RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 41, NUMBER 7 1 APRIL 1990

Higher-harmonic production in a model atom with short-range potential

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(Received 7 August 1989)

Production of higher harmonics in a model atom described by a three-dimensional δ-function potential in the presence of a monochromatic linearly polarized field is investigated. The rates for production of the individual harmonics can be expressed as one-dimensional integrals. The only property of the atom that enters the model is its ionization energy. The results are in qualitative agreement with many of the data. In particular, the model exhibits a "rising plateau."

Interest in multiphoton-ionization of atoms has risen considerably with the observation of above-threshold ionization,[1] the first phenomenon in the history of the field that is not explained by lowest-order perturbation theory. A closely related process is the emission of higher harmonics of the incident laser light by the irradiated atoms. A number of experimental studies has shown[2] rather copious production of surprisingly high harmonics. The interest in these harmonics rose considerably when it turned out[3-5] that the intensity of the harmonics did not, as anticipated, more or less continuously decrease with increasing harmonic number. Rather, after an initial, rapid decrease the harmonic intensities establish a "plateau" region of fairly constant intensities. Finally, the plateau has a quite well-defined rim from where on the harmonic intensities decrease quickly. The most recent and extensive measurements on xenon, argon, krypton, and neon for intensities of a few times 10^{13} W/cm^2 at a wavelength of 1064 nm are reported in Ref. 5.

A theoretical description of the observed production of higher harmonics is much more complicated than a theory of above-threshold ionization. The latter experiments could be performed at low pressure such that collective effects could essentially be ruled out. In contrast, production of higher harmonics requires comparatively high pressures. As a consequence, the atoms no longer emit (or absorb) independently. Maximum net emission of the entire gas sample in a given direction requires "phase matching"[6] which, in a possibly complicated way, depends on details of the atom, the laser pulse, and the gas jet. Phase matching becomes less effective for increasing harmonic number. It has been estimated that (in the case of a gas jet with Lorentzian shape) the phase-matching factor decreases by almost 5 orders of magnitude from the 5th to the 31st harmonic.[5] This would imply that the response of the individual atom has to *increase* by a corresponding factor in order to yield the observed plateau in the collective response.[7] It is not quite clear, however, in how far the phase-matching results, which assume comparatively weak fields, apply to the current experiments.

In view of these difficulties, previous work has concentrated on harmonic emission by a single atom, using lowest-order perturbation theory,[8-10] essential-state models,[11] one-dimensional computer simulations of model atoms,[12] as well as a fairly realistic Xe atom.[13] There is also an analytically solvable model where the external field couples to the Lenz vector rather than the position vector.[14] References 9, 10, 12, and 13 show clear evidence of a plateau region. More and more sophisticated computer simulations of realistic atoms including the collective aspects are under way.[15]

In this paper, we will consider one of the simplest conceivable models: one three-dimensional δ-function potential with a regularizing factor.[16] The limitation to one "atom" prevents direct comparison of the results with the experiments. The oversimplification of our atom, however, should give insight into whether the production of very high harmonics and the existence of the plateau is a general intense-field effect or a property dependent on the more subtle features of the atom.

Our model potential is

$$V(\mathbf{r}) = \frac{2\pi}{\kappa m}\delta(\mathbf{r})\frac{\partial}{\partial r}r\,, \tag{1}$$

where the regularizing factor $(\partial/\partial r)r$ acts on the ensuing wave function. The potential supports exactly one bound state with binding energy $|E_0| = \kappa^2/2m$ and wave function $\exp(-\kappa r)/r$. In the presence of a monochromatic field with circular or general elliptic polarization quasienergy wave functions have been found in (not entirely, but largely) analytic form.[17-19] In order to generate harmonics in the context of the dipole approximation, we will consider a linearly polarized monochromatic field with the vector potential

$$\mathbf{A}(t) = a\cos(\omega t)\hat{\mathbf{x}}\,. \tag{2}$$

Any quasienergy solution of the Schrödinger equation satisfies

$$\Psi(\mathbf{r},t) = \int d^3r'\,dt'\,G^{(E)}(\mathbf{r}t,\mathbf{r}'t')V(\mathbf{r}')\Psi(\mathbf{r}'t')\,, \tag{3}$$

where $G^{(E)}(\mathbf{r}t,\mathbf{r}'t')$ denotes the propagator of a free particle in the presence of the field (2). Following Refs. 17-19, we notice that, owing to the δ-function potential (1), Eq. (3) allows for the computation of the wave function $\Psi(\mathbf{r},t)$, provided it is known at the origin. We are interested in the quasienergy solution $\Psi_0(\mathbf{r},t)$ which develops out of the ground state when the field is turned on. One can easily convince oneself that near the origin this wave function differs from the wave function in the ab-

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sence of the field only by a time-dependent factor $u(t)$, viz.,

$$\Psi_0(\mathbf{r},t) \underset{r\to 0}{\sim} [(1/r)-\kappa]u(t) . \tag{4}$$

Using Eq. (4) in Eq. (3), one can carry out the integration over $\mathbf{r}'$. In the limit $\mathbf{r}\to 0$, one then obtains an integral equation for $u(t)$. The ansatz

$$u(t)=e^{-iEt}w(t), \quad w(t)=\sum_{n=-\infty}^{\infty} a_n e^{2in\omega t}, \tag{5}$$

with periodic $w(t)$, viz., $w(t)=w(t+\pi/\omega)$, determines both the quasienergy E and the function $w(t)$, i.e., the Fourier coefficients a_n. *A priori,* the quasienergy is only specified modulo multiples of $\hbar\omega$. We have to select that particular branch $E=-|E_0|+\Delta-\frac{1}{2}i\Gamma$, where Δ and Γ tend to zero when the field is turned off. The real part Δ is the small Stark shift of the ground state while Γ is the total ionization rate per time. The quasienergy approach only makes sense when both quantities are very small compared with $|E_0|$.

If the quasienergy E and the Fourier coefficients a_n are known, the wave function $\Psi_0(\mathbf{r},t)$ of the dressed ground state is determined by Eq. (3) up to a quadrature. We can then evaluate the matrix element for emission of a photon with frequency Ω and polarization $\boldsymbol{\varepsilon}$ by the atom which remains in the process in the dressed ground state $\Psi_0(\mathbf{r},t)$:

$$M=\left[\frac{2\pi\Omega}{V}\right]^{1/2}\int d^3r\,dt\,e^{i\Omega t}\Psi_0(\mathbf{r},t)^* e\mathbf{r}\cdot\boldsymbol{\varepsilon}\Psi_0(\mathbf{r},t) , \tag{6}$$

where V is a normalization volume. We notice that this is essentially the Fourier transform of the ground-state expectation value of the dipole moment as calculated in Refs. 15 and 16. As we already did for the description of the laser field, here we adopted the dipole approximation for the emitted photon, too.

All of the integrations indicated in Eqs. (3) and (6) can be carried out analytically up to one final integration which is left for numerical evaluation. The result for the matrix element M will display a sequence of odd harmonics which formally originate from two different sources: Both the Volkov propagator $G^{(E)}$ and the periodic function $w(t)$ [Eq. (5)] have higher-harmonic frequencies in their Fourier expansions. Both contributions are qualitatively similar. Both, for example, have the same leading power of η. Quantitatively, however, the contributions arising from the propagator are dominant. It has been shown[19] that the approximation $w(t)=$const, i.e., $a_n=a_0\delta_{n0}$, introduces a relative error of order $(\omega/4|E_0|)^2$ in the total ionization rate Γ. The corresponding error in the harmonic spectrum may well be larger. Our preliminary estimates of the coefficients a_n indicate that for $\eta=(ea)^2/4m\omega\lesssim 3$ we have $|a_1|\lesssim 10^{-4}|a_0|$. The higher coefficients decrease further in magnitude. The approximation $a_n=a_0\delta_{n0}$ therefore seems reasonably safe. The remaining constant a_0 is determined by the normalization of the field-free wave function $|a_0|^2=(8m|E_0|)^{1/2}$.

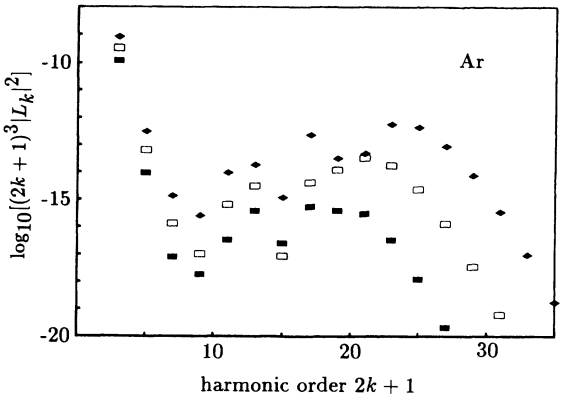

FIG. 1. $\log_{10}(2k+1)^3|L_k|^2$ as a function of the harmonic order $2k+1$ for argon for three different intensities: solid diamond, 3×10^{13} W/cm^2; open rectangle, 2.21×10^{13} W/cm^2; solid rectangle, 1.55×10^{13} W/cm^2.

Squaring the matrix element (6), multiplying with the phase-space density of the emitted photon with momentum $\mathbf{K}(|\mathbf{K}|=\Omega)$, dividing by a long normalization time, and integrating over the frequency then gives the rate of emission $dR_{2k+1}/d\Omega_{\mathbf{K}}$ per unit time and solid angle of a photon with frequency $\Omega=(2k+1)\omega$

$$\frac{dR_{2k+1}}{d\Omega_{\mathbf{K}}}=4\pi\varepsilon_x^2(2k+1)^3\frac{r_0}{\lambda}\omega|L_k|^2, \tag{7}$$

with

$$L_k=2\left[\frac{|E_0|\eta}{\omega}\right]^{1/2}\int_0^\infty\frac{d\rho}{\rho^{3/2}}e^{-ik\rho}\{\cos\alpha_k\beta_k(\rho)J_k[z(\rho)]-e^{-i\rho}\sin\alpha_k\bar{\beta}_k(\rho)J_{k+1}[z(\rho)]\} , \tag{8}$$

where

$$\alpha_k=\left\{\frac{|E_0|}{\omega}+\eta\left[1-\left[\frac{\sin\rho/2}{\rho/2}\right]^2\right]\right\}\rho+(-1)^{k+1}\frac{\pi}{4}, \quad \beta_k(\rho)=\frac{1}{k(2k+1)}\left[1-\frac{4k}{(2k+1)\rho}e^{-i\rho/2}\sin\rho/2\right],$$

$$\bar{\beta}_k(\rho)=\frac{1}{(k+1)(2k+1)}\left[1-\frac{4(k+1)}{(2k+1)\rho}e^{-i\rho/2}\sin\rho/2\right], z(\rho)=\eta\left[\sin\rho-\frac{4\sin^2\rho/2}{\rho}\right]. \tag{9}$$

The quantity $\eta=(ea)^2/4m\omega$ is the ratio of the quivering energy of a free electron in the field (2) over the energy $\hbar\omega$ of a field quantum, $r_0=e^2/m$ is the classical electron radius, and $\lambda=2\pi/\omega$ denotes the laser wavelength. In the above expressions we dropped the small quantities Δ and Γ.

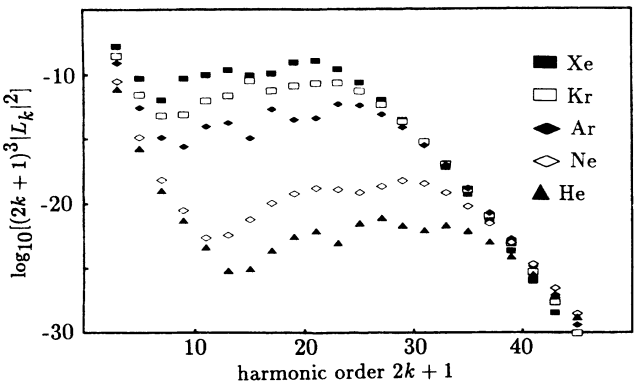

FIG. 2. $\log(2k+1)^3|L_k|^2$ as a function of the harmonic order $2k+1$ for $I=3\times10^{13}$ W/cm^2 and $|E_0|/\omega$ corresponding to the rare gases Xe, Kr, Ar, Ne, and He.

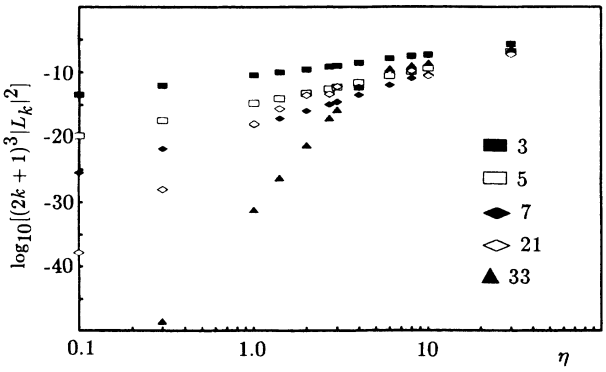

FIG. 3. $\log(2k+1)^3|L_k|^2$ as a function of η [Eq. (7)] for several higher harmonics in argon.

The results which come out of this model are shown in Figs. 1–3. In each case the quantity $(2k+1)^3|L_k|^2$ is plotted. Figure 1 displays the harmonic spectrum of Ar for several intensities I of the laser field while Fig. 2 compares the spectra of Xe, Kr, Ar, Ne, and He for fixed intensity $I=3\times10^{13}$ W/cm^2. In each case, the laser frequency is $\hbar\omega=1.16$ eV [yttrium aluminum garnet (YAG)]. The specific atom enters only via its ionization energy $|E_0|$, which determines the parameter $\kappa=(2m|E_0|)^{1/2}$. The ionization potentials for Xe, Kr, Ar, Ne, and He are 12.13, 14.00, 15.76, 21.56, and 24.59 eV, respectively, corresponding to a minimum number of photons for ionization of $N=11$, 13, 14, 19, and 22, respectively, for the YAG frequency. All spectra show an initial rapid decrease of the harmonic intensities with a pronounced minimum between the 7th harmonic for Xe and the 13th for He (at $I=3\times10^{13}$ W/cm^2). This initial dip is followed by a more-or-less ragged plateau which is, on the average, sloping upwards rising from the dip to its maximum by several orders of magnitude. From some order on, which for $\eta\gtrsim1$ is roughly given by $2k+1\sim2\times(|E_0|/\omega+\eta)+1$, the harmonic intensities start dropping rapidly. All of these features are qualitatively present in the data of Ref. 5 (taking into account the phase-matching factor). Quantitatively, the most obvious discrepancy between this model and the data is in the steep drop of the initial harmonics which the model exaggerates. Otherwise, quantitative differences are within 1 order of magnitude. Figure 3 gives the intensity dependence of selected harmonics. For $\eta\lesssim0.3$ the slope is $2k+1$ as expected in lowest-order perturbation theory [note that $|L_k|^2\sim\eta^{2k+1}$ for $\eta\ll1$ since $z(\rho)\sim\eta$]. It is interesting that even for intensities where the individual harmonics still exhibit the slope predicted by perturbation theory, their emission rates do not always decrease with increasing order. For example, Fig. 3 shows that for $\eta\gtrsim1$, the 21st harmonic becomes more intense than the 7th, even though both still display the respective slope of $2k+1$ of lowest-order perturbation theory. Similar effects have been observed by other authors.[9,10] In the plateau region the intensity dependence is more complicated. Figure 3 is not really trustworthy for $\eta\gtrsim10$ due to the approximation that $a_n=a_0\delta_{n0}$; the values given are only supposed to indicate the trend.

There are two gross features which dominate the behavior of the integral L_k: the smallness of the Bessel functions when $\max[z(\rho)]\sim1.5\eta\ll k$ and the exponential which is rapidly oscillating except when $|E_0|/\omega+\eta\sim k$. The interplay of these two properties accounts for the initial dip and the rising plateau with the steep upper edge.

In view of the extreme crudeness of the model it should not be compared with specific features of the data. Noticing, however, the strong suppression of the 15th harmonic in Ar in the intensity range covered by Fig. 1, it is difficult not to be reminded of the absence of the 13th harmonic in Ar in the data.[5] The suppression shown in Fig. 1 is due to the two terms in the integral L_k beating against each other. This is strongly intensity dependent: the suppression goes away for lower and higher intensities. While the near agreement with the data may well be coincidental, it is remarkable that a model as simple as this allows for a strong dynamic suppression of individual harmonics.

In conclusion, the simple model of a δ-function potential qualitatively displays many of the features observed in higher-harmonic emission by rare gases including a rising plateau in the single-atom response which should lead to a flat plateau in the collective response.

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