1-4-2016

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Repository Citation

Martin, Nicholas L. S. and deHarak, B. A., "Test of Target Independence for Free-Free Scattering in a Nd:YAG Laser Field" (2016). *Physics and Astronomy Faculty Publications*. 438.
https://uknowledge.uky.edu/physastron_facpub/438

Digital Object Identifier (DOI)
https://doi.org/10.1103/PhysRevA.93.013403

Test of target independence for free-free scattering in a Nd:YAG laser field

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(Received 18 November 2015; published 4 January 2016)

We report measurements of one-, two-, and three-photon processes during the elastic scattering of electrons through 90° by helium, argon, and molecular-nitrogen targets, in the presence of 1.17-eV photons from a Nd:YAG laser. The incident energy of the electrons was 200 and 350 eV, and the linear polarization direction of the laser was parallel to the momentum transfer direction. Our measured free-free count rates for the three processes are target independent within the experimental uncertainties, perfectly consistent with the Kroll-Watson approximation, which assumes no interaction of the laser radiation with the target.

DOI: 10.1103/PhysRevA.93.013403

I. INTRODUCTION

When an electron of energy E_i is elastically scattered by an atom or molecule A in the presence of a laser field of frequency ω, there is the possibility of the absorption or emission of one or more photons of energy $\hbar\omega$ by the electron. This process is known as laser-assisted free-free scattering, or simply free-free scattering [1,2], and may be represented by

$$A + e(E_i) + \mathcal{N}\hbar\omega \rightarrow A' + e(E_f) + \mathcal{N}'\hbar\omega, \qquad (1)$$

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 $E_f = E_i \mp n\hbar\omega$. The first free-free experiments were carried out in Ar in 1977 by Weingartshofer *et al.* [3] using 0.117-eV photons from a CO_2 laser.

We recently reported two free-free experiments using 1.17-eV photons from a Nd:YAG laser [4,5]. In the first experiment the single-photon emission probability was measured, for laser light of fixed polarization, as a function of incident electron energy [4]. In the second experiment the single-photon emission free-free signal was measured at a number of discrete incident energies while the direction of the polarization of the light was varied over 180° in a plane perpendicular to the scattering plane [5].

The results of both these free-free experiments were in good agreement with the theoretical predictions of the semiclassical Kroll-Watson Approximation (KWA) [6]. To our knowledge, these two experiments were the first to use 1.17-eV photons to investigate the free-free process for elastic scattering, although Luan *et al.* [7] investigated the inelastic scattering analog known as simultaneous electron-photon excitation [1].

Both our earlier experiments were carried out using helium as a target, and both investigated only single-photon processes. We have now extended our test of the KWA for 1.17-eV photons by measuring the free-free signal for one-, two-, and three-photon processes in He, Ar, and N_2. These three targets span a large mass range with $M_{\mathrm{He}} = 4$ u, $M_{\mathrm{N}_2} = 28$ u, and $M_{\mathrm{Ar}} = 40$ u and lowest electronic excitation energies of about 6 eV (N_2), 12 eV (Ar), and 21 eV (He).

A key assumption of the KWA is that the ratio of the free-free cross section to the elastic-scattering cross section is independent of the target atom or molecule. One requirement for this to be true is that the photon energy is much less than the lowest excitation energy E' of the target, and the laser intensity is sufficiently small that multiphoton excitation or ionization processes can be ignored. It is also assumed in the KWA that the laser does not interact with the target in any way, i.e., the target is not "dressed" by the laser field. Byron and Joachain [8] investigated the effect of dressing the target atom by the electric field, for laser intensities corresponding to electric-field strengths much less than the internal fields of an atom but much larger than normal laboratory fields. They evaluated the effect of a hydrogen atom dressed with an admixture of p states due to the laser's electric field. More generally, the effect of dressing could be expressed in terms of the electric-dipole polarizability α of an atom, a result previously obtained by Zon [9] in the context of Bremsstrahlung. Byron and Joachain [8] concluded that the effects in helium would be negligible and suggested the heavier noble gases as possible candidates. Wallbank and Holmes [10] looked for dressing effects using 0.117-eV photons in a comparison of free-free experiments on He and Ar for certain geometries where the KWA predicted small cross sections, but their results were inconclusive. Very recently the first experiments that have unambiguously observed the effect of dressed atoms in laser-assisted scattering experiments have been reported by Morimoto *et al.* [11]. The experiments were carried out in Xe, for which $\alpha = 28$ a.u. [12], and the effect of dressed states was only observed at scattering angles less than 1°. At larger scattering angles, their results were in good agreement with the KWA. The experiments reported below were carried out at 90°, for which the effect of dressed states is therefore expected to be very small, as is shown below.

Another requirement for the KWA to be true, even in the absence of dressed-atom effects, is that only first-order scattering processes are important, for if a second-order treatment is necessary the sum over all intermediate excited states clearly depends on the energy-level structure of the target. Such second-order terms for He, Ar, and especially N_2, with its vibrational and rotational levels, are therefore expected to be very different.

II. THEORY

In the KWA the free-free cross section depends on the dimensionless parameter

$$x = -0.022\lambda^2 I^{1/2} E_i^{1/2} \frac{\hat{\boldsymbol{\epsilon}}\cdot\boldsymbol{Q}}{k_i}, \qquad (2)$$

where $\lambda(=2\pi c/\omega)$ is the wavelength of the radiation in μm, I is its intensity in GW/cm^2, $\hat{\boldsymbol{\epsilon}}$ is the polarization direction, E_i is the incident electron energy in eV, and $\boldsymbol{Q}$ 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, and depends strongly on both the incident electron energy and the laser intensity.

The KWA then relates the free-free cross section $d\sigma^{(n)}_{\mathrm{KWA}}/d\Omega$, for absorption ($n<0$) or emission ($n>0$) of n photons, to the field-free elastic-scattering cross section $d\sigma_{\mathrm{el}}/d\Omega$, by [6]

$$\frac{d\sigma^{(n)}_{\mathrm{KWA}}}{d\Omega} = \frac{k_f}{k_i} J^2_{|n|}(x) \frac{d\sigma_{\mathrm{el}}}{d\Omega}. \quad (3)$$

Here $\boldsymbol{k}_i$ and $\boldsymbol{k}_f$ are the initial and final electron momenta (so $\boldsymbol{Q}=\boldsymbol{k}_f-\boldsymbol{k}_i$), and $J_{|n|}$ is a Bessel function of the first kind of order $|n|$.

The ratio of n'- to n-photon emission or absorption is then given by

$$\frac{d\sigma^{(n')}_{\mathrm{KWA}}}{d\Omega} \bigg/ \frac{d\sigma^{(n)}_{\mathrm{KWA}}}{d\Omega} = [J_{|n'|}(x)/J_{|n|}(x)]^2, \quad (4)$$

where we have used $k_f(n')/k_f(n) \approx 1$ since in our experiments n and n' are small and $E_i \gg \hbar\omega$. Similarly the parameter x is evaluated with the value of $\boldsymbol{Q}$ for field-free elastic scattering—a good approximation except for very small scattering angles. With these approximations there is no difference between n-photon absorption or emission.

It is possible to estimate the effect of target dependence through dressed states. Zon's model [9] yields a simple analytical formula for the cross section [11], which includes the effect of dressing via the polarizability α:

$$\frac{d\sigma^{(n)}_{\mathrm{ZON}}}{d\Omega} = \frac{k_f}{k_i} \left| J_n(x) f_{\mathrm{el}} - \frac{\alpha m_e^2 \omega^2 x}{2\pi\varepsilon_0 Q^2} J'_n(x) \right|^2, \quad (5)$$

where f_{el} is the field-free scattering amplitude ($d\sigma_{\mathrm{el}}/d\Omega = |f_{\mathrm{el}}|^2$), m_e is the electron mass, and J'_n is the first derivative of the Bessel function. The first term is simply the Kroll-Watson approximation, and the second term is the extra term due to the dressing of the atom by the laser. Table I shows the calculated percentage difference between the undressed and the dressed cross sections for one-, two-, and three-photon processes given by Eqs. (3) and (5), using $\alpha = 1.4$, 11.1, and 11.5 a.u. for He, Ar, and N_2, respectively [12,13]. The remaining parameters were chosen to correspond to the experiments reported below: laser intensity 5 GW/cm^2 and available cross-section data for 200- and 350-eV electrons scattered through 90° in He, Ar, and N_2 [14,15]. It can be seen that the dressing effects get larger with increasing n, reaching about 2% for Ar and N_2 for $n=3$. In fact the dressing effects are very similar for Ar and N_2, whereas for He the effects are two to three times smaller. Given our experimental uncertainties, we do not expect to be able to detect any differences between the targets; the maximum difference is $n=3$, 200 eV, for which the dressing effect on N_2 is only 1.3% larger than on He.

Note that the use of the polarizability circumvents a detailed calculation involving the precise energy-level structure of the target. However it is the *dipole* polarizability that is used in Eq. (5), and is not therefore equivalent to a rigorous second-order calculation that involves a summation over intermediate states of every polarity. Nevertheless, the dipole terms are expected to dominate at high incident electron energies, and therefore the calculations shown in Table I should be a good indication of the size of the expected effects.

TABLE I. Calculated percentage differences between the Kroll-Watson approximation [undressed targets, Eq. (3)] and Zon's model [dressed targets, Eq. (5)], for 200- and 350-eV electrons scattered through 90° in He, Ar, and N_2 in a laser field of 5 GW/cm^2. See text for details.

		(KWA-Zon)/KWA % diff.		
E_i (eV)		He	Ar	N_2
200	$n=1$	0.20	0.55	0.59
	$n=2$	0.46	1.25	1.32
	$n=3$	0.71	1.92	2.04
350	$n=1$	0.17	0.40	0.37
	$n=2$	0.44	1.01	0.94
	$n=3$	0.69	1.59	1.48

III. EXPERIMENTAL METHOD

The free-free experiments were carried out using a Continuum Powerlite 9030 Nd:YAG laser with photon energy 1.17 eV ($\lambda = 1.06\ \mu$m), repetition rate 30 Hz, pulse duration ≈8 ns, and, in the present experiments, deduced intensities of 4.3 and 11.3 GW/cm^2. The laser beam is focused down to a diameter of 0.75 mm in the interaction region.

A schematic of the experimental setup for the present experiments is shown in Fig. 1. The electron spectrometer consists of an unmonochromated electron gun and a scattered electron detector, both mounted on independent coplanar concentric turntables, and a single-bore gas nozzle to create the target beam. See [4] 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°. The laser polarization direction $\hat{\boldsymbol{\epsilon}}$ is parallel to the

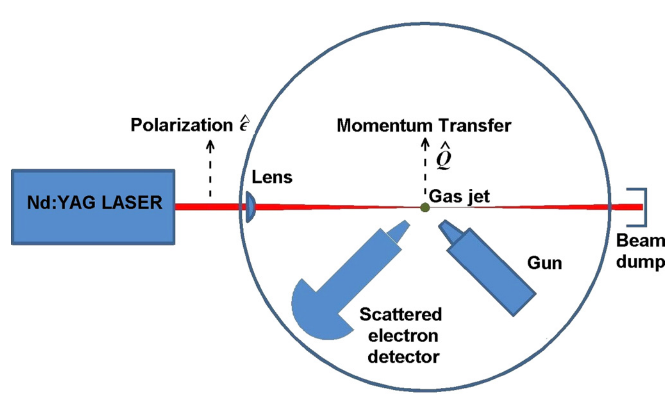

FIG. 1. Schematic of the laser-assisted electron-scattering apparatus.

momentum transfer direction $\hat{\boldsymbol{Q}}$, as shown in the figure; this maximizes the free-free signal via the $\hat{\boldsymbol{\epsilon}}\cdot\boldsymbol{Q}$ term in Eq. (2).

The laser beam is terminated in a beam dump with an attached thermocouple to monitor the beam intensity as a function of time. The temperature of the beam dump with the laser on is typically about 50°C above room temperature.

For 200-eV incident electron energy, data were taken in separate experiments, with the scattered-electron detector manually tuned for $n = 1,2$, or 3 between each experiment and each target. For 350 eV, the $n = 1,2,3$ data were taken in a single experiment (for each target) using a computer controlled digital-to-analog converter that repeatedly cycled through the three voltages appropriate for one-, two-, and three-photon energies away from the elastic-scattering peak. Thus the 350-eV experiments, for a given target, are subject to less systematic error, between $n = 1,2,3$ photon processes, due to laser-flashlamp degradation than the 200-eV experiments. However, there may still be systematic uncertainties *between* targets.

IV. RESULTS AND DISCUSSION

Free-free measurements were carried out in He, Ar, and N_2 at incident electron-beam energies of 200 and 350 eV for $|n| = 1,2,3$ photon processes. At each energy, the gas pressure for each of the three targets was adjusted to keep the (laser-off) elastic-scattering signal the same (approximately 800 000 counts/s for 350 eV—somewhat less for 200 eV). This enabled a direct comparison of the free-free count rates from the three targets, and is equivalent to testing the target independence of the ratio $(d\sigma^{(n)}_{\mathrm{KWA}}/d\Omega)/(d\sigma_{\mathrm{el}}/d\Omega)$ [see Eq. (3)]. For experimental reasons the single-photon measurements at 200 eV are for photon emission; all other measurements are for photon absorption; within the approximations of Eq. (4) the ratios for absorption and emission should be the same.

Figure 2 shows the timing spectra for one-, two-, and three-photon absorption by a 350-eV beam in Ar. The different laser-off signals in the three spectra correspond to the high-energy tail of the elastically scattered electron beam at one-, two-, and three-photon energies above the elastic peak. The spectra were obtained over a period of 46.5 h by repeatedly cycling through the three appropriate energies in 1-h intervals. The free-free signal occupies several 12.5-ns time bins due to the time spread of electron trajectories through the analyzer optics. The time bins are numbered with respect to a timing signal that controls the laser; see [4] for details.

Our experimental results for 200- and 350-eV incident electron-beam energy for He, Ar, and N_2 are shown in Fig. 3, and absolute values with statistical uncertainties are given in Table II. The results are presented as actual free-free count rates per hour of data collection for one photon (emission at 200 eV, absorption at 350 eV), two-photon absorption, and three-photon absorption—note the logarithmic scale in the figure. Each hour of data collection corresponds to a laser-on time of about 1 ms. Long run times were thus required to get adequate statistics; for He, for example, the 200-eV three-photon absorption results were obtained from three experiments totalling 110 h of data taking. (We do not know the overlap of the electron beam, the laser beam, and the target gas beam well enough to give our results as absolute cross-section ratios.)

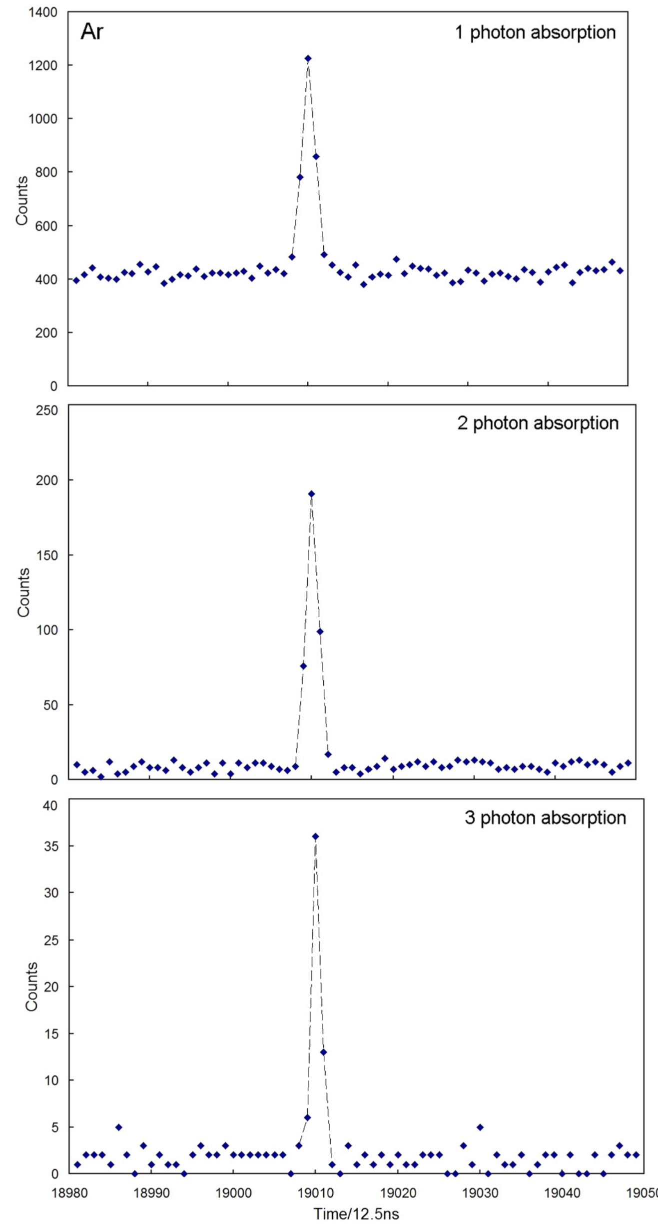

FIG. 2. Timing spectra of scattered-electron events in Ar corresponding to the absorption of one, two, and three 1.17-eV photons by 350-eV electrons elastically scattered through 90°, showing the counts per 12.5 ns time bin. The total data collection time of 46.5 h was equally shared between the three spectra. The dashed line is to guide the eye.

In Fig. 3 the dashed lines through the one- and two-photon processes are the average values over the three targets at each energy. Experiments at the two incident energies were carried out some time apart, during which the laser flashlamps had degraded and the laser intensity had dropped from about 11.3 to 4.3 GW/cm^2—these values were extracted by fitting the KWA to the one-photon–two-photon dashed line averages [see Eq. (4)]. The three-photon data then provide an absolute test of the KWA shown by the dashed line through the three-photon data at each incident electron energy.

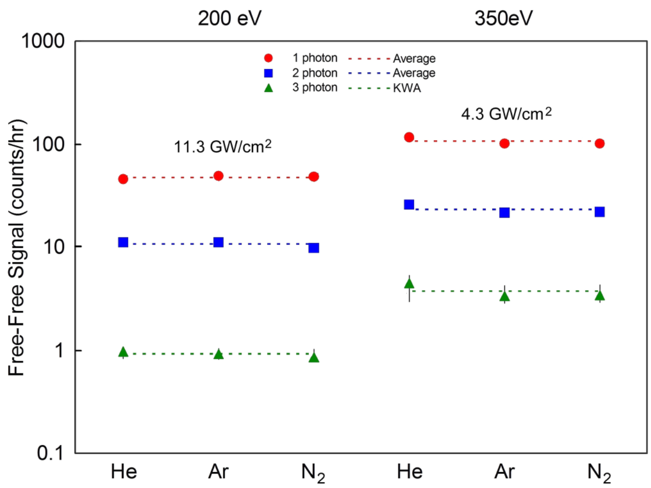

FIG. 3. Measured free-free count rates for elastic scattering of 200- and 350-eV electrons by He, Ar, and N_2 in the presence of 1.17-eV photons from a Nd:YAG laser. Note the logarithmic scale, and note that 1 h of data collection corresponds to a laser-on time of about 1 ms. The dashed lines for the one- and two-photon processes are the averages of the experimental data. The dashed line for the three-photon process is a KWA calculation. The statistical error bars are in most cases smaller than the symbols. See text for details.

In addition to the statistical uncertainty, we estimate a possible systematic uncertainty of 10% due to laser flashlamp degradation between experiments and electron-beam retuning for different targets, etc. Within these experimental uncertainties, the data are independent of the target at both energies and are perfectly consistent with the KWA. In fact only the He 350-eV data differ from those of the other targets by more than the statistical uncertainties, but are consistent within the joint statistical and systematic uncertainties. Clearly, our experiments are unable to observe the small effects due to dressing predicted in Table I.

TABLE II. Measured free-free count rates for 200- and 350-eV electrons scattered through 90° by He, Ar, and N_2 in the presence of 1.17-eV photons from a Nd:YAG laser. The laser-on time is about 1 ms per hour of data collection. The statistical uncertainties are given; in addition there is an estimated 10% systematic uncertainty. See text for details.

E_i (eV)		He	Ar	N_2
200	$n = 1$	46(2)	49(4)	48(4)
	$n = 2$	11(1)	11(1)	10(1)
	$n = 3$	1.0(1)	0.9(1)	0.9(1)
350	$n = 1$	115(5)	100(3)	101(4)
	$n = 2$	26(2)	22(1)	22(1)
	$n = 3$	4.5(8)	3.3(5)	3.4(5)

V. CONCLUSIONS

In this work, the KWA has been tested for three different targets in a single experiment, and therefore under the same experimental conditions. Taken together with our other two experiments on energy dependence [4] and laser polarization [5], the Kroll-Watson approximation has now been shown to give a good description of free-free processes for a moderate intensity laser field of 1.17-eV photons and a wide range of physical parameters. Possible future experiments include investigating the KWA at higher intensities by focusing the laser beam down to a smaller diameter in the interaction region.

ACKNOWLEDGMENTS

This work was supported by the NSF under Grants No. PHY-0855040 (N.L.S.M.) and No. PHY-1402899 (B.A.d.).

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