Electron-helium scattering in a 1.17 eV laser field: The effect of polarization direction

B. A. deHarak,¹ Benjamin Nosarzewski,^{2,*} Mahsa Siavashpouri,^{3,†} and N. L. S. Martin³

¹*Physics Department, Illinois Wesleyan University, P.O. Box 2900, Bloomington, Illinois 61702-2900, USA*

²*Department of Physics, Cornell University, 109 Clark Hall, Ithaca, New York 14853, USA*

³*Department of Physics and Astronomy, University of Kentucky, Lexington, Kentucky 40506-0055, USA*

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We report measurements of one-photon emission during the elastic scattering of electrons by He atoms through 90◦ in the presence of 1.17 eV photons from a Nd:YAG laser. The incident energy of the electrons was in the range 30–200 eV and the linear polarization direction of the laser was varied over 180◦ in the plane, perpendicular to the scattering plane, that contains the momentum transfer direction. Our results are perfectly consistent with the Kroll-Watson approximation. In particular, we see no evidence of free-free transitions when the polarization is perpendicular to the momentum transfer direction.

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I. INTRODUCTION

The elastic scattering of an electron by an atomic or molecular target, in the presence of a laser field, is known as laser-assisted free-free scattering, or simply free-free scattering $[1,2]$. For electrons of energy E_0 incident on a target *A*, and a laser field of frequency *ω*, there is the possibility of the absorption or emission of one or more photons

$$
A + e(E_0) + \mathcal{N}\hbar\omega \to A' + e(E) + \mathcal{N}'\hbar\omega, \tag{1}
$$

where $\mathcal{N}' = \mathcal{N} \pm n$, corresponds to the emission (+) or absorption (−) of *n* photons by the $A + e$ system and the final electron energy is $E = E_0 \mp n\hbar\omega$.

We recently reported free-free experiments in He over a range of incident-electron energies from 50 to 350 eV in the presence of 1.17 eV photons from a Nd:YAG laser [\[3\]](#page-4-0). Our experimental results were in good agreement with a theoretical prediction of the electron-energy dependence of the free-free signal using the semiclassical Kroll-Watson approximation (KWA) [\[4\]](#page-4-0). To our knowledge, these experiments were the first to use 1.17 eV photons to investigate the free-free process for elastic scattering, although Luan *et al.* [\[5\]](#page-4-0) investigated the inelastic scattering analog known as simultaneous electronphoton excitation, or SEPE [\[1\]](#page-4-0).

We have now extended our test of the KWA for 1.17 eV photons by measuring the free-free signal as a function of the linear polarization direction with respect to the momentum transfer direction. This is of interest because the experiments of Wallbank and Holmes [\[6–8\]](#page-4-0), using 0.117 eV photons from a $CO₂$ transversely excited atmospheric (TEA) laser, found free-free signals orders of magnitude larger than those expected when the laser polarization is almost perpendicular to the momentum transfer direction; the KWA predicts vanishingly small free-free signals for this geometry. All theoretical approaches have so far failed to explain these results; these are summarized in a recent paper by Morrison and Greene [\[9\]](#page-4-0). It has been suggested that double-scattering

events contributed to the free-free signal in the perpendicular direction [\[10\]](#page-4-0); this mechanism seems to have been ruled out by later experiments [\[11\]](#page-4-0).

The experiments of Wallbank and Holmes confined the measurements to those cases where the polarization direction lay in the scattering plane (formed by the incidentand scattered-electron directions), and was perpendicular, or almost perpendicular, to the momentum transfer direction. Our experiments differ from these experiments in that we measure the full range of polarization directions in a plane, perpendicular to the scattering plane, that contains the momentum transfer direction. Thus our measurements range from the case where we expect a maximum free-free signal to that where we expect zero signal.

Section II gives the KWA, and Sec. [III](#page-1-0) describes the apparatus and the geometry of our free-free measurements. Section [IV](#page-2-0) presents the results, and Sec. [V](#page-2-0) our conclusions.

II. THEORY

The KWA relates the free-free cross section $d\sigma_{\text{FF}}^{(n)}/d\Omega$, for absorption $(n < 0)$ or emission $(n > 0)$ of *n* photons, to the field-free elastic-scattering cross section $d\sigma_{el}/d\Omega$, by [\[4\]](#page-4-0)

$$
\frac{d\sigma_{\rm FF}^{(n)}}{d\Omega} = \frac{k_f}{k_i} J_n^2(x) \frac{d\sigma_{\rm el}}{d\Omega}.
$$
 (2)

Here k_i and k_f are the initial and final electron momenta, and J_n is a Bessel function of the first kind of order n , with argument

$$
x = -0.022\lambda^2 I^{1/2} E_i^{1/2} \frac{\hat{\epsilon} \cdot \mathcal{Q}}{k_i},\tag{3}
$$

where λ is the wavelength of the radiation in μ m, *I* is its intensity in GW/cm², $\hat{\epsilon}$ is the polarization direction, E_i is the incident electron energy in eV, and $Q = k_f - k_i$ is the momentum transfer.

The quantity *x* is a measure of the maximum number of photons expected to be absorbed or emitted in a free-free transition. It may be expressed in the alternative formulation

$$
x = \frac{eA_0}{m\hbar\omega}\hat{\epsilon} \cdot \mathbf{Q} \approx |n_{\text{max}}|,\tag{4}
$$

^{*}Present address: Department of Physics, Stanford University, California 94305, USA.

[†] Present address: Martin A. Fisher School of Physics, Brandeis University, 415 South Street, Waltham, Massachusetts 02453, USA.

FIG. 1. (Color online) The form of the free-free signal for onephoton emission $(n = 1)$ as a function of the angle θ between the laser polarization and the momentum transfer direction. The dashed line is a KWA calculation given by Eq. [\(2\)](#page-0-0) with $x = \cos \theta$. The solid line is the $\cos^2 \theta$ approximation given by Eq. (6).

where *e* is the charge, and *m* is the mass, of an electron, and *A*⁰ is the vector potential in a semiclassical description of the laser field. In the Appendix we give a simple classical derivation of this result.

In the limit of small x , the Bessel function may be approximated by the first term of a power series expansion and Eq. [\(2\)](#page-0-0) becomes

$$
\frac{d\sigma_{\rm FF}^{(n)}}{d\Omega} \approx \frac{k_f}{k_i} \left(\frac{1}{|n|!}\right)^2 \left(\frac{x}{2}\right)^{2|n|} \frac{d\sigma_{\rm el}}{d\Omega}.\tag{5}
$$

This equation shows that for $n = \pm 1$, and for a given laser intensity and incident electron energy, as the polarization direction is varied the free-free signal takes the form

$$
\left(d\sigma_{\text{FF}}^{(1)}/d\Omega\right)/(d\sigma_{\text{el}}/d\Omega) \propto (\hat{\boldsymbol{\epsilon}} \cdot \hat{\boldsymbol{Q}})^2 = \cos^2 \theta,\qquad(6)
$$

where θ is the angle between the polarization axis and the momentum transfer. In fact this relationship is almost true when *x* is not small, as can be seen in Fig. 1 where $\cos^2 \theta$ is compared with $[J_1(\cos \theta)]^2$, so that $x = 1$ when $\theta = 0$. The experiments performed below were carried out in this regime, and, because of the statistical uncertainties, our experimental data do not discriminate between the two forms.

III. EXPERIMENTAL METHOD

The free-free experiments were carried out using the electron spectrometer and Continuum Powerlite 9030 Nd:YAG laser described in our earlier work [\[3\]](#page-4-0). A schematic of the experimental setup for the present experiments is shown in Fig. 2. The electron spectrometer is a modified version of an apparatus previously used for $(e, 2e)$ studies $[12, 13]$. It consists of an unmonochromated electron gun (*H*), a scattered electron detector (*G*), both mounted on independent concentric turntables in the *xy* plane, and a single-bore gas nozzle to create the helium beam (F) . See $[3]$ for details of the spectrometer, data acquisition system, and data analysis.

The scattering geometry for the present experiments is as shown in the figure. The angle between the electron beam and the laser beam is 45◦, and the scattered electron detector is positioned to receive electrons elastically scattered through 90 \degree in the *xy* plane. The momentum transfer *Q* is in the \hat{y} direction, perpendicular to the laser beam direction.

The laser (*A*) has photon energy 1.17 eV ($\lambda = 1.06 \mu$ m), repetition rate 30 Hz, pulse duration ≈8 ns, and, in the present experiments, a pulse energy of between 0.15 and 0.2 J. The laser beam first passes through a rotatable $\lambda/2$ plate (B_1) set to select maximum horizontal polarization (\hat{y} in the figure), followed by a polarizing beam splitter cube (B_2) to reject any vertical (\hat{z}) component. The resultant pure horizontally polarized beam then passes through a second $\lambda/2$ plate (*C*), set in a computer-controlled rotatable mount, which is used to create the desired direction of polarization within the vertical *yz* plane that contains the momentum transfer \hat{Q} shown in the figure. (Thus horizontal polarization is parallel to \hat{Q} .) The laser beam enters the vacuum system (J) through an infrared window, and a lens (E) then focuses the beam to a diameter

FIG. 2. (Color online) The experimental layout for the present experiments. A: Nd:YAG laser, B1 : *λ/*2 plate, B2: polarizing beam splitter cube, C: *λ/*2 plate and computer controlled rotatable mount, D: polarizing beam splitter cube, E: lens, F: He nozzle, G: scattered-electron detector, H: electron gun, J: vacuum chamber, K: beam dump, L: power meter (for setup). The direction of the momentum transfer \hat{Q} is indicated.

0.75 mm in the interaction region (F) . The power meter (L) and polarizing beam splitter (*D*) were used to calibrate the fast axes of the two $\lambda/2$ plates.

Measurements were taken for repeated sequences of 24 fast axis directions covering 360◦ in 15◦ intervals. The computercontrolled $\lambda/2$ plate (*C*) was held at each direction for one hour while data were accumulated. Thus each 360◦ rotation corresponded to four complete cycles of six polarization

FIG. 3. (Color online) The free-free signal as a function of the angle θ between the laser polarization and the momentum transfer direction for incident-electron energies 30, 60, 120, and 200 eV, and the sum of all the results. The data have been scaled to give the best fits to $\cos^2 \theta$ (solid lines).

directions at 30◦ intervals. Note that the accurate angular calibration was done at the end of the experimental runs, which were carried out using a previous approximate calibration. Thus the final angles differ from our initial estimate by 7◦.

IV. RESULTS AND DISCUSSION

Free-free experiments, corresponding to one-photon emission, were carried out at incident-electron energies 30, 60, 120, and 200 eV. At each energy, measurements were made of the free-free signal as a function of $\theta = \cos^{-1}(\hat{\epsilon} \cdot \hat{\mathbf{Q}})$ covering a $180°$ range in 30° intervals. For all experiments the energy of a laser pulse was estimated to be between 0.15 and 0.2 J, corresponding to an instantaneous power of approximately 20 MW and an intensity of approximately 5 GW*/*cm² .

The results are shown in Fig. 3. The vertical error bars are the statistical uncertainties and the horizontal error bars are the systematic uncertainty of $\pm 2^\circ$ in θ , derived from an uncertainty of ±1◦ in the calibration of the fast axis of the *λ/*2 plate. The solid lines are $\cos^2 \theta$, and the measured free-free signals are plotted with a scale that gives the best fit. Only the statistical errors are used; the systematic errors are not incorporated in the fits, which had reduced chi-square values ranging from 0.8 to 2.8.

Given that the fits at all the individual energies are satisfactory, a slightly more rigorous test of the cosine-squared law is provided by summing all the results to obtain better statistics. This sum is shown at the bottom of the figure, with an accompanying fit to $\cos^2 \theta$, which has the somewhat low reduced chi-square value of 0.2.

We also tried fitting the function $A \cos^2 \theta + B$; a nonzero value of *B* corresponds to free-free transitions occurring when the laser polarization is perpendicular to the momentum transfer. The fits were no better than those shown, indicating that, to within our statistics, our results are compatible with zero transitions for the perpendicular case.

V. CONCLUSIONS

We have tested the KWA over the complete range of angles between the laser polarization and the momentum transfer direction, in a plane perpendicular to the scattering plane. Previous measurements by Wallbank and Holmes [\[6–8\]](#page-4-0) had only investigated the parallel and (nearly) perpendicular cases within the scattering plane, and found severe disagreements with the KWA predictions. We find the KWA, a first-order theory, gives a good description over the full range covered by our experiments. In particular we find no evidence for nonzero signals when the laser polarization is perpendicular to the momentum transfer, in contrast to the experiments of Wallbank and Holmes. This agreement with the first-order theory therefore rules out processes such as a double collision, which are included in a second-order theory. Such a theory allows the possibility of a collision with a He atom in which the incident electron scatters elastically from each of the two electrons in turn. For such an event the overall momentum transfer is $Q = Q_1 + Q_2$, where $Q_{1,2}$ are the momentum transfers for each part of the double collision. If a photon is absorbed or emitted during the first (or second) part of the double collision, it is possible for the overall momentum

transfer to be perpendicular to the laser polarization (i.e., $\hat{\epsilon} \cdot \mathbf{Q} = 0$), but with $\hat{\epsilon} \cdot \mathbf{Q}_1 \neq 0$ (and $\hat{\epsilon} \cdot \mathbf{Q}_2 \neq 0$), and therefore a nonzero free-free signal in the perpendicular direction. (Such double collisions for *inelastic* processes are known to be important in the electron-impact excitation of the doubly excited He $2\ell 2\ell'$ autoionizing levels, via $1s \rightarrow 2\ell$ and $1s \rightarrow 2\ell$; a second-order theory is vital for agreement with experiment [\[14\]](#page-4-0).)

We note, however, that Wallbank and Holmes measured the largest deviation from the KWA for two and three photon transitions. In fact, the second-order double collision process, described above, may be more important for those than for single-photon absorption or emission since there are more paths leading to a nonzero free-free signal in the perpendicular direction. For example, a two-photon absorption can occur in one part of the collision, or sequential single-photon absorption can occur in each part of the collision; these amplitudes add coherently and therefore significantly enhance the two-photon absorption cross section. We therefore plan to repeat our measurements to include two and three photon transitions and also to mimic their experiments more closely by rotating the laser polarization direction within the scattering plane.

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APPENDIX

What follows is similar, but not identical, to the classical derivation given by Kroll and Watson in Sec. [II](#page-0-0) of [\[4\]](#page-4-0). The basic difference is that we average over a cycle at the end of the calculation, rather than beforehand. We begin by examining an electron-atom collision in the presence of an oscillating spatially homogeneous electric field, and then extend the treatment to cover a classical electromagnetic wave.

It is well known that a free electron (charge *e* and mass *m*) cannot absorb energy from an oscillating electric field of frequency ω : For a classical field $\mathcal{E} = \mathcal{E}_0 \sin(\omega t)$ the momentum change, integrated over a complete cycle, for an electron is

$$
\overline{\Delta p} = e \int_0^{2\pi/\omega} \mathcal{E} dt = 0,
$$
 (A1)

and hence there is no change in kinetic energy. On the other hand, if the electron undergoes a collision with an atom in such a field then the electron can absorb (or emit) a quantum of energy. (The equivalent statement in quantum mechanics is that energy and momentum cannot both be conserved for the absorption by a free electron of a photon of energy $\hbar\omega$ and momentum $\hbar \omega/c$, whereas they *can* both be conserved in an $electron + photon + atom system.)$

A simple classical model can be used to find the energy absorbed (or emitted) by an electron undergoing elastic

FIG. 4. Schematic of an electron-atom collision in an oscillating electric field. k_1 is the electron momentum at the beginning of an oscillatory cycle. k_2 and k_3 are the momenta just before and after the collision at time t_c . k_4 is the momentum at the end of the cycle. See text for details.

scattering through an angle $\theta_{\rm sc}$ during one cycle of the field $\mathcal{E} = \pm \mathcal{E}_0 \sin(\omega t)$. We take $t = 0$ to correspond to the start of the cycle and the \pm takes into account the two possible behaviors of $\mathcal E$ as *t* increases from 0. Figure 4 shows a schematic of the process. At $t = 0$ the free electron has momentum k_1 . The collision occurs at $t = t_c$, where $0 < t_c$ $2\pi/\omega$. Just before the collision the electron has momentum

$$
k_2 = k_1 + \Delta p, \tag{A2}
$$

where the change in momentum is

$$
\Delta p = e \int_0^{t_c} \mathcal{E} dt = \pm \frac{e \mathcal{E}_0}{\omega} [1 - \cos(\omega t_c)] \hat{\epsilon}, \quad (A3)
$$

and $\hat{\epsilon}$ is the polarization direction of the field. Immediately after the collision the momentum is k_3 , with $|k_3|=|k_2|$ for elastic scattering. Finally, at $t = 2\pi/\omega$, the momentum is

$$
k_4 = k_3 + \Delta p', \tag{A4}
$$

with

$$
\Delta p' = e \int_{t_c}^{2\pi/\omega} \mathcal{E} dt = \mp \frac{e\mathcal{E}_0}{\omega} [1 - \cos(\omega t_c)] \hat{\epsilon} = -\Delta p,
$$
\n(A5)

which also follows from $\Delta p + \Delta p' = \overline{\Delta p} = 0$. The assumption here is that the collision time is negligible compared to the period of the field so that the same value of t_c appears in Eqs. $(A3)$ and $(A5)$.

The momentum transferred in the collision is

$$
Q = k_4 - k_1,\tag{A6}
$$

which, using the above relationships, may also be written

$$
Q = k_3 - k_2. \tag{A7}
$$

The kinetic energy change of the electron is given by

$$
\Delta E = \frac{1}{2m} (k_4^2 - k_1^2). \tag{A8}
$$

Using the above equations, and the fact that $k_2^2 = k_3^2$, gives

$$
\Delta E = \pm \frac{e\mathcal{E}_0}{m\omega} [1 - \cos(\omega t_c)] \hat{\epsilon} \cdot \mathbf{Q}.
$$
 (A9)

Averaging over all values of t_c yields the average energy absorbed or emitted by the electron

$$
\overline{\Delta E} = \pm \frac{e\mathcal{E}_0}{m\omega} \hat{\boldsymbol{\epsilon}} \cdot \boldsymbol{Q}.
$$
 (A10)

FIG. 5. (Color online) Scattering kinematics showing the scattering plane (*xy* plane) and the plane perpendicular to the momentum transfer *Q* (*xz* plane) that bisects the scattering angle formed by k_1 and k_4 . For any polarization vector that lies in this plane $\hat{\boldsymbol{\epsilon}} \cdot \hat{\boldsymbol{k}}_1 = \hat{\boldsymbol{\epsilon}} \cdot \hat{\boldsymbol{k}}_4.$

Thus there is maximum energy transfer when the polarization is parallel to the momentum transfer, whereas there is zero energy transfer when it is perpendicular. The latter can be readily understood from Fig. 5, which shows the plane perpendicular to Q that bisects $\theta_{\rm sc}$. Any polarization vector that lies in this plane makes equal angles with k_1 and k_4 , and

the energy transfer averaged over a cycle vanishes. It is only when the polarization vector makes unequal angles with the electron momenta before and after the collision that energy transfer occurs.

Equation $(A10)$ may be used to estimate the maximum number of photons of energy $\hbar \omega$ expected to be absorbed or emitted [15]

$$
n_{\max} \sim \pm \frac{eA_0}{m\hbar\omega} \hat{\epsilon} \cdot \mathbf{Q},\tag{A11}
$$

where the vector potential $A_0 = \mathcal{E}_0/\omega$.

The use of a homogeneous electric field is equivalent to the dipole approximation if the oscillating electric field is replaced by an electromagnetic wave. If the approximation is not valid we can still obtain the same result as follows. The electric field is now given by a plane wave $\mathcal{E} = \mathcal{E}_0 \sin(\kappa \cdot r + \omega t)$ with a corresponding vector potential $A(r,t) = A_0 \cos(\kappa \cdot r + \omega t)$, where κ is the wave vector and κ is the position vector. The electron momentum is k_1 at ($r = 0, t = 0$), and the collision occurs at $(r = r_c, t = t_c)$ with corresponding momentum changes

$$
\Delta p = -e\Delta A = eA_0[1 - \cos(\kappa \cdot r_c + \omega t_c)]. \tag{A12}
$$

The relationship $\Delta p = -\Delta p'$ is satisfied if the final momentum k_4 is taken when the electron coordinates (r_4 , t_4) next obey the relationship $(\mathbf{k} \cdot \mathbf{r}_4 + \omega t_4) = 2\pi$, so that $\Delta A + \Delta A' = 0$.

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