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Multiphoton ionization of hydrogen by a strong multimode field

S. Basile, F. Trombetta, and G. Ferrante
Istituto di Fisica dell'Università, Via Archirafi 36, 90123 Palermo, Italy

R. Burlon and C. Leone
Dipartimento di Energetica e di Applicazioni di Fisica, Viale delle Scienze, 90128 Palermo, Italy

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Multiphoton ionization of hydrogen atoms by a strong multimode field when several final continuum states are populated is treated theoretically within a model based on the S-matrix formalism. The field is taken in dipole approximation, with zero bandwidth. Field-dressed Coulomb wave functions are used for the electron final states. Ionization rates are calculated for different numbers of photons absorbed in excess of the minimum number required to go into the continuum. In general, the calculated quantities are in good qualitative agreement with the corresponding experimental observations.

This paper is concerned with the multiphoton ionization of hydrogen atoms by a strong multimode laser field, when the ejected electrons have absorbed more photons than the minimum number required to go into the continuum. This kind of process is currently thoroughly investigated, and is commonly known as above-threshold ionization (ATI) or excess-photon ionization (EPI). The experimental results for the energy spectra of ejected electrons consist of a series of equally spaced peaks centered at the energies

$$\varepsilon_s=[n_0+s]\hbar\omega-I_0,\ \ s=0,1,2,\ldots, \tag{1}$$

where $n_0=(I_0/\hbar\omega)+1$ and s is the above threshold photon number; I_0 is the field-free ionization energy ($\cong 13.6$ eV for hydrogen); $[x]$ denotes the integer part of x.

Most of the experiments have been performed on rare-gas atoms;[1-3] but recently measurements on atomic hydrogen have also been reported.[4,5]

On the theoretical side, a number of different treatments can be found to qualitatively explain the experimental phenomenology.[6] Even if one simplifies to the essential the dynamics of the process, assuming that the electron goes from the initial to the continuum final state via the one-step absorption of a given number of photons, there are at least two points which deserve particular attention: a correct treatment of the electron final state and of the laser field.

First, in the continuum final state the electron interacts with the laser field and with the residual ion, so that its wave function should account for the joint influence of the radiation and of the Coulomb field. Recently an accurate field-dressed final-state wave function has been constructed for the problem under consideration here;[7] its leading term is found as a Coulomb function times the time-dependent part of the Volkov wave, and will be used below to represent the final electron state. This approximate wave function is expected to account for the dominant asymptotic interaction of the electron with the two fields, and in the recent past it has been used in a variety of laser-assisted bound-free transitions processes.[8] It amounts to assuming that the spatial part of the wave function is mainly controlled by the action of the residual ion static field, while the time-dependent part by the strong radiation field (taken in dipole approximation). Here, it amounts to consider only a direct, one-step process to populate a given continuum state, without intermediate transitions or couplings between different continua.

Second, very strong lasers are, as a rule, operated in multimode configuration, and it is generally recognized that most of the actual strong-field multiphoton experiments are performed at many intensities simultaneously.[1,4,5] Further, the laser is generally pulsed. As the ionization can occur during the rise of the pulse, the field amplitude should properly account for the temporal variations, as well as for the spatial inhomogeneities of the field.

In this paper we shall assume an exponential distribution for the laser intensity; it amounts to the assumption that the field correlation time is much larger than any other time involved in the process. Such a distribution, of course, does not account for the above-discussed temporal and spatial inhomogeneities of the field, but it is expected to partially account in some effective way for their averaging effects. Their rigorous inclusion, as well as that of the spectrum bandwidth effects, is left to further investigations.

In what follows, we use the S-matrix formalism to deal with the multichannel multiphoton ionization. The applicability of such a formalism is not obvious here, because one can not always use the notion of transition probability per unit time; this is particularly true, for instance, in regimes when the depletion of the ground state of the atomic target is relevant. In this sense, the S-matrix treatment, in its present form, is expected to hold particularly for short pulse lasers and, anyway, when the ionization rate times the pulse duration is much less than one.[9] Below, we shall see that our findings are in fact closer to the experimental results obtained by short laser pulses.

The transition probability per unit time, in the S-matrix formalism, is given by

$$w = \int \frac{d^3p}{(2\pi\hbar)^3} \lim_{T\to\infty} \left[|S_{\text{if}}|^2 \Big/ \int_{-T}^{T} dt \right] , \tag{2}$$

where[10]

$$S_{\text{if}} = (i\hbar)^{-1} \int_{-T}^{T} dt \langle \Psi_f(\mathbf{r},t) \,|\, e\mathbf{E}(t)\cdot\mathbf{r} \,|\, \psi_i(\mathbf{r},t)\rangle . \tag{3}$$

The initial and final states are

$$\psi_i(\mathbf{r},t) = (\pi a_0^3)^{-1/2} \exp(-r/a_0) \exp(iI_0 t/\hbar) , \tag{4}$$

$$\Psi_f(\mathbf{r},t) = \exp\left[\frac{-i}{2m} \int^t [\hbar\mathbf{k} + e\mathbf{A}(\tau)/c]^2 d\tau + \frac{ie}{\hbar c}\mathbf{A}(t)\cdot\mathbf{r} \right] \psi_{\mathbf{k}}^-(\mathbf{r}) , \tag{5}$$

$$\psi_{\mathbf{k}}^-(\mathbf{r}) = \exp(\pi\nu/2)\Gamma(1+i\nu) \times \exp(i\mathbf{k}\cdot\mathbf{r}) F(-i\nu, 1, -i(kr + \mathbf{k}\cdot\mathbf{r})) , \tag{6}$$

$$\nu = (ka_0)^{-1}, \quad \mathbf{A}(t) = -c\int^t \mathbf{E}(t')dt' . \tag{6a}$$

In Eqs. (6) and (6a), $\Gamma(x)$ and $F(a,b,c)$ are, respectively, the gamma and the confluent hypergeometric functions; a_0 is the Bohr radius and $\mathbf{E}(t)$ is the electric field. The exponential factor containing $\mathbf{A}(t)\cdot\mathbf{r}$ in Eq. (5) ensures the correct gauge consistency.

Assuming, first, a purely coherent field, let us write the electric field in the form $\mathbf{E}(t) = \mathbf{E}_0 \sin\omega t$; proceeding from (3) according to the usual rules, after the required amount of algebra we can write the "doubly differential" ionization rate as

$$d^2w/d\Omega d\varepsilon = \sum_{s=0}^{\infty} (d^2w/d\Omega d\varepsilon)_s , \tag{7}$$

where s denotes the number of photons absorbed beyond the minimum, required to reach the $(s+1)$th continuum and

$$(d^2w/d\Omega d\varepsilon)_s = \frac{e^2}{\hbar a_0} \frac{2^5}{\pi^2} \frac{I}{I_a} \frac{1}{1-\exp(-2\pi\nu)} \times |T_{n_0+s}(\mathbf{k},\mathbf{E}_0)|^2 \delta(\varepsilon + \Delta - \varepsilon_s) . \tag{8}$$

In Eq. (8), I is the field intensity (at this stage, well fixed), I_a is equal to 3.51×10^{16} W/cm^2, and

$$T_l(\mathbf{k},\mathbf{E}_0) = -\int_{-\pi}^{\pi} da \cos(a) B(\mathbf{k}, \mathbf{E}_g \sin a) f_l(a) \tag{9}$$

with

$$f_l(a) = \exp[ila - i(\Delta/2\hbar\omega)\sin 2a - i\lambda\cos a] , \tag{10}$$

$$\Delta = e^2E_0^2/(4m\omega^2) , \quad \lambda = e\mathbf{k}\cdot\mathbf{E}_0/(m\omega^2), \quad \mathbf{E}_g = e\mathbf{E}_0/(\hbar\omega) . \tag{10a}$$

From Eq. (8), Δ can be seen as a shift of the ionization threshold, due to the field, able to yield the ATI peak suppression;[7,11] $\hat{\mathbf{e}}$ and $\hat{\mathbf{k}}$ are the unit vectors in the direction of the field and of the ejected electron momentum. Further,

$$B(\mathbf{k}, \mathbf{E}_g \sin a) = -(16\pi a_0^4)^{-1} \int \{d^3r \exp[-r/a_0 - i(\mathbf{k} + \mathbf{E}_g \sin a)\cdot\mathbf{r}]\hat{\mathbf{e}}\cdot\mathbf{r} F(i\nu, 1, i(kr + \mathbf{k}\cdot\mathbf{r}))\} \tag{11}$$

is evaluated analytically (see Ref. 7). It can be shown that in the weak-field limit the ionization rates derived by Eq. (8) exhibit the I^{n_0+s} behavior; experimentally, this behavior extends to higher intensities only for short pulse lasers,[11] thus directly providing indications on the range of validity of our present treatment and confirming the expectations based on physical grounds.

In order to account for the field fluctuations within the assumption of a vanishing field spectrum bandwidth, we have now to average the "doubly differential" ionization rate, given in Eq. (8) for the case of a coherent field, over the chaotic field intensity distribution or, equivalently, over the thresholds distribution:

$$P(\Delta) = \exp(-\Delta/\langle\Delta\rangle)/\langle\Delta\rangle , \tag{12}$$

where, in terms of the mean-field intensity $\langle I\rangle$

$$\langle\Delta\rangle = e^2\langle E_0^2\rangle/4m\omega^2 = (2\pi e^2/mc\omega^2)\langle I\rangle . \tag{12a}$$

Averaging of Eq. (8) gives

$$\langle (d^2w/d\Omega d\varepsilon)_s\rangle = \int_0^\infty d\Delta (d^2w/d\Omega d\varepsilon)_s P(\Delta) = \frac{e^2}{\hbar a_0} \frac{2^5}{\pi^2\Delta_a} \frac{1}{1-\exp(-2\pi\nu)} \eta \exp(-\eta) |T_{n_0+s}(\mathbf{k},\tilde{E}_0)|^2 , \tag{13}$$

where

$$\Delta_a = (e^2/4m\omega^2)(e/a_0^2), \quad \tilde{E}_0^2 = (4m\omega^2/e^2)(\varepsilon_s - \varepsilon), \quad \eta = (\varepsilon_s - \varepsilon)/\langle\Delta\rangle . \tag{14}$$

As physically expected, Eq. (13) shows that the $(s+1)$th continuum state contributes energies from 0 to ε_s. To obtain the ionization rate to the $(s+1)$th continuum state outside the laser beam, we have now to integrate Eq. (13) over all the ejection energies inside the beam, i.e., from 0 to ε_s. Finally, following previous authors,[12] we assume that an electron, after the absorption of (n_0+s) photons will be detected, outside the laser beam, with an energy given by Eq. (1), whatever its energy is (in this case, between 0 and ε_s) inside the laser beam.

In Figs. 1 and 2 we show, for the first five continuum states and for several mean laser intensities, the ionization rates per unit time given by

$$\langle w_s\rangle = \int_{4\pi} d\Omega \int_0^{\varepsilon_s} d\varepsilon \langle (d^2w/d\Omega d\varepsilon)_s\rangle . \tag{15}$$

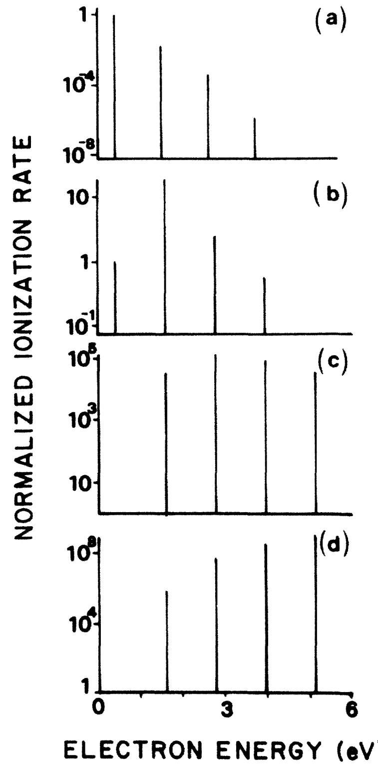

FIG. 1. Photoelectron energy spectra, normalized to the first peak; the energies are in eV; the photon energy is equal to 1.17 eV. The laser mean intensities are (in W/cm²): (a) 10^{11}; (b) 7.5×10^{11}; (c) 2.5×10^{12}; (d) 10^{13}.

For weak fields, the low-energy ATI peaks are dominant, and the perturbative I^{n_0+s} dependence on the intensity is recovered, together with the $(n_0+s)!$ enhancement typical of a weak chaotic field with respect to a purely coherent one. Though no peak is suppressed (contrary to the case of a purely coherent field), at increasing intensities the envelope of the peaks broadens and shifts to larger energies, recalling the observations of different experiments.

Figure 2 shows the ionization probabilities $\langle w_s \rangle$ for several continuum states versus the mean-field intensity [or, equivalently, versus $\langle \Delta \rangle$, see Eq. (12a)]. It is evident

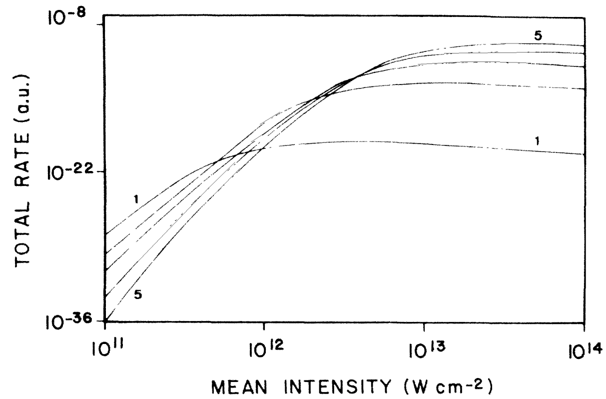

FIG. 2. Ionization rates (in atomic units) vs the laser mean intensity (in W/cm²). The numbers on the curves denote the final continuum states; other parameters as in Fig. 1.

that the peak inversion, shown in Fig. 1, is due to a nonperturbative saturation of the ionization probabilities: The smaller the continuum state energy, the lower the intensity at which the saturation appears. It is worth remarking that this behavior is here a result of an *ab initio* calculation, while in Ref. 13 it is obtained only thanks to an *ad hoc* assumption. It must be noted that this behavior is peculiar of a multimode field (see Ref. 7 for the case of a coherent field).

In conclusion, we have analyzed the above-threshold ionization of atomic hydrogen by a chaotic laser field of vanishing bandwidth within an S-matrix formalism. The ionization probabilities exhibit a nonperturbative saturation as functions of the field intensity, leading to the experimentally observed peak inversion in the photoelectron energy spectra. The results are encouraging in that the present S-matrix treatment has been found to reproduce qualitatively well important experimental features, thus demonstrating that it may serve as a useful basis for further improvements and calculations of other physical quantities. The S-matrix formalism to treat multiphoton ionization has also been used widely before.[9,10] Nevertheless, it is believed that we have given a treatment which is an improvement over previous works in many respects, and well fitted to give a definite answer on the merits and limits of the S matrix for the physical problem at hand.

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