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ENERGY SPECTRUM OF ELECTRONS FROM MULTIPHOTON IONIZATION

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We discuss the multiple peaks observed in the energy spectrum of electrons resulting from multiphoton ionization of atoms by lasers. When the laser intensity is large enough for the ponderomotive force to result in appreciable broading of the peaks we show that the shape of the broadened peaks contains useful information. We show the multiphoton ionization probability as a function of laser intensity can be obtained but that the free-free cross sections which are in principle also obtainable are probably not obtainable in practice.

1. *Introduction*

The subject of multiphoton ionization by intense lasers has been of experimental interest for some time but it is only recently that experiments[1-3] have been performed which yield information on the energy spectrum of the electrons. Compton et al.[1] have seen two peaks in the spectrum which they identify as five photon resonant ionization of He. The peaks were interpreted as the probabilities of leaving the residual Xe^+ in either the $^2P_{1/2}$ or $^2P_{3/2}$ state since the energy spacing between the peaks (1.6 eV) agreed with the energy separation of these two states. Somewhat earlier Agostini et al.[2] measured the electron distribution in the non-resonant multiphoton ionization of Xe with a much more intense laser and also saw two peaks. These were identified as six and seven photon ionization, respectively, since the peaks were separated by the energy of a single photon (2.34 eV). No evidence of the multiplicity due to more than one possible final state of Xe^+ was found but since the first experiment was a resonant ionization and the second non-resonant this is not necessarily a contradiction. Finally, Kruit at al.[3]. measured the electron distribution in the resonant five photon ionization of He. They saw the doublet structure that corresponds to the fine structure of He observed by Compton. But they also saw this structure repeated at electron energies larger by $\hbar\omega$ and $2\hbar\omega$ than the energy of the lowest doublet, the phenomena observed in the Agostini experiment. These were identified as $(5 + 1)$ and $(5 + 2)$ photon ionization.

Compton's experiment was performed with a peak laser intensity which was too small for the ponderomotive force to affect the electrons. The same is true with Kruit's experiment. But Agostini's, performed with intensities about 10^4 times higher (of the order of 10^{13} W/cm^2) has observed forces which broadened the peaks in the electron distribution. If the spatial distribution of the laser intensity at its focus is known then the shape of the broadened low energy peaks can be used to obain information on the multiphoton ionization probabilities as a function of laser intensity. This is discussed in the next section.

The upper peak, due to absorption of an extra photon will also be broadened and this can in principle be used to extract information on the one photon inverse bremsstrahlung cross section. Gontier et al.[4] have discussed this second peak and pointed out that it arises from two separate (non-interfering) effects. The first, which they called *above-threshold-ionization*, (*ATI*), results from an absorption of the last photon by the electron while it is in the field of its parent ion. The second, *inverse-bremsstrahlung-process* (*IBP*) ariess from the electron's collision with another atom (or ion) as it travels out of the laser beam from its point of origin. The processes are, at least in principle, distinguishable by different pressure dependences so that we shall assume that they can be separately measured. The first of these, *ATI*, is the more interesting. It represents absorption of one more photon than is necessary to cause ionization. This process can happen in many different and interfering ways but experience has shown[5] that the dominant path is the almost on-shell one. That means that the transitions which most closely conserve energy in the intermediate states tend to dominate the total transition probability. Such a transition is available here since the absorption of the last photon can take place as a free-free transition in the field of the parent ion. Looking at the ionization this way, a simple analysis shows that the second peak in the Agostini experiment is unlikely to contain much of this first process, (*ATI*).

The mean time, T, between photon arrivals at the atom being ionized is given by

$$T = \frac{\hbar\omega}{I\sigma}$$

where $\hbar\omega$ is the photon energy, I the laser intensity and σ some cross section for absorption of the photon to form the second peak by *ATI*. If v is the velocity of the electron in the lower peak then vT will be the distance that the electron will move before the absorption of the last photon. If that distance is much larger than ϱ, the range of the potential around the ion in which the absorption can take place, then the absorption is an unlikely event. We can crudely evaluate this ratio for the conditions of this experiment obtaining

$$\frac{vT}{\varrho} \cong 25 \frac{a_0^3}{\sigma \varrho}$$

where a_0 is the Bohr radius. Reasonable values of σ and ϱ yield a result greater than unity, indicating that the electron has most likely left the influence of its parent ion before the *ATI* can occur.

The second process depends upon the *IBP* cross section and in the last section we show how the shape the second peak can in principle be used to extract information concerning the free-free cross section as a function of electron energy and laser intensity. However, the details appear to be too compex for the method to be useful.

2. *Analysis of the lower peak*

We assume that the electrons are collected at a small rectanguar hole in the containment vessel of dimension a, in a direction perpendicular to the laser beam and length b, along the beam. The hole is a distance d from the focus of the beam whose radius is R, as in Fig. 1. We assume that $d \gg a \gg b$ and that b is sufficiently small so that variation of the laser intensity along the beam direction can be neglected. (This restriction is removed below). Fig. 2. shows a plane perpendicular to the laser beam. At a point (r_0, Φ_0, z_0) an electron is ejected in the azimuthal direction ψ_0 with a z component of velocity v_z. There is a range of the angle ψ_0, called $\Delta\psi_0$, for which the electron will reach the collector. It can be obtained by integration of the classical equations of motion of an electron moving in a potential $V(r)$ which is taken to be the ponderomotive potential[6]:

$$V(r) = \frac{e^2}{4m\omega^2} \vec{E}_L^2(r) = kI(r) \tag{2.1}$$

where $\vec{E}_L(r)$ is the elecctric field of the laser, $I(r)$ is its intensity, assumed to be azimuthally symmetric, and $k = \frac{\pi e^2}{mc\omega^2}$. These equations yield

$$\Phi - \Phi_0 = \int_{r_0}^{r} \frac{L \, dr}{\sqrt{2} r^2 \left(E_0 + V(r_0) - E_z - V(r) - \frac{L^2}{2r^2} \right)^{1/2}} \tag{2.2}$$

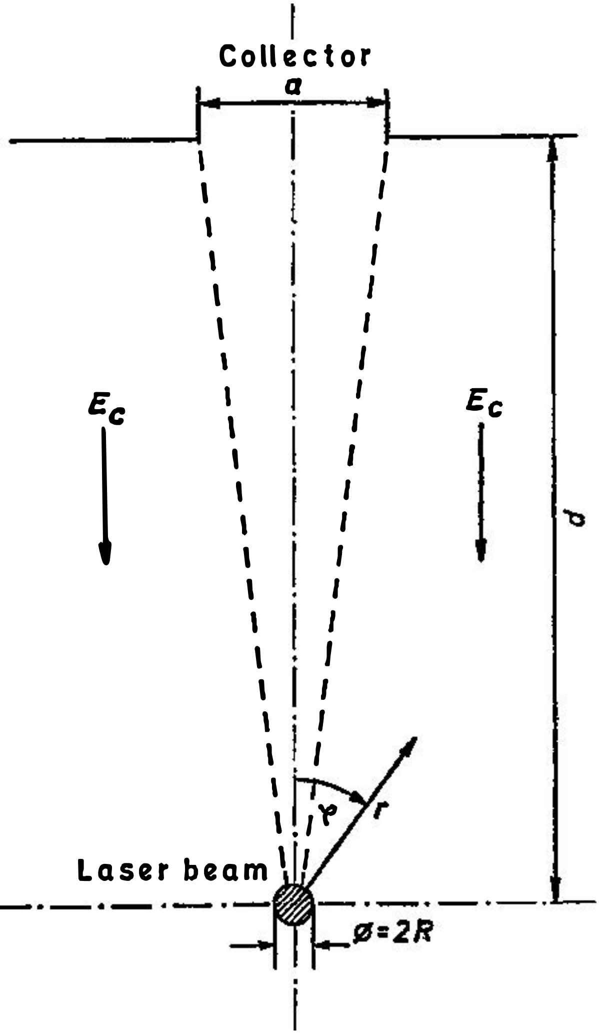

Fig. 1. The focused laser beam in the containment vessel for the analysis of the multiphoton ionization from the energy spectrum of electrons.

where L is the angular momentum of the electron about the center of the laser beam

$$L = mv_{\perp}\, r_0 \sin (\psi_0 - \Phi_0) \tag{2.3}$$

E_0 is the initial energy of the electron,

$$E_0 = \nu \hbar \omega - B \tag{2.4}$$

and

$$E_z = \frac{1}{2} m\, v_z^2. \tag{2.5}$$

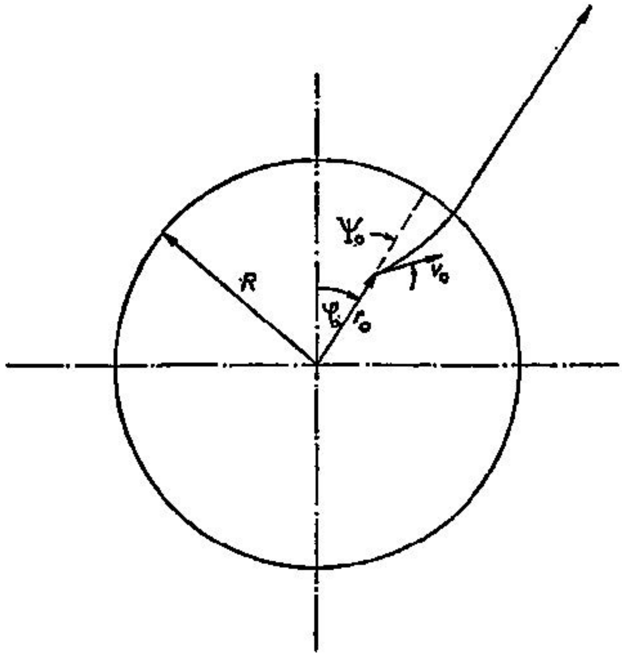

Fig. 2. Geometry in the laser beam.

Here ν is the number of photons absorbed during the ionization and B the binding energy of the electron with account being taken of the residual state of the ion. Then $v_\perp$ and v_z are related by

$$E_0 = \frac{1}{2} m (v_\perp^2 + v_z^2) = \frac{1}{2} m v_M^2. \tag{2.6}$$

An electron which is collected will pass through the point $r = \infty$, $\Phi = 0$ (see Fig. 1) with an allowable range in Φ given by

$$\Delta\Phi = \frac{a}{d} \ll 1. \tag{2.7}$$

This will map into an allowable range in ψ_0 which will be collected. This range can be obtained from (2.2) as

$$\Delta\psi_0 = \Delta\Phi \left(\frac{\partial f}{\partial \psi_0}\right)^{-1} \tag{2.8}$$

where

$$f(r_0, \Phi_0, \psi_0, v_z) = \int_{r_0}^{\infty} \frac{\mathrm{d}r\, L}{\sqrt{2}\, r^2 \left(E_0 + V(r_0) - E_z - V(r) - \frac{L^2}{2r^2}\right)^{1/2}} = -\Phi_0 \tag{2.9}$$

The right hand side of (2.8) is to be considered a function of (r_0, Φ_0, v_z) with ψ_0 eliminated by solving (2.9) as $\psi_0 = \psi_0 (r_0, \Phi_0, v_z)$. Electrons, born at a point z_0 with z component of velocity v_z may or may not reach the opening in the chamber in order to be counted. But since we are approximating the laser by a beam which is uniform in z, the entire system (beam plus chamber aperture) can be considered to be periodic in z with period b. Then for every electron (z_0, v_z) which is not collected there is one at $(z_0 - b, v_z)$ which is. Then the aperture in effect, will collect all electrons originating at z_0 for all v_z and no other electrons.

Let $P(I, \psi_0)$ be the probability of ionization per unit time per atom, at the point (r_0, Φ_0, z_0) with electron velocity specified by ψ_0, v_z, (Fig. 2). Then the total rate of collection of electrons with energy between E and $E + \mathrm{d}E$ is

$$nbr_0 \, \mathrm{d}r_0 \int\limits_{-V_M}^{V_M} \mathrm{d}v_z \int\limits_0^{2\pi} \mathrm{d}\Phi_0 \, P(I, \psi_0) \, \Delta\psi_0 (r_0, \Phi_0, \psi_0, v_z) \tag{2.10}$$

where v_M is given in (2.6), and n is atomic density. This can be identified with the detection rate per unit energy integrated over all possible v_z. The energy of the detected electron can be related to its point of origin by

$$E = E_0 + k\, I(r_0) \qquad 2.11)$$

and if $I(r_0)$ is a monotonically decreasing function of r_0 then it can be inverted to give $r_0(I)$ and

$$\mathrm{d}r_0 = \frac{\mathrm{d}E}{k} \left(\frac{\mathrm{d}I}{\mathrm{d}r_0} (r_0(I)) \right)^{-1} . \tag{2.12}$$

These can be assembled to give

$$\frac{1}{2v_M} \int\limits_{-V_M}^{V_M} \mathrm{d}v_z \frac{1}{2\pi} \int\limits_0^{2\pi} \mathrm{d}\Phi_0 \, P(I, \psi_0) \, \Delta\psi_0 (r_0, \Phi_0, \psi_0, v_z) = \frac{k}{2\pi\, nb} \frac{N(E)}{r_0(I)} \frac{\mathrm{d}I(r_0(I))}{\mathrm{d}r_0} \tag{2.13}$$

where $N(E)$ is the experimental counting rate per unit energy range and ψ_0 is considered to be a function of $r_0(I), \Phi_0, v_z$ from (2.9). This is an integral equation for P, the quantity of theoretical interest in terms $N(E)$, the measured spectrum. Its inversion would require extensive numerical analyses which, we show, can be avoided. Before doing so we briefly discussed it qualitatively.

* The only dependence of P upon the direction of the velocity of the ionized electron is through ψ_0, $\cos \psi_0 = \hat{v} \cdot \hat{E}_L$. This is so because the orientation and alignment of the initial state of the ion are not observed. Consequently, sums over the magnetic quantum numbers of these states must be performed in obtaining P. This erases all angular dependence upon $\vec{v}$ except tha described here.

If the multiphoton ionization rate is a rapidly rising function of I, as seems likely in the Agostini experiment, then the most electrons will be created at the region of the greatest intensity. This region will produce electrons of greatest energy since the (repulsive) ponderomotive potential is greatest there. Consequently we expect the broadened (lower) peak of the distribution function to have its maximum near the high energy end. This is not the case in the Agostini experiment, so there must be other broadening mechanisms at work there.

The complexities encountered above can be eliminated by using a small collecting electric field, E_c, (Fig. 1) which is large enough to collect all electrons formed in the multiphoton ionization process. However, it should be too small to distort the distribution of the photo-electrons. This is expressed by

$$2ReE_c \ll V(0) = kI(0) \tag{2.14}$$

where $I(0)$ is the peak laser intensity. These conditions are easily satisfied. We can also allow for a variation of the laser intensity along the direction of the laser beam, i. e. allow for a real focussing of the beam. This complicates the dynamics of the electrons and the geometry of the collection but the complication can be avoided by making b, the size of the aperture, large enough. This is easily seen in the following way: The laser intensity will decrease as the distance from the focal plane increases. The ionization probability, a rapidly rising function of intensity, will therefore be smaller the greater the distance from the focal plane. If b is large enough then essentially all the ionized electrons will be collected, since few will be produced far from the focal plane.

We can obtain a relation between the ionization probability and the counting rate in a manner analogous to the one used in obtaining (2.13). There are several differences:First, the use of the collecting field means that electrons with any ψ_0 are collected. Second, the laser intensity is now a function of r_0 and z_0. The integral over r_0, z_0 can be converted to an integral over I, z_0, with a Jacobean $\partial I(r_0, z_0)/\partial r_0$. We again assume that I is a monotonic function of r_0 for each z_0. The result is

$$\frac{n}{k}\int_{-\infty}^{\infty} dz_0 \frac{r_0(I, z_0)\, \Theta(I(O, z_0) - I)}{\left|\frac{\partial I(I, z_0)}{\partial r_0}\right|} 2\pi \int_0^{2\pi} d\psi_0\, P(I, \psi_0) = N(E(I)) \tag{2.15}$$

where $E(I)$ is obtained from (2.11) and the Heavyside function Θ arises from the fact that there is a maximum value of I for each z_0 which is $I(O, z_0)$. Eq. (2.15) relates the total ionization rate (integrated over directions of the electron) for a given laser intensity to the experimental counting rate, which is the purpose of this section. The collecting field has eliminated the geometric complications of (2.13).

3. *Analysis of the upper peak*

We shall assume that the different pressure dependences of the *ATI* and the *IBP* processes have been used to isolate each and we shall discuss only the inverse bremsstrahlung process contribution to the upper peak.

The spectrum of the upper peak is obtained in the following way: An atom is ionized at the point (r_0, Φ_0, z_0) where the laser intensity is again assumed to be $I(r_0, z_0)$. The electron moves under the influence of the ponderomotive potential and the collecting field to some point in the laser field, (r_1, Φ_1, z_1), where it collides with another atom and absorbs a laser photon. It then escapes from the laser field and is collected (by E_c) with the energy

$$E = E_0 + \hbar\omega + kI(r_0). \tag{3.1}$$

We have assumed that another collision is unlikely and this is born out by the experimental absence of another peak. An analysis similar to that given in the preceding section leads to

$$N_1(E) = \frac{n^2}{k} \int_{-\infty}^{\infty} \frac{\mathrm{d}z_0\, r_0(I)}{\left|\frac{\partial I}{\partial r_0}\right|} \Theta(I(0, z_0) - I) \int_{r_0}^{\infty} \mathrm{d}l \int_0^{2\pi} \mathrm{d}\Phi_0 \cdot$$
$$\cdot \int_0^{2\pi} \mathrm{d}\psi_0\, P(I, \psi_0) \frac{1}{2v_M} \int_{-V_M}^{V_M} \mathrm{d}v_z\, \sigma_T(\vec{p}_l, \vec{E}_L). \tag{3.2}$$

Here Θ is the unit step function, $\sigma_T(\vec{p}_l, \vec{E}_L)$ is the total cross section for one-photon induced inverse bremsstrahlung and $\vec{p}_l$ is the momentum of the electron just before this collision. $\vec{p}_l$ is a function of the point at which the electron is initally produced, its velocity at that point and the point of the second collision. The magnitude of $\vec{p}_l$ is given by

$$\frac{p_l^2}{2m} = E_0 + k(I(r_0, z_0) - I(r_1, z_1)) \tag{3.3}$$

but its direction must be obtained from a solution of the equation of motion of the electron. Finally the integral over dl follows the path of the electron determined from these equations. This is an extremely complicated equation which can in principle be used to obtain σ_T once P has been obtained by methods of the second section.

We can illustrate the method by a simple example. We neglect any z dependence of the laser intensity as in the first part of the preceding section and we assume that E_0, (2.4), is very small. We also note that (2.14) allows us to neglect the effect of the collecting field on the electron while it is inside the laser beam. The electron will then be expelled radially from its point of creation. Moreover, $\vec{p}_l$ will then depend upon the conditions of the ionization event only through $I(r_0)$, which is held constant in (3.2). The integral over dl will then be a simple integral over the radial line strating from r_0. The integrals over v_z, ψ_0, z_0 and Φ_0 can then be done and related to $N(E - \hbar\omega)$ via (2.15). (The definition of E in this section, (3.1), is different from that of the preceding one, (2.11)). The result is

$$\frac{N_1(E)}{nN(E-\hbar\omega)} = \int_0^{2\pi} \frac{d\Phi_0}{2\pi} \int_{r_0}^{\infty} dr_1\, \sigma_T(\vec{p}_l(r_1); \vec{E}_L(I)) \tag{3.4}$$

where $\vec{p}_1$ is directed radially, with magnitude and direction

$$p_l(r_l) = (2mk(I - I(r_1)))^{1/2},\ \hat{p}_l \cdot \hat{E}_L = \cos\Phi_0$$

and E is related to I by (3.1). If the dependence of σ upon the laser intensity is known and simple then it is possible to invert this equation to get σ_T but in general the method described in this section seems to be too complicated to be useful for extracting σ_T from the shape of the second peak.

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References

1) R. N. Compton, J. C. Miller, A. E. Carter and P. Kruit, Chem. Phys. Lett. **71** (1980) 87;
2) P. Agostini, F. Fabre, D. Mainfray, D. Petite and N. K. Rahman, Phys. Rev. Lett. **42** (1979) 1127;
3) P. Kruit, J. Kimman, M. J. Van Der Weil, Proc. XII ICPEAC, Gatlinburg Tenn., (1981) 1058.
4) Y. Gontier, M. Poirier and M. Trahin, J. Phys. B. **13** (1980) 1381;
5) See for example B. Beers and L. Armstrong, Phys. Rev. A **12** (1975) 2447;
6) The formula for C (r) given here is 1/2 that of Ref. 2.

ENERGETSKI SPEKTAR ELEKTRONA IZ MULTIFOTONSKE JONIZACIJE

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Originalni naučni rad

Razmatramo višestruke pikove eksperimentalno viđene u energetskim spektrima fotoelektrona pri multifotonskoj jonizaciji atoma laserima. Kada je intenzitet lasera dovoljno veliki da ponderomotivne sile dovedu do merljivog širenja pikova pokazujemo da oblik pikova sadrži korisne informacije. Može se dobiti verovatnoća multifotonske jonizacije u funkciji laserskog intenziteta. Poprečni preseci slobodno — slobodnih prelaza se takođe mogu dobiti, mada za njih analiza izgleda suviše komplikovana da bi se primenila u praksi.

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