PHYSICAL REVIEW A VOLUME 45, NUMBER 7 1 APRIL 1992

Semiclassical treatment of laser excitation of the hydrogen atom

G. D. Billing and N. E. Henriksen
Department of Chemistry, H. C. Ørsted Institute, University of Copenhagen, 2100 Copenhagen, Denmark

C. Leforestier
Laboratoire Chimie Theoriqe, Universite de Paris Sud, Orsay, France
(Received 11 October 1991)

We present an alternative method for studying excitation of atoms in intense laser fields. In the present paper we focus upon the optical harmonic generation by hydrogen atoms.

PACS number(s): 42.65.Ky

INTRODUCTION

The excitation of atoms and specifically hydrogen atoms in intense laser fields has received much attention recently. Thus the problem has been attacked by introducing the Floquet ansatz [1], using a classical trajectory treatment of the electron [2], a variational Fourier transform method [3], and by wave-packet techniques [4]. It is the purpose of this paper to present a semiclassical approach, which has the advantage over a completely classical treatment that the generated harmonics spectrum converges with just a few (around 50) trajectories and that it easily is extended to more than one electron system. Furthermore, fewer equations have to be solved than those necessary for the classical trajectory treatment.

THEORY

In order to facilitate the solution of the laser-atom interaction problem further it is possible to introduce a mixed quantum-classical description, in which the radial motion is treated classically. Thus one obtains the following mixed Hamiltonian:

$$H_{\text{mixed}}=\frac{p_r^2}{2\mu}-\frac{e^2}{r}-\frac{\hbar^2}{2\mu r^2}\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial}{\partial\theta}\right)-\frac{\hbar^2}{2\mu r^2\sin^2\theta}\frac{\partial^2}{\partial\phi^2}-g(t)eE_0r\cos\theta\sin(\omega t)\,, \tag{1}$$

where μ is the reduced mass, θ and ϕ the usual spherical angles, E_0 the field strength of a plane polarized field in the z direction, ω the laser frequency, and $g(t)$ a time-dependent shape function. The above quantization conserves the l-selection rules for the dipole transitions but ignores the n selection. It is the purpose of this section to investigate the validity of this approximation. Note that it facilitates the solution for the larger systems (more electrons) appreciably. The wave function is now expanded in spherical harmonics as

$$\psi(\theta,\phi,t)=\sum_{l,m}c_{lm;l_0m_0}(t)Y_{lm}(\theta,\phi)\,. \tag{2}$$

From the time-dependent Schrödinger equation we then obtain

$$i\hbar\dot{c}_{lm;l_0m_0}=\left(\frac{1}{2\mu}p_r^2-\frac{e^2}{r}+\frac{\hbar^2}{2\mu r^2}l(l+1)\right)c_{lm;l_0m_0}-g(t)eE_0r\sin(\omega t)\sum_{l',m'}c_{l'm';l_0m_0}\langle Y_{lm}|\cos\theta|Y_{l'm'}\rangle \tag{3}$$

where (l_0m_0) is the initial state, and the matrix elements may be evaluated as

$$\langle Y_{lm}|\cos\theta|Y_{l'm'}\rangle=\delta_{m,m'}\times\begin{cases}\delta_{l,l'-1}\left[\dfrac{(l-m+1)(l+m+1)}{(2l+1)(2l+3)}\right]^{1/2}\\ \delta_{l,l'+1}\left[\dfrac{(l-m)(l+m)}{(2l-1)(2l+1)}\right]^{1/2}\end{cases}. \tag{4}$$

For nonlinear polarization we would obtain off diagonal matrix elements in the m quantum number. It is nevertheless possible to obtain the same reduction in the complexity of the problem as for linearly polarized light by introducing the classical limit (large l) of the coupling elements (see below and Ref. [5]).

Before we consider these equations further we wish to introduce an effective Hamiltonian for the classical motion by

$$H_{\text{eff}}=\langle\psi|\hat{H}_{\text{mixed}}|\psi\rangle=\frac{1}{2\mu}p_r^2-\frac{e^2}{r}+\frac{\hbar^2}{2\mu r^2}S(t)-eE_0g(t)r\sin(\omega t)T(t) \tag{5}$$

where

$$S(t)=\sum_l l(l+1)|c_{lm;l_0m}|^2 \tag{6}$$

and

$$T(t)=\sum_{l,l'} c^{*}_{lm,l_0m}c_{l'm,l_0m}\langle Y_{lm}|\cos\theta|Y_{l'm}\rangle . \quad (7)$$

The time-dependent equations (3) are due to the selection rules (4) tridiagonal in the quantum number l. Such equations are solvable analytically if the large l limit of the matrix elements is taken. Thus we introduce [6]

$$\langle Y_{l'm}|\cos\theta|Y_{lm}\rangle \sim d^{l}_{l'-l,0}\left(\frac{\pi}{2}\right) d^{l}_{l'-l,0}(\beta) , \quad (8)$$

where $\cos\beta=m/l$. Thus for $m=0$ we obtain

$$\langle Y_{l'0}|\cos\theta|Y_{l0}\rangle \sim \tfrac{1}{2} , \quad (9)$$

which already for $l=0$ is satisfied to within 15% and for $l=1$ within 3%. Thus the coupling element is nearly independent of l, which in the "sudden" limit gives the following solution [7] to Eq. (3)

$$c_{l,l_0}=J_{l-l_0}(p) , \quad (10)$$

where $J(p)$ is a Bessel function and where the index m has been dropped. The argument is given by [7]

$$p(t)=\frac{-2}{\hbar}\sqrt{\tfrac{1}{2}}\, d^{1}_{1,0}(\beta)eE_0\int_{-\infty}^{t} dt' r(t')g(t')\sin(\omega t') . \quad (11)$$

This expression follows as mentioned from the solution of Eqs. (3) in the sudden limit. Improvements can be obtained by introducing in Eq. (11) an energy correction factor $\exp(i\Delta Et/\hbar)$ (see Ref. [8]). Inserting in the equations for S and T we obtain

$$S(t)=\sum_{l} l(l+1)J^{2}_{l-l_0} \quad (12)$$

and

$$T(t)=\frac{1}{p}\sum_{l}(l-l_0)J^{2}_{l-l_0} , \quad (13)$$

which for $l_0=0$ yield

$$S(t)=\frac{p^2}{4}+\frac{p^2}{2}(J_0^2+J_1^2)-\frac{p}{2}J_0J_1$$

and

$$T(t)=\frac{p}{2}(J_0^2+J_1^2)-\tfrac{1}{2}J_0J_1 .$$

The following three equations are solved as a function of time:

$$\dot{r}=\frac{p_r}{\mu} , \quad (14)$$

$$\dot{p}_r=-\frac{e^2}{r^2}+\frac{\hbar^2}{\mu r^3}S(t)+eE_0g(t)\sin(\omega t)T(t) , \quad (15)$$

$$\dot{p}=\frac{eE_0}{\hbar}g(t)r(t)\sin(\omega t) , \quad (16)$$

where we have used $m=0$ in Eq. (4). The expectation value of the dipole moment is obtained as

$$\langle d\rangle=\frac{e}{2}r(t)T(t) . \quad (17)$$

Thus we have to solve just three coupled differential equations for a number of randomly selected initial variables of $r(t_0)$.

RESULTS

We consider excitation of the hydrogen atom by a 1064-nm laser field with a field strength of 1 or 10 TW/cm^2. The field is turned on linearly during the first 5 optical cycles. The optical generated harmonics are studied through the Fourier transform of the expectation value of the dipole moment, i.e.,

$$D(\omega)=\left|\frac{1}{T}\int_0^T dt\langle d\rangle\exp(i\omega t)\right|^2 , \quad (18)$$

where the analysis is started (i.e., $t=0$) after the initial period of 5 optical cycles and T is $(N-5)2\pi/\omega$, where N is the total number of optical cycles. The Fourier transform is furthermore taken as an average over a number of trajectories with randomly selected initial values of the distance r. The values are selected using the squared quantum-mechanical wave function for the $1s$ electron as the distribution function. The momentum p_r can then be calculated by conservation of energy, i.e.,

$$p_r=\pm\left[2\mu\left(E_{1s}+\frac{e^2}{r}\right)\right]^{1/2} , \quad (19)$$

where the sign is selected randomly.

Comparing the "spectra" in Figs. 1 and 2 obtained with 50 and 200 trajectories, respectively, shows that we obtain "converged" spectra to within 1% with just 50 trajectories. The peaks, however, are not clearly resolved after

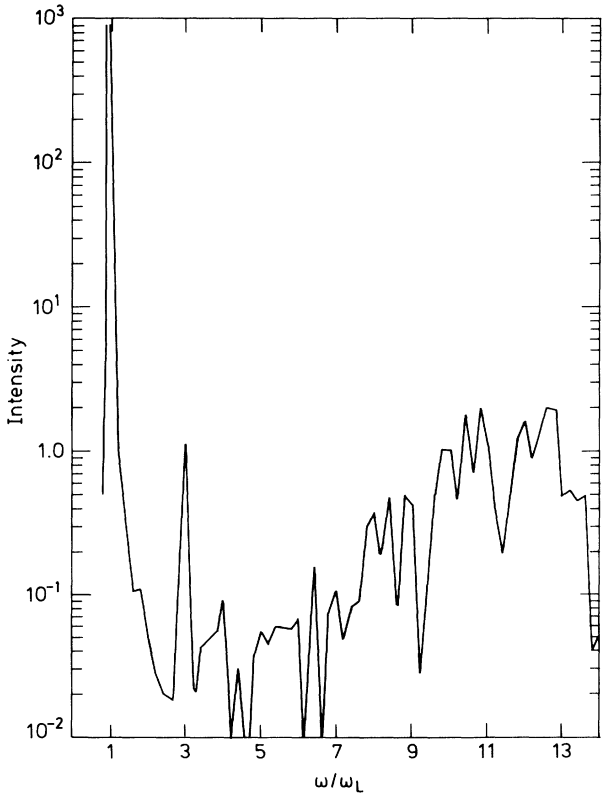

FIG. 1. Harmonic spectrum (18) (in arbitrary units) obtained with a 10 TW/cm^2 1064-nm laser field operating for 20 optical cycles. 50 Monte Carlo trajectories were used.

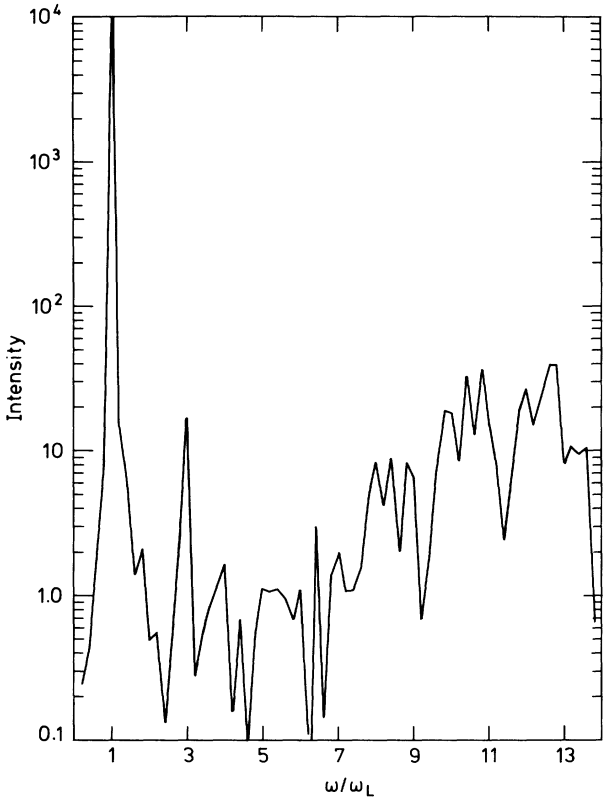

FIG. 2. Same as Fig. 1 but with 200 trajectories.

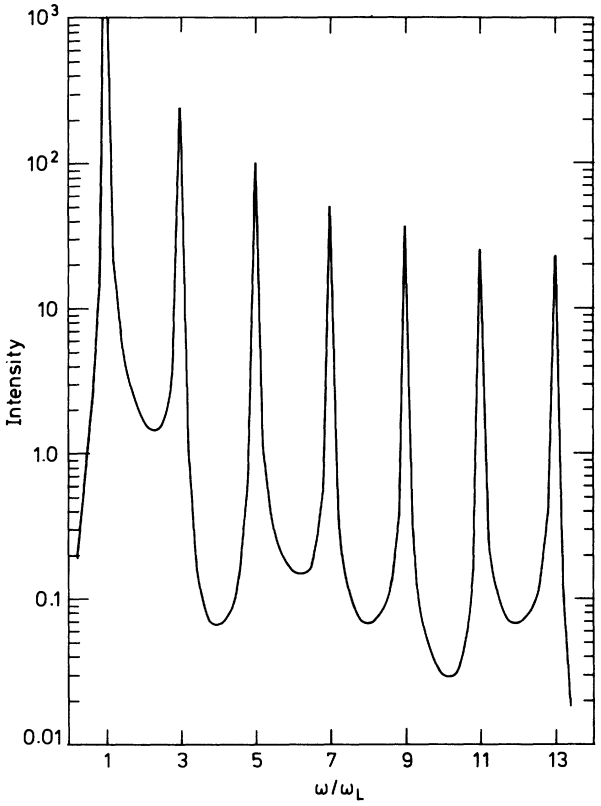

FIG. 4. Harmonic spectrum (18) obtained with a 1 TW/cm^2 1064-nm laser field applied for $N=100$ cycles. 50 trajectories were used.

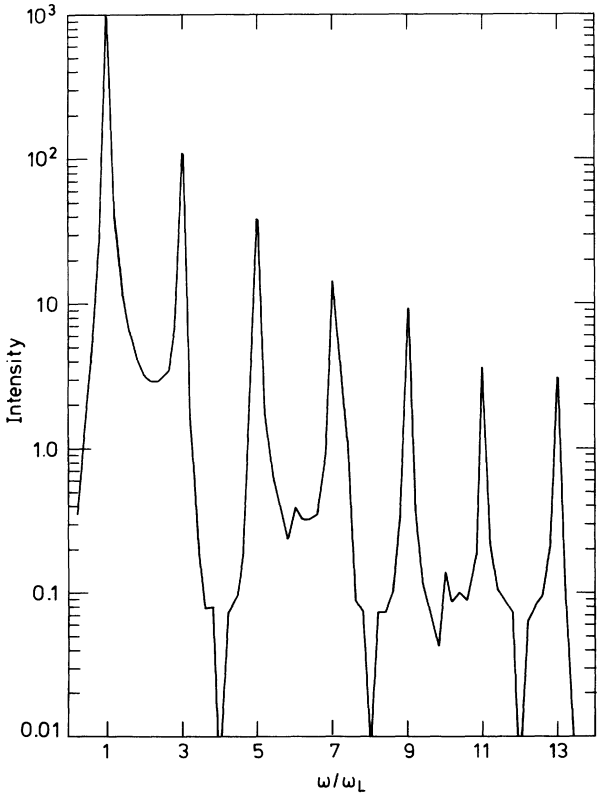

FIG. 3. Harmonic spectrum (18) obtained with a 10 TW/cm^2 1064-nm laser field applied for $N=40$ cycles. 50 randomly selected trajectories were used.

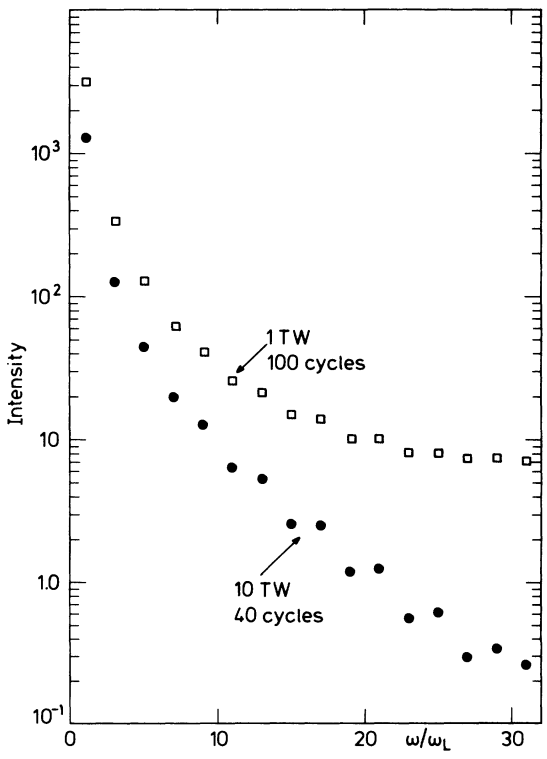

FIG. 5. Intensity of the harmonic spectrum generated up to the 31st harmonic for the two cases studied in Figs. 3 and 4 (arbitrary units).

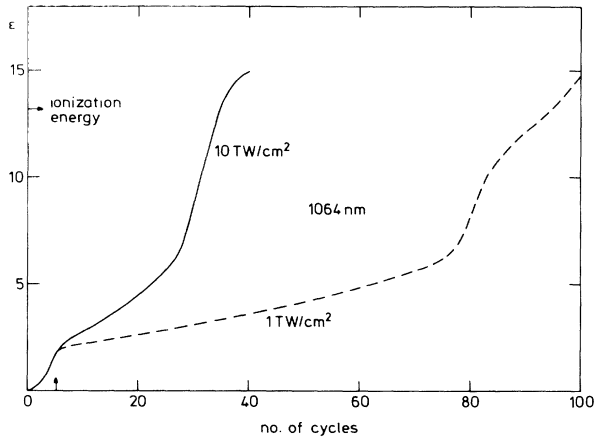

FIG. 6. Energy (in units of 100 kJ/mole) transferred to the hydrogen atom as a function of the number of optical cycles the laser field is applied for a 1 and 10 TW/cm^2 1064-nm laser field. The field is turned on using a linear ramp for the first 5 cycles (indicated by an arrow). The ionization energy is also indicated.

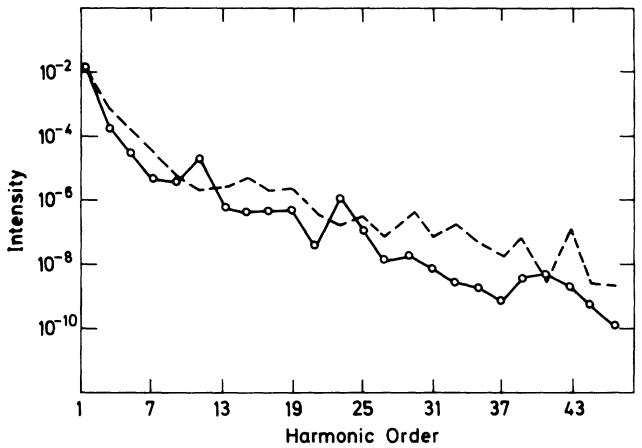

FIG. 7. Comparison of quantum mechanical (o) [4] and semiclassical (dashed line) intensities (arbitrary units) obtained using a 100 TW/cm^2 1064-nm laser pulse for 20 cycles. The curves are shifted to match the harmonic order $n=1$.

$N=20$ cycles. Figure 3 shows the spectrum after 40 cycles with a 10 TW/cm^2 field strength. In Fig. 4 the intensity is lowered to 1 TW/cm^2 and the spectrum is shown after 100 optical cycles using the "last" 95 for the analysis of the dipole moment. Figure 5 shows intensity of the peaks up to the 31st harmonic generated by the field. We notice that all the peaks are as well resolved as shown in Figs. 3 and 4. Figure 6 shows the energy pumped into the hydrogen atom as a function of the number of cycles the laser is operated. We notice (see Fig. 7) that the magnitude of the generated harmonics compare well with those obtained by Kulander [4]. The fact that the intensity of the higher-order harmonics are overestimated could be a consequence of the sudden approximation used when solving Eqs. (3). As mentioned above it is easy to introduce an approximate correction for this or alternatively solve Eqs. (3) numerically. This possibility will be considered in a subsequent paper.

We have not attempted to obtain ionization yields, although this is in principle possible. It would in the present method require the definition of a critical distance r^*, for which the atom could be considered ionized, and also the propagation of many trajectories, in order to obtain good statistics.

CONCLUSION

We have presented a simple semiclassical method for treating laser excitation of atoms in the presence of an intense laser field. We expect the method to be reliable for excitation processes involving multiphoton processes, where the excitation pathway is less sensitive for quantum selection rules in the principal quantum number (n). By treating part of the Hamiltonian quantally (in the large quantum number limit) it is possible in the sudden limit to obtain an analytically solvable time-dependent Schrödinger equation. The method can easily be extended to atoms with more than one electron.

ACKNOWLEDGMENTS

This research was supported by the Danish Natural Science Research Council. Part of the work was carried out while one of the authors (G.D.B.) visited the Universite de Paris-Sud.

[1] R. M. Potvliege and R. Shakeshaft, Phys. Rev. A **40**, 3061 (1989); S.-I. Chu, Comput. Phys. Commun. **63**, 265 (1991).

[2] G. Bandarage, A. Marquet, and J. Cooper, Phys. Rev. A **41**, 1744 (1990).

[3] F. H. M. Faisal and P. Scanzano, Comput. Phys. Commun. **63**, 265 (1991).

[4] K. C. Kulander, Phys. Rev. A **35**, 445 (1987); J. L. Krause, K. J. Schafer, and K. C. Kulander, Chem. Phys. Lett. **178**, 573 (1991); P. L. DeVries, Comput. Phys. Commun. **63**, 95 (1991).

[5] G. D. Billing, J. Chem. Phys. **65**, 1 (1976).

[6] D. M. Brink and G. Satchler, *Angular Momentum* (Clarendon, Oxford, 1968).

[7] G. D. Billing, J. Chem. Phys. **62**, 1480 (1975).

[8] M. H. Alexander and A. E. DePristo, J. Chem. Phys. **83**, 1499 (1979); C. Nyeland and G. D. Billing, Chem. Phys. **60**, 359 (1981).