found that the static polarizability \( \chi^{(1)}(\omega \rightarrow 0) = 4.51 \text{ au} \), was in excellent agreement with that obtained using the perturbative analysis [17].

The third-order nonlinear susceptibility was obtained using:

\[
\chi^3(\omega) = \lim_{I \rightarrow 0} \frac{\chi^{(T)}(I, \omega) - \chi^{(1)}(\omega)}{I}.
\]  

Alternatively, the third-order term can be obtained by taking the gradient of \( \chi(I, \omega) \) with respect to the intensity \( I \) in cases where the increase of \( \chi(I, \omega) \) is linear.

3. Results and discussion

3.1. The instantaneous dipole moment

Before discussing the simulation results, it is important to define instantaneous susceptibility as the behavior of the dipole moment that follows the electric field. Figure 1 shows the time-dependent dipole oscillation \( d(t) \), and its components \( d_{gb}(t) \) and \( d_{gc}(t) \) respectively, from top to bottom. The left panels show results obtained using an intensity of \( 6.4 \times 10^{13} \text{ W cm}^{-2} \), while the right panels show those obtained using an intensity of \( 1.47 \times 10^{14} \text{ W cm}^{-2} \). The red dotted curves are the rescaled electric fields, while the red thin curves are the corresponding electric field envelopes. We can see that at low intensity, \( d(t) \), \( d_{gb}(t) \) and \( d_{gc}(t) \) show relatively better overlap with both the electric field and its envelope, which demonstrates their instantaneous responses. The phase of the electric field and the dipole oscillation are also found to be the same. At high intensity (right panel), the rise of the oscillation amplitude of \( d(t) \) trails behind that of the electric field. More interestingly, at short times, the electric field and dipole moment are in phase but revert to being in anti-phase after some time. In addition, in the second half of the plot, the center of oscillation is displaced up. This reveals that at short times when the ionization is still low, \( d(t) \) is instantaneous, but then is predominately induced by ionization at later times.

On the other hand, the dipole oscillation components induced by the electronic transition from the ground state to either the excited or continuum state, \( d_{gb}(t) \) and \( d_{gc}(t) \) are, in general more instantaneous, even at high intensity. The phase of the electric field and that of the dipole oscillation components is also the same. This indicates that the electronic wavepacket contributes to the instantaneous nonlinear susceptibility spatially close to the atomic core, because the oscillator strengths of the states that are coupled with the ground state are proportional to \( e^{-r/\rho} \).

Thus, the electron that is removed from the atomic core does not contribute to the instantaneous
Figure 1. Plots of the various dipole oscillations, $d(t)$ and their components, $d_{gB}(t)$ and $d_{gC}(t)$. Panels (a)–(c) show results obtained at an intensity $6.4 \times 10^{13}$ W cm$^{-2}$, while those in (d)–(f) show results obtained using $1.47 \times 10^{14}$ W cm$^{-2}$. The red-dotted curves in each panel are the rescaled electric fields while the red-thin curves are the electric field envelopes.

response. The high frequency components appearing in $d_{gC}(t)$ are due to high frequency harmonics, as the electrons in the continuum are accelerated by the electric field back to the atomic core. This is not discussed here as high harmonic generation is not the focus of this work.

In order to investigate the effect of ground state depletion on saturation of the nonlinear susceptibility, we performed additional simulations with a total pulse duration equal to 12 cycles. Using shorter pulses, the ground state becomes less depleted than when longer pulses are used. The results generated using $T = 24$ cycles are depicted as thin black curves while those of 12 cycles are displayed as thick red curves. In figure 2(a), we show the ground state probability as a function of intensity, while in figure 2(b), the occupation probability in the continuum (solid curve) and excited states (dotted curve) are shown. Note that the probability density of the electrons in the ground, the excited, and the continuum states were evaluated at the end of the pulses. We can see that at low intensity ($I < 6 \times 10^{13}$ W cm$^{-2}$), the excitation process is dominant, whereas ionization dominates at high intensity. The growth of the electron density occupying the excited states seems to be linear, while that of the ionization is more likely exponential. Moreover, the excitation process is likely to have stabilized as it can be seen that the yields obtained using $T = 12$ and 24 cycles remain unchanged. This means that at high
intensity, the ground-state depletion is eventually due to an ionization rather than an excitation process.

In figures 2(c) and (d), we show the results of susceptibility components $\chi_{gB}(\omega_0)$ and $\chi_{gC}(\omega_0)$, respectively. At small intensity, the two curves in figures 2(c) and (d) coincide with each other, indicating that at low intensity, all responses are purely instantaneous, independent of the pulse shape and duration. At high intensity, $\chi_{gB}(\omega_0)$ becomes saturated, but the saturation from the simulation using $T = 24$ cycles occurs at relatively lower intensity than that of $T = 12$ cycles. Since $d_{gB}(t)$ is determined from the coupling between the ground state and the bound-excited wavepacket, and on the other hand, the excitation probability density remains unchanged (figure 2(b)), we can conclude that the saturation of the $\chi_{gB}(\omega_0)$ must be the result of ground state depletion.

On the other hand, $\chi_{gC}(\omega_0)$ already exhibits saturation even at small intensity. This is an indication that the nonlinear susceptibility components induced by the electronic transition from the ground state to the continuum have a negative sign. Higher ionization yields, as can be seen in figure 2(b), seem to bring only irrelevant small effects. This finding is a strong indication that the instantaneous component, $\chi_{gC}(\omega_0)$, is not influenced by the onset of
ionization, but is mainly the result of ground state depletion. This can easily be understood since the continuum wavepacket is spatially removed from the atomic core, such that its contribution to the instantaneous susceptibility is negligible.

It is also clear from figure 2 that the positions of the turning points are substantially determined by the onset of ground state depletion. Simulation using $T = 24$ cycles provides a turning point of $9 \times 10^{13}$ W cm$^{-2}$, whereas using $T = 12$ cycles yields the position of the turning point to be $1.2 \times 10^{14}$ W cm$^{-2}$. Based on the above findings, the total instantaneous susceptibility can now be approximately written in the form:

$$\chi_{\text{inst}}(I, \omega) = \hat{P}(I) \left[ \chi^{(1)}(\omega) + \chi^{(3)}(\omega) I \right],$$  \hspace{1cm} (5)

where $\hat{P}(I) = (1/T) \int_0^T P_g(I, t) \, dt$, is the time averaged ground state probability. Thus, at low intensity the susceptibility increases proportionally with intensity but then decreases as the ground state becomes depleted. High-order terms that saturate the nonlinear susceptibility are thus accommodated naturally by the presence of ground state depletion.

For the sake of comparison, in figure 3, we display the total instantaneous susceptibility obtained directly from simulation $\chi_{\text{inst}}(I, \omega_0) = \chi_{gB}(I, \omega_0) + \chi_{gC}(I, \omega_0)$, depicted as black curves, and those calculated using equation (5) as thick red curves. We can see relatively good agreement between them. Note that in equation (5), the linear part $\chi^{(1)}(\omega_0) = 4.59$ au was obtained by performing simulations at very low intensity, whereas $\chi^{(3)}(\omega_0) = 52$ au was obtained by taking the gradient in the interval where $\chi_{\text{inst}}(I, \omega_0)$ was linear.

Based on equation (5), the nonlinear part can then be written as

$$\chi_{\text{inst}}^{\text{NL}}(I, \omega) = \hat{P}_g(I) \left[ \chi^{(1)}(\omega) + \chi^{(3)}(\omega) I \right] - \chi^{(1)}.$$  \hspace{1cm} (6)

At low intensity where the depletion of the ground state is negligible such that $\hat{P}_g(I) = 1$, then $\chi_{\text{inst}}^{\text{NL}}(I, \omega) = \chi^{(3)}(\omega) I$ is simply the Kerr nonlinearity. Equation (6) thus generalizes the well-known Kerr nonlinearity formulae for high intensity and ultrashort laser pulses.
3.2. Time integrated dipole moment

Figure 4 (top) shows the dipole oscillation, $d_{CC}(t)$, resulting from coupling between the continuum wavepackets. For the sake of the discussion, we use the simulation results with an intensity of $1.47 \times 10^{14} \text{ W cm}^{-2}$, i.e. the largest in our simulation series. We see that besides the main oscillation driven by the electric field, there is a displacement of the center of the oscillation. Interestingly, the displacement starts during the second half of the pulse. To understand the origin of this displacement, we display in the inset the corresponding spectral intensity. The thick red curve shows the spectrum of the electric field. We can see that the displacement corresponds to low frequency oscillation (dc components) of the ionized electrons.
The origin of this displacement can be clarified using a model that is partially adopted from the semi-classical three step model [18]. In the absence of Coulomb interaction, the classical motion of a particle of mass $m$ and charge $q$ can be described using Newton’s law $ma = qE(t)$, with $a$ being the acceleration. The classical trajectory of the ionized electron induced by the electric field, after integrating twice over time $t$, can be expressed as:

$$d(t) = d_0 + v_0 t - \frac{q}{\omega_0^2} E(t),$$

(7)

where $d_0$ is the initial position at which the electron is ionized, and $v_0$ is the corresponding initial velocity. Thus, as a new ionized electron is generated, its initial velocity is zero. This low energy electron then appears to have dc spectral components. Note that the term ionized electron is used here to refer to electrons that are ionized but still influenced by the Coulomb interaction with their parent ions while plasma, as used shortly, indicates that the ionized electrons are completely free.

We elaborate further the relationship between the ionized electron and the plasma under the influence of the laser field. This is done by reconstructing the dipole oscillation $d_{CC}(t)$ within the spectral range to be the same as that of the driving electric field. To be clear, we perform a Fourier transform of $d_{CC}(t)$ depicted in figure 4 (top) and then reconstruct a new dipole oscillation $d_{NC}(t)$ with spectral components in the range $0.85 \leq (\omega/\omega_0) \leq 1.15$. The result is plotted as a black curve in the bottom panel of figure 4.

The dipole oscillation induced by the plasma, on the other hand, is simply given by adopting the third term of the right-hand side of equation (7), $d_{PL}(t) = -\rho/\omega_0^2 E(t)$, where $\rho$ is the density of the generated plasma. The data for $\rho$ generated directly from the TDSE is depicted in figure 4 (bottom) by the thick blue curve, whereas the calculated dipole oscillation $d_{PL}(t)$ is plotted as the dotted red curve. We can see good agreement between them, but in general $d_{NC}(t)$ exhibits a smaller amplitude, which is attributed as being due to the presence of a plasma correction factor. In the model, it is usually assumed that once an electron is ionized, the electron is completely free, but a Coulomb interaction still exists, even though it is weak. This residual Coulomb interaction decelerates the electron as it is driven by the electric field, such that its oscillation amplitude decreases.

We have checked the relationship between the ionization-induced dipole moment and that induced by the plasma for other intensities ($I < 1.47 \times 10^{14}$ W cm$^{-2}$). For a singly ionized atom, we have observed that the amplitude of the dipole oscillation $d_{CC}(t)$ is about 0.8–0.93 of that predicted by the semi classical model equation (7).

**4. Conclusions**

In conclusion, we have investigated the saturation of the nonlinear susceptibility of atomic hydrogen induced by intense ultrashort laser pulses. The electronic wavepacket is split into terms belonging to the ground, bound-excited and the continuum states, such that the dipole oscillation components contributing to the instantaneous susceptibility can be well identified. We found that at low intensity, the susceptibility is purely instantaneous. At high intensity, the susceptibility becomes saturated whereas the position of the saturation intensity is found to depend on the depletion of the electronic ground state. We further characterized the dipole oscillation resulting in the coupling between the continuum wavepackets and found good agreement with the plasma-induced dipole oscillation with a correction factor.
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