Experiments on multiphoton free-free transitions

A. Weingartshofer, E. M. Clarke, J. K. Holmes, and C. Jung
Department of Physics, St. Francis Xavier University, Antigonish, Nova Scotia, Canada
(Received 23 January 1979)

Multiphoton free-free transitions are detected in the scattering of electrons on argon atoms and on hydrogen molecules in the presence of an intense pulsed CO_2 laser field. The authors present measurements for several values of the incoming electron energy, the electron scattering angle, and the angle between the laser polarization vector and the electron momentum transfer. All observations are in qualitative agreement with a recent theoretical model based on a low-frequency approximation.

I. INTRODUCTION

In an electron-atom (molecule) scattering process the electron is accelerated and so it can emit electromagnetic radiation (bremsstrahlung). For electrons with an energy of the order of 10 eV the spontaneous emission of a photon with a measurable energy has a negligible probability. However, in a sufficiently intense-laser-field induced emission and absorption can become so strong, that even multiphoton processes can be observed, in which the scattered electron changes its energy by an integer multiple of the photon energy. Because in such transitions the electron proceeds from one continuum state to another continuum state, these processes are called free-free transitions (F-F transitions) and have recently been reviewed.[1,2]

The first observation of multiphoton F-F transitions was reported in 1977.[3] We have since continued these measurements with other values of the incoming electron energy, the scattering angle of the electron, the angle between the laser beam and the electron beam and with another target. As in Ref. 3, the measurements in this paper are still preliminary and qualitative. The purpose of our experiments is to gain experience in order to plan and carry out an extensive series of good quantitative measurements with an improved laser pulse.

Free-free cross sections are of importance to test the validity of many theoretical investigations about scattering processes in strong laser fields that have been published recently (see Refs. in Ref. 1) and, furthermore, F-F cross sections are also needed for such applications as laser heating of plasmas.[2,4]

II. APPARATUS

The scattering geometry of our experimental setup is shown in Fig. 1. The angles θ (scattering angle of the electron) and ψ (angle between $\vec{p}_i$ and the laser beam) are measured in the scattering plane, and throughout this paper clockwise rotation is indicated with a positive sign. The angle between the polarization vector $\vec{\epsilon}$ of the laser beam and the scattering plane of the electron is designated as ϕ and with one exception to be mentioned later, $\phi = 0$.

The electron gun and the detector were described in detail in Ref. 5. Our laser is a pulsed CO_2 laser (model Lumonics TEA-103-1) with a photon energy of 117 meV and an energy output of 15 J per pulse. The laser was operated in multimode optical configuration and the temporal shape of the pulse is shown in Fig. 2. Note that fast oscillations of the intensity are averaged out in this oscilloscope picture.

A fast digital counter was constructed having four digital display channels. The first three designated A, B, C sample equal and consecutive time slices (1.2 μsec each) of the scattered electrons in the presence of the laser pulse as shown in Fig. 2. The fourth channel D was delayed for a time long enough to ensure that the detector was counting scattered electrons without laser field and to improve the statistics this channel remained open for a longer length of time (10 $\times 1.2$ μsec). However, in the spectra we report the counts for an equivalent time as in channels A, B and C.

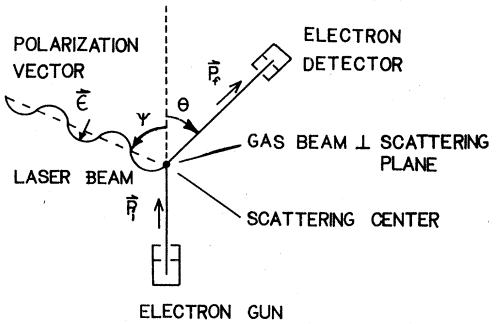

FIG. 1. Scattering geometry.

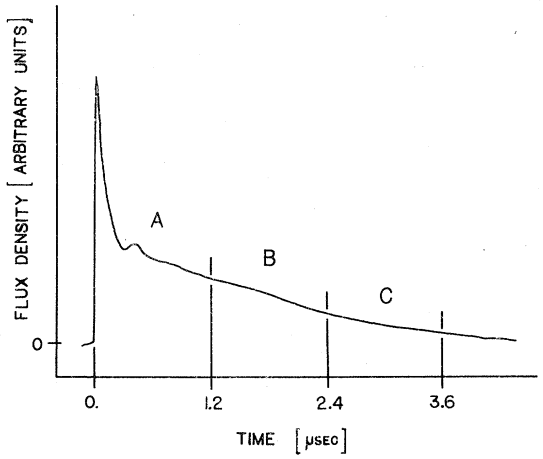

FIG. 2. Temporal shape of the laser pulse. Effect of different average fluxes in time intervals A, B, C was investigated with a fast counter as explained in main text.

Ar atoms and H_2 molecules were chosen as targets because we already had experience in handling these gases from former electron scattering experiments with the same apparatus (Refs. 5 and 6).

We have measured energy spectra of the scattered electrons with specific values of the angles θ, ψ and incoming electron energy E_i. For each spectrum the acceptance energy E_f of the detector was varied above and below the energy E_i. For every value of E_f the scattered electrons were counted successively in channels A, B, C and D for an equal number of laser shots. To confirm that the interaction was strictly due to the incoming electron beam with the laser pulse, we repeated some measurements by turning off the electron beam and leaving everything else as before. The accumulated counts were zero or negligible.

III. THEORY

For theoretical statements about multiphoton F-F transitions we will follow Ref. 7. It was shown there, that the cross section for an electron scattering in a pulsed laser field from the initial state with momentum $\vec{p}_i$ and energy $E_i = \vec{p}_i^{\,2}/2m$ into a final state with momentum $\vec{p}_f$ and energy $E_f = p_f^2/2m = E_i + n\hbar\omega$ is given by

$$\frac{d\sigma^n}{d\Omega}(E_i, \theta) \approx \frac{p_f}{p_i} R_n \frac{d\sigma^{el}}{d\Omega}(E_i, \theta), \tag{1}$$

where

$$R_n = 1/(T_f - T_i)\int_{T_i}^{T_f} dt\; J_n^2(p(t)), \tag{2}$$

and

$$\rho(t) = [eA(t)/mc\hbar\omega]\vec{\epsilon}\cdot(\vec{p}_f - \vec{p}_i). \tag{3}$$

J_n is the Bessel function of the first kind and order n, e is the electron charge, $A(t)$ is the time-dependent absolute value of the vector potential of the laser beam, m is the electron mass, c is the speed of light, ω is the laser frequency, and $d\sigma^{el}/d\Omega$ is the cross section for the corresponding electron-target scattering without laser field. The integration in (2) goes over a laser pulse or over the electronically selected time portion of the pulse.

In the derivation of (1) three assumptions or approximations have been made: (i) the interaction between the laser beam and the target has been neglected. This means that the target atom or molecule is not deformed by the presence of the laser beam. This approximation is good, if the photon energy $\hbar\omega$ is small compared to the target excitation energies and if the laser intensity is not too high, so that virtual or real excitation or even ionization of the target has a negligible probability; (ii) applicability of the soft photon approximation. This means that we can separate the electron-laser and the electron-target interaction, so that the laser acts on the electron only before and after the electron-target interaction. This approximation is good, if the effective range of the atomic (molecular) potential is much smaller than the laser wavelength and if the electron energy is higher than the photon energy. More information about low-frequency approximations is contained in Refs. 8–10. (iii) $d\sigma^{el}/d\Omega$ should depend only very weakly on the electron energy, so that for all n in (1, 2) which give a significant contribution, we have

$$\frac{d\sigma^{el}}{d\Omega}(E_i + n\hbar\omega, \theta) \approx \frac{d\sigma^{el}}{d\Omega}(E_i, \theta).$$

The e-Ar and the e-H_2 systems have sharp resonances (Refs. 5 and 6) but in our present experiment the energy width of the incoming electron beam is 60–150 meV (was changed for the various energy spectra) which is much broader than the widths of the resonances and therefore all resonance effects were smeared out and will be neglected in this paper and we assume, the condition (iii) is fulfilled.

The Bessel functions decrease rapidly as soon as the order becomes greater than the argument and so we expect a measurable signal only for such processes with $n \leq \max|\rho(t)|$. Because the energy flux density F of the laser beam changes strongly in time in each of the time intervals A, B, C (see Fig. 2), all oscillations of the Bessel functions are completely averaged out in (2) and

R_n behaves like the envelope of the Bessel functions. Some examples of this averaging results are shown pictorially in Ref. 7. Because the laser was run in multimode operation, there were also strong spatial and fast time fluctuations of the intensity that gave additional averaging.

Now we want to say something about the magnitude of ρ: for a flux of $F = 10^8$ watt/cm^2 of a CO_2 laser, an electron energy of 10 eV and angles $\theta = 180°$, $\psi = 90°$ and $\vec{\epsilon} \parallel \vec{p}_i$, $\rho \approx 5$; and ρ grows with the square root of F if all other quantities are kept fixed. In our experiment F was not quite 10^8 watt/cm^2 in the maximum and we expect that under the best angular conditions we can detect electrons having changed their energy by as many as 3 $\hbar\omega$ or 4 $\hbar\omega$. Considering that $A \propto \sqrt{F}/\omega$, we see that ρ is proportional to ω^{-2} for a given F. This is the reason why we have chosen an infrared laser.

The Bessel functions fulfil the sum rule

$$\sum_{n=-\infty}^{+\infty} J_n^2(x) = 1 \tag{4}$$

for all x and that leads to

$$\sum_{n=-\infty}^{+\infty} R_n = 1 , \tag{5}$$

and if $|p_f| \approx |p_i|$ for all n that give strong contributions to the sum, we get the following differential sum rule for F-F cross sections:

$$\sum_{n=-\infty}^{+\infty} \frac{d\sigma^n}{d\Omega}(E_i, \theta) \approx \frac{d\sigma^{el}}{d\Omega}(E_i, \theta) . \tag{6}$$

The differential sum rule says that the total number of electrons, summed up over all final energies, which go into a given direction, is independent of the properties of the laser beam and therefore is the same as in the absence of the laser beam.

One last point to be noticed in (1) is: The properties of the special target are contained only in $d\sigma^{el}/d\Omega$. R_n is completely independent of the target, which means, that for different targets the count rate ratios between the various final electron energies in the spectrum are always the same and only the count rates are proportionally changed. Consequently, we see that we get strong signals of F-F processes only for such values of θ and E_i, for which the elastic cross section is large.

IV. DEPENDENCE ON THE LASER FLUX

It follows from Eq. (3) that the argument of the Bessel functions varies with $\sqrt{F}$. In our experiment we did not change the energy output of the laser but instead our counter (see Sec. II) selected out the electrons that were scattered in the time intervals A, B, and C of the laser pulse shown in Fig. 2. In these 1.2 μsec intervals the average flux is appreciably different and spectra A, B, and C of Figs. 3 and 4 clearly show this dependence. Figure 3 applies to e-Ar scattering in the backward direction, while Fig. 4 is an example of e-H_2 scattering in the forward direction. Other parameters are indicated in the figures.

The spectra in the D channel of these figures show the electron energy distributions of the incoming electron beam without laser field. Because the elastic e-H_2 cross section is already

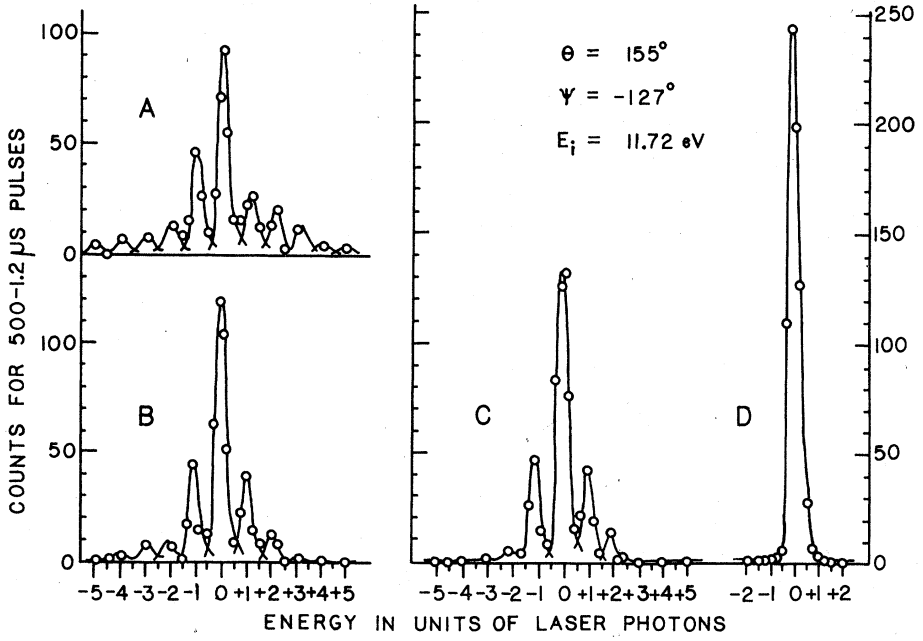

FIG. 3. Energy gain-loss spectra for e-Ar scattering showing dependence on laser flux F. Spectra A, B, C show count rates of electrons scattered in time intervals A, B, C of laser pulse in Fig. 2. Spectrum D measured without laser. ○ gives measuring points and solid line traces out multiphoton processes with each peak approximated with an outline of same shape as incoming electron beam (elastic peak).

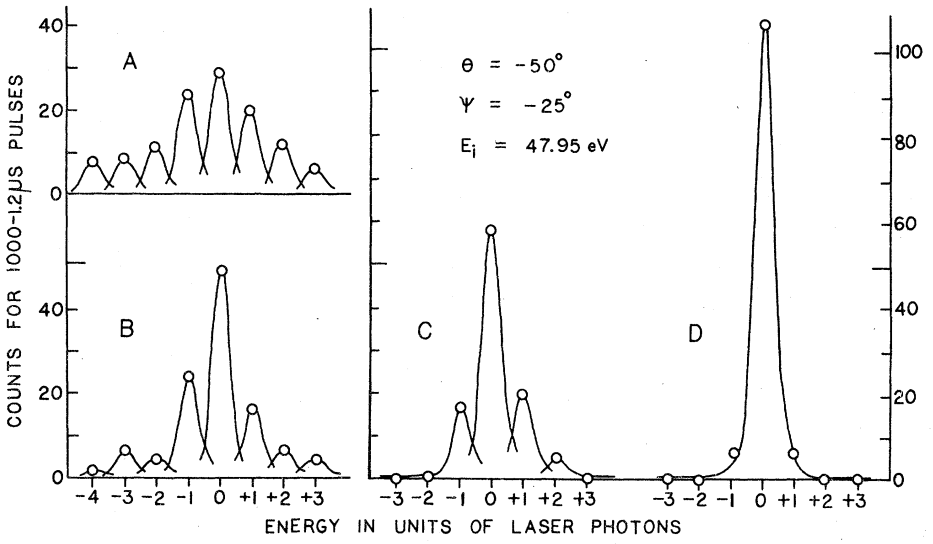

FIG. 4. Energy gain-loss spectra for e-H_2 scattering showing dependence on laser flux F. Other explanations as in Fig. 3.

rather small for an electron energy of $E_i = 47.95$ eV we had to increase the incoming electron current in order to get a sufficient count rate. Unfortunately this caused a broader energy distribution and a noticeable overlap of the various peaks in the energy spectra A, B, C.

Inspection of Figs. 3 and 4 reveals clearly that the sum rule (6) holds. Furthermore, comparing the energy gain-loss spectra in channels A, B, and C, we see that F-F processes producing greater energy changes in the scattered electrons have a lower probability as the laser intensity decreases. Because we do not know the exact time and space dependence of F for these measurements, it is not possible to give a quantitative theoretical interpretation of the count rates.

V. DEPENDENCE ON THE ENERGY OF THE INCOMING ELECTRONS

A second possibility of changing ρ and hence the scattered electron energy spectra is to vary the incoming electron energy or momentum. In Fig. 5 we show the energy gain-loss spectra for several values of E_i in e-H_2 scattering at angles $\theta = -50°$, $\psi = -25°$. The count rates are taken over the complete pulses, for instance, in spectrum e count rates of A, B, and C of Fig. 4 are summed up. For $E_i = 23.95$ eV in spectrum c we have only measured the count rates for positive n. For the spectra a, b, c, and d the width of the energy of the incoming electron beam is about 60 meV and so the different peaks in the spectra can be re-

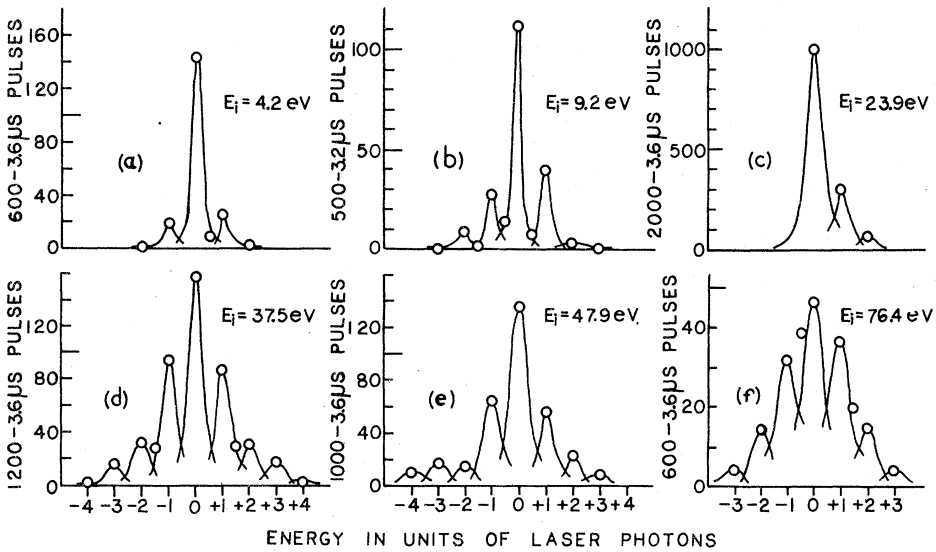

FIG. 5. Energy gain-loss spectra for e-H_2 scattering showing dependence on electron energe E_i. $\theta = -50°$, $\psi = -25°$. ○ measuring points and for solid line see Fig. 3.

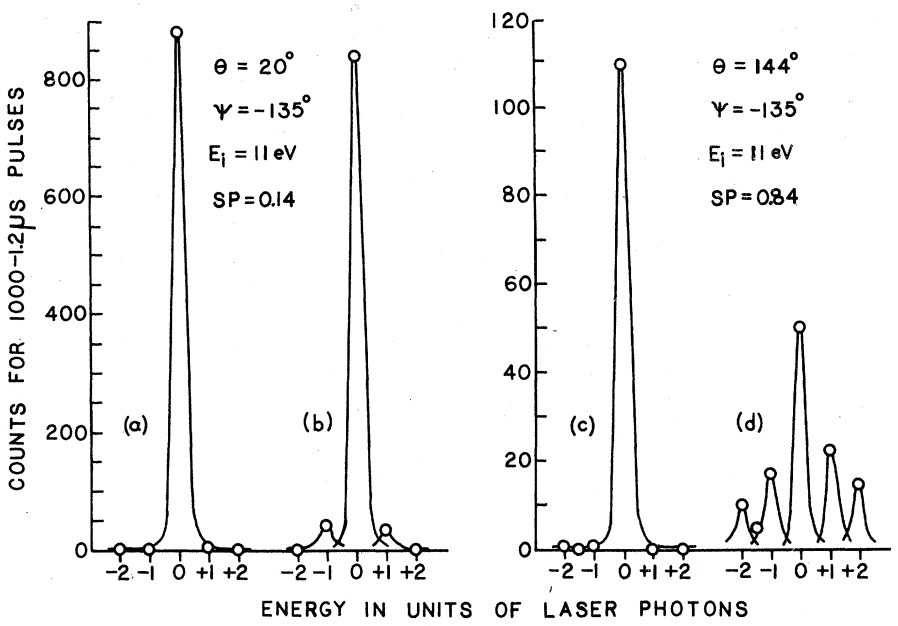

FIG. 6. Energy gain-loss spectra for e-Ar scattering showing dependence on electron scattering angle θ. (a) and (c) without laser, (b) and (d) with laser. $SP = \vec{\epsilon} \cdot (\vec{p}_f - \vec{p}_i)/2p_i$. ○ measuring points and for solid line see Fig. 3

solved. Because the elastic cross section for larger E_i values is small we had to increase the electron current. This resulted in a broader energy width for spectra e and f of about 90 meV and 130 meV respectively.

We can conclude from this figure that the strength of interaction between the electrons and the laser beam increases with electron momentum. The probability of emission and absorption of a higher number of photons increases as well with momentum.

VI. ANGULAR DEPENDENCE

Another possibility of varying ρ consists in modifying the scalar product $\vec{\epsilon} \cdot (\vec{p}_f - \vec{p}_i)$ in (3) by changing the angles θ, ψ, or ϕ. In Fig. 6 we see electron energy gain-loss spectra for two different values of θ keeping all other parameters constant. The target is argon and $E_i = 11$ eV. Each spectrum in Fig. 6 was measured for the same number of laser pulses (in b and d only the count rates of part A of the laser pulse are shown).

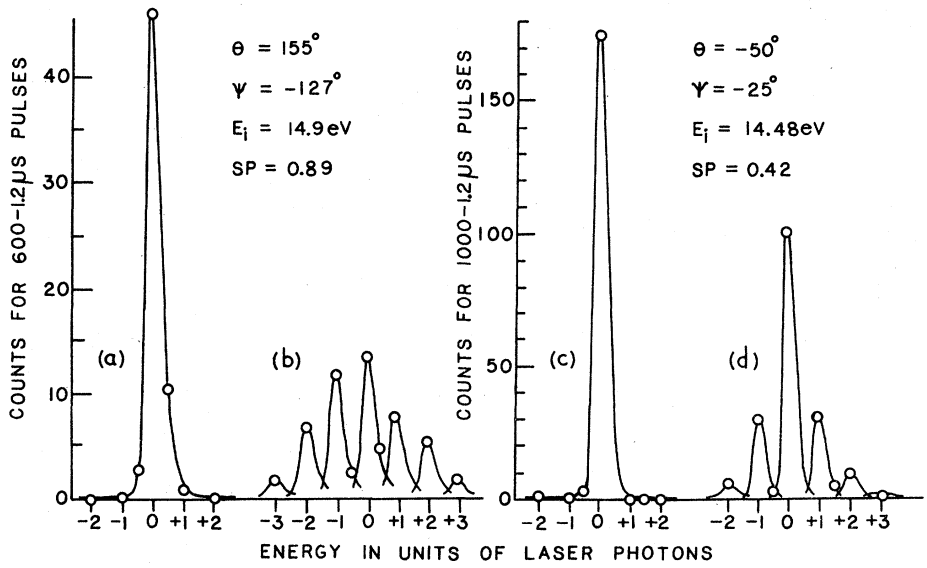

FIG. 7. Energy gain-loss spectra for e-Ar scattering showing dependence on electron scattering angle θ and direction of laser beam ψ. (a) and (c) without laser, (b) and (d) with laser. $SP = \vec{\epsilon} \cdot (\vec{p}_f - \vec{p}_i)/2p_i$. ○ measuring points, and for solid line see Fig. 3.

Comparison of spectra a and c gives us simply the angular dependence of the elastic scattering which is known (Ref. 5). For spectrum d the scalar product $\vec{\epsilon}\cdot(\vec{p}_f - \vec{p}_i)$ is nearly maximized and the relative probability of emission/absorption of photons compared to the 0-photon peak is much greater than in b, where this scalar product is small. In comparing a with b, or c with d we confirm again the validity of the differential sum rule (6).

In Fig. 7 we show results obtained by changing both angles θ and ψ. The target is argon and E_i is between 14 and 15 eV. Note the effect of the bigger scalar product in spectrum b as compared to d.

We also tried out the combination of angles: $\theta = 15°$ with $\psi = -85°$ and $\theta = 120°$ with $\psi = -30°$. In both cases the scalar product $\vec{\epsilon}\cdot(\vec{p}_f - \vec{p}_i) \approx 0$ and we did not detect any electrons having changed their energy. Finally we turned the polarization vector $\vec{\epsilon}$ of the laser so that $\phi = 90°$. In this case also, we could not detect any emission or absorption of photons.

VII. CONCLUSIONS

In principle there is yet another way of changing ρ: by varying the frequency ω. Unfortunately this cannot be done with our present experimental setup.

We do not have F-F measurements with Ar and H_2 for exactly the same values of θ, ψ and E_i, however we have compared spectra with similar values of these quantities. From these measurements we did not see any detectable differences in the quantities R_n by changing the target gases.

All the observed effects can be explained qualitatively with the theory given in Sec. III. We cannot check for quantitative agreement between experiment and theory at this time, because we do not know the exact laser flux distribution in space and time. To perform better quantitative measurements, we are now in the process of modifying the laser system to produce laser pulses with the following characteristics: (i) Single-mode TEM_{00} operation of the laser. This has the advantage that there are no fast oscillations of the flux in time and that the spatial distribution of the energy in the beam is smooth and uniform. All pulses should have the same temporal shape; (ii) a sufficiently high-energy output per pulse so that we can concentrate our laser beam onto a focal region that covers the complete electron-target interaction volume. This will ensure that all electrons, which are scattered at the same time, are scattered in approximately the same laser beam intensity; (iii) an energy-flux density of 10^8 watt/cm^2 in the focal region so that processes involving up to 4 or 5 photons can be detected.

If these conditions are fulfilled, it is possible to calculate the quantities R_n and together with the knowledge of the elastic scattering cross sections exact quantitative predictions of the F-F count rates can be made and compared with the experimental results.

ACKNOWLEDGMENTS

We are grateful to the Natural Sciences and Engineering Research Council of Canada for continuous financial support. We express our appreciation to the William and Flora Hewlett Foundation of Research Corporation for a generous grant. We also thank Professor G. Caudle and Professor H. Krüger for their advice and fruitful discussions.

[1] M. Gavrila and M. Van der Wiel, Comments At. Mol. Phys. 8, 1 (1978).
[2] S. Geltman, J. Res. Nat. Bur. Stand. 82, 173 (1977).
[3] A. Weingartshofer, J. K. Holmes, G. Caudle, E. M. Clarke, and H. Krüger, Phys. Rev. Lett. 39, 269 (1977).
[4] Y. Shima and H. Yatom, Phys. Rev. A 12, 2106 (1975).
[5] A. Weingartshofer, K. Willmann, and E. M. Clarke, J. Phys. B 7, 79 (1974).
[6] A. Weingartshofer, E. M. Clarke, J. K. Holmes, and J. W. McGowan, J. Phys. B 8, 1552 (1975).
[7] H. Krüger and C. Jung, Phys. Rev. A 17, 1706 (1978).
[8] F. E. Low, Phys. Rev. 110, 974 (1958).
[9] N. M. Kroll and K. M. Watson, Phys. Rev. A 8, 804 (1973).
[10] C. Jung and H. Krüger, Z. Phys. A 287, 7 (1978).