PHYSICAL REVIEW A **90**, 013426 (2014)

Ab initio and perturbative calculations of the electric susceptibility of atomic hydrogen

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(Received 17 June 2014; published 31 July 2014)

We apply a numerical basis-state method to determine the electric susceptibility of atomic hydrogen via calculations of the terms of the perturbative power series expansion up to eleventh order as well as from *ab initio* calculations of the field-induced polarization. The results of the perturbative calculations indicate that the applicability of the power series expansion is limited to intensities of a uniform electric field below 2×10^{13} W/cm^2 at a wavelength of 800 nm. While the predictions for the electric susceptibility from the perturbative power series tend to strongly increase beyond this intensity regime, those from *ab initio* calculations have a negative slope as a function of intensity. We therefore interpret that the latter feature is due to the nonperturbative interaction between the atom and the external field and an explanation via higher-order nonlinear terms is not applicable.

DOI: 10.1103/PhysRevA.90.013426 PACS number(s): 32.80.Wr, 42.65.An

I. INTRODUCTION

The nonlinear response of a medium to a strong external field, e.g., that of an intense laser pulse, gives rise to various nonlinear optical phenomena, such as high-harmonic and attosecond pulse generation (for a review, see Ref. [1]) or high-power femtosecond pulse filamentation (for reviews, see Refs. [2–4]). The effect of the external electric field on the medium is given by the induced polarization and/or a current. For many materials the induced polarization **P** is found to be proportional to the electric field **E** at frequency ω:

$$\mathbf{P}(\omega) = \epsilon_0 \chi(\omega) \mathbf{E}(\omega), \tag{1}$$

where χ is the electric susceptibility of the material. For the response to a strong electric field, the susceptibility at the driving frequency is often expanded in a power series as

$$\chi(\omega) = \chi^{(1)}(\omega) + \chi^{(3)}(\omega;\omega_1,\omega_2,\omega_3)\,|\mathbf{E}(\omega)|^2 + \chi^{(5)}(\omega;\omega_1,\omega_2,\omega_3,\omega_4,\omega_5)\,|\mathbf{E}(\omega)|^4 + \cdots, \tag{2}$$

where $\sum_j \omega_j = \omega$ and $\omega_j = \pm\omega$ with $\omega > 0$.

Several nonlinear phenomena, such as Kerr self-focusing or self-phase modulation [5], can be well understood by restricting the series to the lowest-order (i.e., third-order) nonlinear term $\chi^{(3)}$ of the expansion. Kerr self-focusing itself is a key ingredient in the standard model of femtosecond pulse filamentation, since it describes the collapse of the pulse. A self-guided light pulse, called a filament, is formed by the arrest of the pulse collapse leading to a dynamical balance between self-focusing and defocusing effects. Standard mechanisms considered for the arrest of the collapse are plasma effects or a negative fifth-order nonlinear susceptibility (for a detailed discussion of the theory of optical pulse propagation and filamentation, see, e.g., Ref. [6]).

However, results of a recent experiment [7] were interpreted such that third- and even fifth-order nonlinear susceptibilities were insufficient to account for the nonlinear response of an atomic or molecular gas via the induced polarization at standard light pulse intensities in filaments. In particular, at high intensities a strong negative slope in the susceptibility as a function of intensity was observed, in agreement with earlier theoretical predictions [8,9]. It was proposed to introduce higher-order Kerr effects (HOKEs) via the higher-order nonlinear susceptibilities $\chi^{(N)}$ to account for this feature [7]. The results started a controversial debate [10–33] about the significance and interpretation of the higher-order terms in the power series expansion [Eq. (2)], which currently is not fully settled.

Although the assumption to describe the filamentation process by using a perturbation theory has been questioned recently [30,33], different approaches to determine the higher-order coefficients $\chi^{(N)}$ in the power series, Eq. (2), from measurements [7], *ab initio* calculations [33], as well as the Kramers–Kronig transform of multiphoton-ionization rates [19,24] have been reported. We contribute to the present debate by determining both *ab initio* results for the susceptibility as well as the higher-order terms of the power series expansion independently from each other but within the same theoretical framework. We apply our approach to atomic hydrogen; our results indicate that the perturbative power series breaks down at intensities lower than those at which the strong negative slope of the susceptibility occurs. Thus, based on our results we conclude that a description of this feature via higher-order Kerr terms is inapplicable and, for an accurate theoretical description, if needed, one is required to account for the nonperturbative interaction between the atom and the intense laser pulse.

This paper is organized as follows: In the next section we outline the numerical basis-state method that is used to perform *ab initio* calculations of the field-induced polarization and the related susceptibility as well as to determine the terms of the perturbative power series expansion. In Sec. III we then present results for the higher-order terms in the perturbative series, followed by a discussion of the breakdown of the series and the applicability regime of perturbative calculations of the susceptibility of atomic hydrogen. Finally, we compare results of *ab initio* and perturbative calculations for the interaction with short laser pulses and conclude the paper with a brief summary of the results.

II. THEORY

We intend to provide both *ab initio* calculations of the susceptibility $\chi(\omega)$ as well as perturbative calculations of the coefficients $\chi^{(N)}(\omega)$ of the power series within the same theoretical approach. A common derivation of the higher-order coefficients is based on the field-free energy basis states of the atomic or molecular gas; in this study, atomic hydrogen. For our *ab initio* calculations we therefore make use of an expansion of the solution of the corresponding time-dependent Schrödinger equation (TDSE) into the field-free energy basis states as well. Such solutions are either based on analytical basis sets, e.g., B splines [34,35], or a numerical basis set [36], and we will make use of the latter approach. In the remainder of this section we first briefly recapitulate the basics of the *ab initio* numerical basis-state method and its application to calculate the polarization and susceptibility of atomic hydrogen due to a laser pulse. Then we proceed to review the definition and calculation of the susceptibility terms using this basis set.

A. *Ab initio* calculation of susceptibilities

In order to determine the susceptibility $\chi(\omega)$ of an atom in an external electric field using Eq. (1) we need to calculate the polarization response to the field. To this end, we solve the standard TDSE for atomic hydrogen interacting with the electric field of an intense laser field given by

$$i\frac{\partial}{\partial t}\left|\Psi(\mathbf{r},t)\right\rangle = H(\mathbf{r},t)\left|\psi(\mathbf{r},t)\right\rangle, \tag{3}$$

with the Hamiltonian in the length gauge using the dipole approximation as

$$H(\mathbf{r},t) = H_0(\mathbf{r}) + E(t)\,z, \tag{4}$$

where we restrict ourselves to a linearly polarized field $E(t)$, and

$$H_0(\mathbf{r}) = \frac{\hat{\mathbf{p}}^2}{2} - \frac{1}{r} \tag{5}$$

is the field-free Hamiltonian and $\hat{\mathbf{p}}$ is the momentum operator.

For the numerical solution we expand $\Psi(\mathbf{r},t)$ in a discrete set of field-free eigenstates of atomic hydrogen in a box along the radial distance as

$$\left|\Psi(\mathbf{r},t)\right\rangle = \sum_{nlm} c_{nlm}(t)\left|\psi_{nlm}(\mathbf{r})\right\rangle \tag{6}$$

$$= \sum_{nlm} c_{nlm}(t)\left|R_{nlm}(r)\,Y_{lm}(\Omega)\right\rangle, \tag{7}$$

where $R_{nl}(r)$ and $Y_{lm}(\Omega)$ are the radial wave functions and spherical harmonics, respectively. The radial functions $u_{nl}(r) = r R_{nl}(r)$ are generated as numerical solutions of the corresponding eigenvalue equation by using the Numerov method on a logarithmic one-dimensional finite-space grid with the boundary condition $u_{nl}(r=0) = u_{nl}(R_0) = 0$ [36]. Due to the finite box size R_0 both bound and continuum parts of the spectrum are discrete and all states in the basis set can be indexed by n. For a linearly polarized field the basis set can be restricted due to the dipole selection rule ($\Delta m = 0$). In the present study we consider the ground state of atomic hydrogen as the initial state and therefore only include states with $m = 0$ in our basis set. For convenience, we therefore drop the index m below.

We then use the field-free eigenstates to represent the dipole operator as

$$\hat{\mu} = -\sum_{n,l,n',l'} \left|\psi_{nl}\right\rangle\left\langle\psi_{nl}\right|\hat{z}\left|\psi_{n'l'}\right\rangle\left\langle\psi_{n'l'}\right| \tag{8}$$

and propagate the time-dependent Schödinger equation

$$\left(\frac{\hat{\mathbf{p}}^2}{2} - \frac{1}{r} + E(t)\,\hat{\mu}\right)\left|\Psi(\mathbf{r},t)\right\rangle = i\frac{\partial}{\partial t}\left|\Psi(\mathbf{r},t)\right\rangle \tag{9}$$

in the electric field of a pulse, which is given by

$$E(t) = E_0 \sin^2\left(\frac{\pi t}{T_0}\right)\sin(\omega t), \tag{10}$$

where E_0 is the field strength, T_0 is the time duration of the pulse, ω is the central field frequency, using the Crank–Nicolson method (for details of the numerical method, see Ref. [36]).

Previously, we applied this method to the nonperturbative interaction of atomic hydrogen with a short intense laser pulse and obtained excitation and ionization probabilities as well as momentum distributions of the photoelectron. The convergence of the results with respect to the size of the numerical basis set as well as the agreement of results with those of two-dimensional grid calculations have been shown [36]. In the present study we calculate the susceptibility $\chi(\omega)$ by taking the Fourier transform of the dipole moment:

$$\frac{\epsilon_0\chi(\omega)}{\rho} = \frac{\mathcal{FT}[\mu(t)](\omega)}{E_0}, \tag{11}$$

where

$$\mu(t) = -\left\langle\Psi(t)\right|\hat{z}\left|\Psi(t)\right\rangle \tag{12}$$

is the time-dependent dipole moment and ρ is the number density of atoms. For the results of our *ab initio* calculations shown below, we present the quantity $\epsilon_0\chi(\omega)/\rho$ instead of $\chi(\omega)$, since the former is independent of the atomic density.

B. Perturbative calculation of higher-order nonlinear susceptibilities

In order to obtain the terms for the coefficients in the power series, we write the Nth-order perturbative correction of the ground-state wave function interacting with an external field as (cf., Ref. [5])

$$\left|\psi^{(N)}(\omega_1,\ldots,\omega_N)\right\rangle = \sum_{j_N\neq j_0}\cdots\sum_{j_1\neq j_0}\left[\prod_{i=1}^{N}\frac{\langle\psi_{j_i}|\hat{\mu}E(\omega_i)e^{-i\omega_i t}|\psi_{j_{i-1}}\rangle}{\omega_{j_i}-\omega_{j_0}-\sum_{k=1}^{i}\omega_k}\right]\left|\psi_{j_N}\right\rangle, \tag{13}$$

where $\hat{\mu}$ is given by Eq. (8), ω_{j_0} is the ground-state energy, ω_k and ω_i are the participating frequencies of the electric field, and j_i denotes state in the basis set. In this work we use the states of the numerical basis set for the calculations. We neglect the finite lifetimes of the excited states, assuming that all transitions considered in this study are far from any resonance.

The Nth-order perturbative correction to the single-atom polarization can then be written as

$$\langle \mathbf{P}^{(N)}(\omega)\rangle = \mathcal{P}\sum_{j'=0}^{N}\langle\psi^{(j')}|\hat{\mu}|\psi^{(N-j')}\rangle, \tag{14}$$

where $\mathcal{P}$ refers to the average of all permutations of the frequencies, where $\omega_j = \pm\omega$ and $\sum_j \omega_j = \omega$. By using the symmetry of the electric field with respect to positive and negative frequencies about the central frequency ω, Eq. (14) can be written in the form [5]

$$\langle \mathbf{P}^{(N)}(\omega)\rangle = \epsilon_0\chi^{(N)}(\omega)|\mathbf{E}(\omega)|^{N-1}\mathbf{E}(\omega), \tag{15}$$

where $\chi^{(N)}$ is the Nth-order coefficient in the power series expansion of the susceptibility (2).

III. RESULTS AND DISCUSSION

In this section we first present the results of our calculations for the coefficients in the power series expansion by showing the convergence of the results with respect to the box size of the numerical grid R_0 and the basis size. We then proceed by analyzing criteria for the breakdown of the series expansion at high field strengths and identify a regime for a critical field strength. Finally, we compare the results of the perturbative calculations with those of the *ab initio* calculations in order to analyze whether or not the series expansion describes the negative slope of the nonlinear susceptibility at high field strengths.

A. Perturbative higher-order nonlinear susceptibilities

In Fig. 1 we present the results of our perturbative calculations for the susceptibility coefficients of the power series, Eq. (2), up to $\chi^{(11)}$ as a function of the frequency of a uniform external electric field. Over the frequency regime shown, the coefficients are almost constant, but the higher-order coefficients tend to increase at the largest frequencies considered in the present calculations. The increase indicates that one approaches the regime of resonant transitions. Since the life times of the excited states are neglected in the present perturbative calculations, we have restricted ourselves to calculations up to $\chi^{(11)}$ and the frequency regime below 0.06 a.u. (corresponding to wavelengths of approximately 800 nm and longer). Note here already that, based on the perturbative results in Fig. 1, it is unlikely that a negative slope in the nonlinear susceptibility can be explained via the power series expansion in the present theory, since the first five coefficients of the series are found to be positive over the frequency regime studied.

The convergence of the results presented in Fig. 1 was tested well with respect to the size of the radial box, R_0, and the basis size. For the present set of calculations we have found that a box size of $R_0 = 500$ was sufficient to reduce the error to be within 0.5%. The size of the numerical basis in the present calculation is determined by the maximum angular momentum [$l_{\max} = (N+1)/2$, where N is the order of the coefficient calculated], and the maximum principal quantum number ($n_{\max}$) chosen. For a grid size of $R_0 = 500$, we varied the principle quantum number up to $n_{\max} = 800$ for our perturbative calculations.

In Fig. 2 we show results for $\chi^{(N)}$ as a function of $n_{\max}$ for $R_0 = 500$ at $\omega = 0.056$. One can clearly see that the results for each of the coefficients converge to a finite value for large $n_{\max}$. The cross on each line indicates that the result is within 1% of the corresponding value at $n_{\max} = 800$. As one would expect, each higher-order term requires additional states to be included in the calculation in order to reach convergence. Furthermore, the dashed line at $n_{\max} = 19$ indicates the separation of bound from continuum states in our basis set. For each of the nonlinear coefficients ($\chi^{(N)}$ with $N \geqslant 3$) we observe that the contributions from the bound states to the coefficient are positive while those from the continuum states are negative.

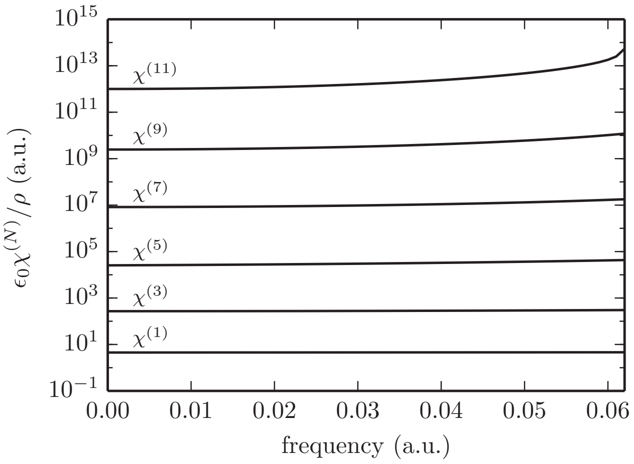

FIG. 1. Results of calculations for the different terms in the power series expansion, Eq. (2), as a function of frequency ω of a uniform electric field.

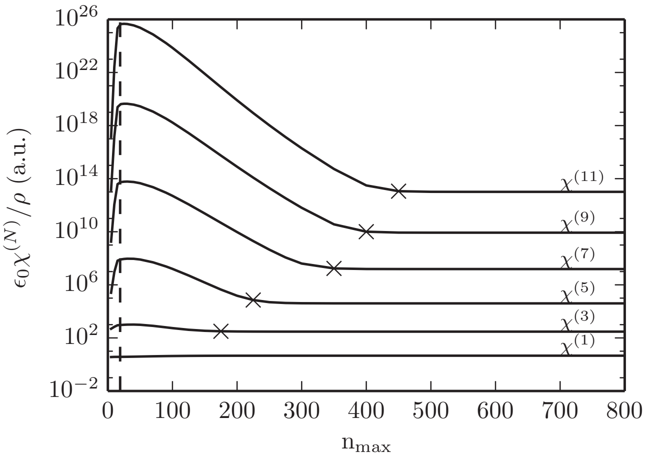

FIG. 2. Perturbative susceptibility coefficients $\chi^{(N)}$ in the power series expansion, Eq. (2), as a function of the maximum principal quantum number $n_{\max}$ in the basis set. The radial box size was $R_0 = 500$. The dashed line denotes the transition from bound to continuum sets in the present basis set, while the crosses indicate convergence of the results within 1% of the respective result at $n_{\max} = 800$.

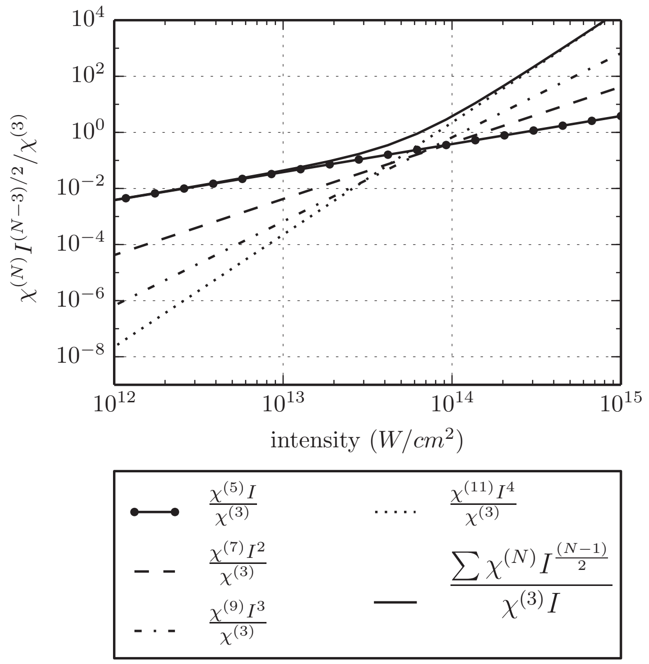

FIG. 3. The perturbative nonlinear susceptibility terms $\chi^{(N)}I^{(N-1)/2}$ scaled by $\chi^{(3)}I$ as a function of the intensity of a uniform electric field at 800 nm.

Thus, in order to reach convergence obviously both bound and continuum states have to be included.

B. Breakdown of the perturbative power series expansion

In order to be applicable, the power series expansion, Eq. (2), must converge. Since we cannot determine all the terms in the expansion, we intend to estimate a region of applicability by investigating the relative contributions from the low-order nonlinear terms in the expansion. To this end, we have determined the ratios of the higher-order nonlinear terms ($N \geqslant 5$) to the lowest-order nonlinear term, i.e., $\chi^{(N)}I^{(N-1)/2}/(\chi^{(3)}I)$ as a function of the intensity $I = |E|^2$. The results are shown in Fig. 3 along with the sum of all higher-order terms calculated with respect to the third-order term (solid line). For low intensities ($\approx 10^{12}$ W/cm^2), each higher-order nonlinear term is much smaller than the lowest-order nonlinear term in the expansion. We therefore can expect that the power series does converge in this intensity regime. On the other hand, at high intensities ($\approx 10^{14}$ to 10^{15} W/cm^2) the trend is reversed: higher-order terms are larger in magnitude than the lowest-order term, indicating that the series may diverge.

In order to further narrow down the intensity regime at which the convergence of the power series expansion becomes questionable, we note that the ratio of the sum of all higher-order terms calculated to the lowest-order nonlinear term exceeds 10% at about 2×10^{13} W/cm^2. From the results in Table I, we further see that in this intensity regime the ratio of successive terms in the perturbative power series expansion does exceed 10%, which may be considered as another indication for the inapplicability of the series expansion.

TABLE I. Intensities (in units of 10^{12} W/cm^2) at which the ratio $R = \chi^{(N)}I/\chi^{(N-2)}$ equals either 0.01 or 0.1.

	$N = 5$	$N = 7$	$N = 9$	$N = 11$
$R = 0.01$	2.6	0.91	0.63	0.29
$R = 0.1$	26	9.1	6.3	2.9

C. Comparison of results of perturbative and *ab initio* calculations

Ab initio calculations of the susceptibility are performed for laser pulses at a central wavelength of 800 nm having a sin^2 envelope and a total pulse length consisting of a finite number of electric field cycles. In order to compare our results for the power series expansion, which are determined for a uniform electric field, to those of the *ab initio* calculations, we therefore need to account for the finite length and envelope of the pulse. To this end, we have averaged each term of the power series expansion over the intensity distribution of the pulse.

The comparison of the results of the perturbative calculations and those of the *ab initio* calculations for a ten-cycle pulse (Fig. 4) show excellent agreement at low intensities. In this regime of intensities both results increase linearly with intensity, in agreement with the expectation that the lowest-order nonlinear term is dominant at these intensities. At higher intensities, however, the results of the two calculations reveal an opposite trend. While the *ab initio* results show, in good quantitative agreement with earlier predictions [8,9], a negative slope for the susceptibility as a function of intensity, the sum of the first five terms in the power series expansion has a positive slope, as expected from the positive sign of each term in the power series determined here.

The peak intensity at which the discrepancy between perturbative and *ab initio* results equals 1% is given in Table II for different pulse lengths at 800 nm. The result is slightly larger than our estimates of the applicability limit of the perturbative

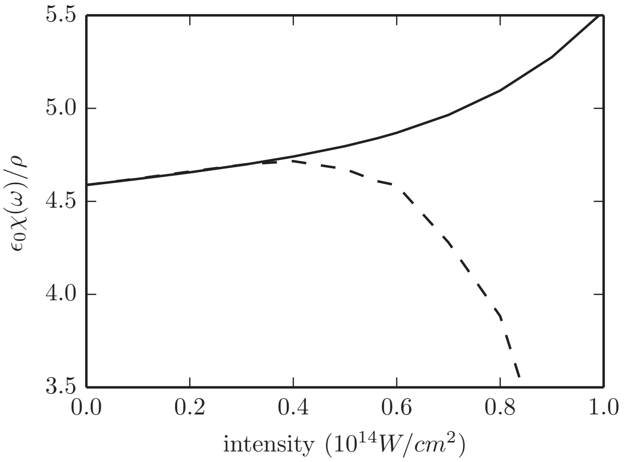

FIG. 4. Comparison of results of perturbative (solid line) and ab initio (dashed line) calculations for the susceptibility as a function of peak intensity of a 10-cycle laser pulse at 800 nm.

TABLE II. Peak intensities (in units of 10^{13} W/cm^2) at which the relative error between the results for the *ab initio* susceptibility and the perturbative susceptibility equals 1% for laser pulses of different pulse lengths (given in field cycles) at 800 nm.

Pulse length	4	10	16	30
Peak intensity	4.7	4.2	3.9	3.4

series expansion in the previous section, since the latter one has been obtained for a uniform field. However, we can still conclude that the negative slope of the susceptibility is a result of the *nonperturbative* interaction between the hydrogen atom and the intense laser pulse. Thus, an explanation of this feature using higher-order-expansion terms (e.g., HOKE) appears to be questionable and not applicable. This also implies that a determination of higher-order terms from *ab initio* results in the intensity regime above 2 to 4×10^{13} W/cm^2 for atomic hydrogen is meaningless. This is in agreement with the results of Köhler *et al.* [33], who were not able to extract consistent HOKE terms from results of *ab initio* simulations in this intensity regime. Instead, if necessary, the atom-field dynamics has to be treated using a nonperturbative theoretical description of the polarization and susceptibility. We note that our results, presented in Table II show that the critical intensity regime for a breakdown of the perturbative approach slightly depends on pulse duration of the pulse. We did not perform calculations for other atoms, which would require an extension of the numerical basis-state method to include spin-orbit effects. However, we expect that the critical intensity regime depends on the ionization energy of the atomic system, as also known for other nonlinear processes. The critical intensity should increase (decrease) for higher (lower) ionization potentials as compared to the results presented for atomic hydrogen here.

IV. CONCLUSIONS

We calculated the electric susceptibility of atomic hydrogen by using a numerical basis-state method, via a perturbative power series expansion as well as the *ab initio* results for the field-induced polarization. The results of our perturbative calculations indicate a breakdown of the series expansion at intensities in the lower range of 10^{13} W/cm^2. Comparison with the *ab initio* results shows a discrepancy in the trend as a function of intensity in the same intensity regime. We therefore conclude that the negative slope of the electric susceptibility of atomic hydrogen at higher intensities cannot be explained by the higher-order terms of a perturbative power series expansion. Instead, the feature has to be considered as a signature of the nonperturbative interaction between the atom and the field.

ACKNOWLEDGMENTS

This work was primarily supported through the AFOSR MURI “Mathematical Modeling and Experimental Validation of Ultrafast Light-Matter Coupling associated with Filamentation in Transparent Media,” Grant No. FA9550-10-1-0561. A.J.-B. was also supported by grants from the National Science Foundation (Grants No. PHY-1125844 and No. PHY-1068706). We thank the MURI team, in particular M. Kolesik and J. V. Moloney, as well as S. L. Chin for stimulating discussions. This work utilized the Janus supercomputer, which is supported by the National Science Foundation (Award No. CNS-0821794) and the University of Colorado Boulder. The Janus Supercomputer is a joint effort of the University of Colorado Boulder, the University of Colorado Denver, and the National Center for Atmospheric Research. Janus is operated by the University of Colorado Boulder.

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