Kinematics of multiphoton ionization in a steady laser beam

Marvin H. Mittleman
Physics Department, The City College of the City University of New York, New York, New York 10031
(Received 15 September 1983)

Previous results are extended to obtain the ionization potential of an arbitrary atom in an arbitrarily polarized single-mode steady-state laser. The results come from energy conservation and ideas associated with the ponderomotive potential. They are used to discuss the width of the energy distribution of electrons obtained from multiphoton ionization.

I. INTRODUCTION

One of the central problems of the theory of laser-atom interactions is the question of what happens to the ionization potential (IP) of an atom inside a laser field. We shall see that the answer to the question depends on the definition of the IP, which is not unique in the presence of the field.[1] The answer to the question will shed some light on the electron spectra observed in multiphoton ionization experiments. This note will deal only with the problem of an atom moving slowly into a single-mode steady-state classical laser beam. Experiments have so far been done with pulsed beams which may yield different electron distributions.

The problem has been treated previously in a restricted way. Muller, Tip, and Van der Wiel[2] have treated a model atom in a circularly polarized laser beam and have obtained results for the local ionization potential (LIP, defined below) which are similar to the results given here. Tip and Muller[3] have given arguments which indicate that similar results apply for a hydrogen atom in a circularly polarized laser. The results derived here apply for an arbitrary atom in an arbitrarily polarized field and depend on little more than energy conservation. They are obtained in Sec. II and used in Sec. III to discuss the energy distribution of electrons from multiphoton ionization.

II. DERIVATION OF THE LOCAL IONIZATION POTENTIAL

Consider an atom in its ground state which moves slowly into a steady-single-mode classical electromagnetic (laser) field. The atom can be described classically as a particle which moves in a potential, $\Delta W_0^{(a)}(I(R))$ which is the shift of the ground-state energy level of the atom. This is a function of the field intensity, which is in turn, a function of the position of the atom in the beam. The average velocity of the atom inside the field is related to that outside the field $\vec{\nabla}(\infty)$ by energy conservation

$$\tfrac{1}{2}M_a[\vec{\nabla}(\infty)]^2=\tfrac{1}{2}M_a[\vec{\nabla}(R)]^2+\Delta W_0^{(a)}(I(R)) \quad . \tag{2.1}$$

The ionization event is assumed to take place at R at which point the local ionization potential is $X(I(R))$. This is defined as the minimum energy that must be supplied to the atom to lift an electron to the continuum. The energy is supplied by absorption of N photons of frequency ω so energy conservation at ionization gives ($\hbar=1$)

$$\tfrac{1}{2}m[\vec{v}(R)]^2+\tfrac{1}{2}M_i[\vec{\nabla}'(R)]^2=\tfrac{1}{2}M_a[\vec{\nabla}(R)]^2 + N\omega-X(I(R)) \quad . \tag{2.2}$$

Here $\vec{v}(R)$ and m are the electron velocity and mass, $\vec{\nabla}'(R)$ and M_i are the ion velocity and mass, and N is the number of photons absorbed. Momentum conservation yields

$$m\vec{v}(R)+M_i\vec{\nabla}'(R)=M_a\vec{\nabla}(R)+N\vec{k} \quad , \tag{2.3}$$

where $\vec{k}$ is the laser photon momentum. The electron is expelled from the laser beam by the ponderomotive potential of the beam which we assume to be monotonically decreasing toward the edge of the beam[4] and slowly varying (on a laboratory scale). The conservation of energy relation for this is

$$\tfrac{1}{2}m[\vec{v}(R)]^2+U_p^{(e)}(I(R))=\tfrac{1}{2}m[\vec{v}(\infty)]^2 \quad , \tag{2.4}$$

where $U_p^{(e)}$ is the ponderomotive potential[5] acting on the electron. Its precise form depends upon the polarization of the field

$$U_p^{(e)}=n\frac{e^2E^2(R)}{4m\omega^2} \quad , \tag{2.5}$$

where E is the electric field amplitude and $n=1$ or 2 for linear and circular polarizations, respectively. The ion is expelled from the beam by a polarization potential but we must also account for the shift of its binding energy due to the field. The energy conservation relation is

$$\tfrac{1}{2}M_i[\vec{\nabla}'(R)]^2+U_p^{(i)}(R)+\Delta W_0^{(i)}(I(R))=\tfrac{1}{2}M_i[\vec{\nabla}'(\infty)]^2 \quad , \tag{2.6}$$

where $U_p^{(i)}$ is the ponderomotive potential acting on the ion and $\Delta W_0^{(i)}$ is the shift of the ground-state energy of the ion due to the field. The difference between initial and final energies is

$$\Delta E=\tfrac{1}{2}m[\vec{v}(\infty)]^2+\tfrac{1}{2}M_i[\vec{\nabla}'(\infty)]^2 - \tfrac{1}{2}M_a[\vec{\nabla}(\infty)]^2+W_0^{(i)}(0)-W_0^{(a)}(0) \quad , \tag{2.6}$$

which can be rewritten as

$$\Delta E=N\omega-X(I)+U_p^{(e)}(I)+U_p^{(i)}(I)+\Delta W_0^{(i)}(I) -\Delta W_0^{(a)}(I)-W_0^{(a)}(0)+W_0^{(i)}(0) \quad . \tag{2.7}$$

 ©1984 The American Physical Society

The fact that the Hamiltonian describing the system is periodic in time results in the statement[6] that ΔE must be an integer times ω. Therefore, $X(I)$ (modω) is given by

$$X(I)-X(0)=U_p^{(e)}(I)+U_p^{(i)}(I)+\Delta W_0^{(i)}(I)-\Delta W_0^{(e)}(I)\ , \tag{2.8}$$

where $X(0)=W_0^{(i)}(0)-W_0^{(a)}(0)$. Finally, the requirement that $X(I)|_{I=0}=X(0)$ allows us to drop the modω restriction on (2.8). The pondermototive potential does not depend on the internal structure of the particle, only on its net charge and mass. We therefore obtain from (2.5)

$$U_p^{(i)}=\frac{m}{M_i}U_p^{(e)}\ . \tag{2.9}$$

The shifts of the ground-state energies of the atom and ion are difficult to calculate exactly as a function of I. However, they are typically small compared to $U_p^{(e)}$ and can be obtained by a second-order perturbation theory as

$$\Delta W_0^x=-\frac{e^2}{2}\sum_n\frac{W_{n0}^x}{(W_{n0}^x)^2-\omega^2}\times\langle 0^x|\vec{\mathrm{E}}_+\cdot\vec{r}\,|n^x\rangle\langle n^x|\vec{\mathrm{E}}_-\cdot\vec{r}\,|0^x\rangle\ , \tag{2.10}$$

where the superscript x can be (a) or (i) denoting atom or ion. Here the electric field has been decomposed as

$$\vec{\mathrm{E}}=\tfrac{1}{2}(\vec{\mathrm{E}}_+e^{-i\omega t}+\vec{\mathrm{E}}_-e^{i\omega t}) \tag{2.11}$$

and $W_{n0}=W_n-W_0$. Implicit in the entire discussion, and in (2.10), is the assumption that no resonances occur so that $W_{n0}^x\neq\omega$.

Equation (2.8) shows that the LIP is essentially increased by the ponderomotive potential $U_p^{(e)}$, which is of the order of 1 eV for $\omega\sim 1$ eV at an intensity of about 10^{13} W/cm^2. The requirement that $N\omega-X(I)$ be positive can then eliminate the lowest value of N which would be present in the absence of this effect. A result similar to (2.8) was obtained by Muller *et al.*[2] and they used it to explain the suppression of the lowest value of N ($N=11$) in the experiment of Kruit *et al.*[7] in Xe.

Since the ionization potential is intensity dependent there is more than one possible definition[1] of the "ionization potential." If it is defined as the minimum energy required to ionize, then it is intensity or position dependent and given by the LIP. However, if it is defined from the energy of the electron outside the beam, which is the experimentally observed quantity, then it is $X(0)$ which is the ionization potential. The latter seems to be preferable on these grounds.

III. ELECTRON SPECTRUM

The experimentally observed quantity is the distribution of the energy of the electrons which have absorbed N photons outside the beam

$$\epsilon_N=\tfrac{1}{2}m[\vec{v}(\infty)]^2\ . \tag{3.1}$$

We may use Eqs. (2.1)–(2.4), (2.8), and (2.9) to obtain as a function of the initial energy of the atom, $E_a=\frac{1}{2}M_aV^2(\infty)$ and the point at which the ionization occurs, R, which enters through the intensity $I(R)$. After some algebra and the neglect of relativistic corrections arising from the photon momentum term in (2.3), the result is

$$\epsilon_N=\frac{M_i}{M_a}[N\omega-X(0)-\Delta W_0^{(i)}(I)+\Delta W_0^{(a)}(I)]-\frac{m}{M_a}[E_a-\Delta W_0^{(a)}(I)](1-2\mu^2)+2\mu\left[\frac{m}{M_a}[E_a-\Delta W_0^{(a)}(I)]\left(\frac{M_i}{M_a}[N\omega-X(I)]-\frac{m}{M_a}[E_a-\Delta W_0^{(a)}(I)](1-\mu^2)\right)\right]^{1/2}\ , \tag{3.2}$$

where $X(I)$ is given by (2.8) and $\mu=\hat{V}(R)\cdot\hat{v}(R)$. The dependence on the ponderomotive potential $U_p^{(e)}$ is weak. It enters in (3.2) through $X(I)$ which is multiplied by the small factor m/Ma. Its primary dependence is canceled due to the fact that the LIP contains $U_p^{(e)}$ so the ionized electron has its energy diminished by this amount but it reacquires this energy from its expulsion from the beam by $U_p^{(e)}$ (This fact has been pointed out by Muller *et al.*[2]) The remaining sources of the width for the electron energy are (1) the dependence of the energy shifts of the ground states of the atom and ion on the intensity, (2) the dependence on the direction of the electron velocity at the ionization event and (3) the dependence on the initial energy of the atom. All are small effects. The first is small since the shifts of the ground states themselves are small, and the effect is made even smaller by the fact that ϵ_N must be averaged over the probability of producing the ionization by N photons. Typically this is a very steep function of I so only the very high values of I near the center of the beam will contribute appreciably. The last two are associated with the translational velocity of the atom when the ionization takes place. This is usually small and this is the reason for the appearance of the small factor m/Ma is (3.2) multiplying the terms containing E_a and μ.

The cancellation of $U_p^{(e)}$ depends crucially upon the fact that a steady-single-mode laser has been treated here. Experiments are so far done with pulsed, and possibly multimode, lasers. In those cases (2.8) is still expected to hold provided that the laser amplitude changes slowly on the atomic time scale. However, Eqs. (2.1), (2.4), and (2.6) may not hold and then the cancellation of $U_p^{(e)}$ that occurs for the steady laser may no longer occur. This will be more fully discussed subsequently.

ACKNOWLEDGMENTS

This research was supported by U.S. Office of Naval Research under Contract No. N00014-76-C-0014.

[1]Extensive discussions on this subject with Dr. M. J. Van der Wiel are gratefully acknowledged.

[2]H. Muller, A. Tip, and M. J. Van der Wiel, J. Phys. B (to be published).

[3]A. Tip and H. Muller (private communication).

[4]See, for example, Marvin H. Mittleman, *Introduction to the Theory of Laser-Atom Interactions* (Plenum, New York, 1982).

[5]The kinematical effect of the ponderomotive potential on a free electron has been discussed by L. S. Brown and T. W. B. Kibble, Phys. Rev. 133, A705 (1964) and is reviewed in Ref. 4.

[6]J. H. Shirley, Phys. Rev. B 138, 979 (1965); this is no more than an application of Floquet theory to quantum mechanics. It can also be viewed in a second-quantized picture of the laser field as the remark that energy is transferred in units of the photon energy.

[7]P. Kruit, J. Kimman, H. G. Muller, and M. J. Van der Wiel, Phys. Rev. A 28, 248 (1983).