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Free–free experiments: the search for dressed atom effects

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Abstract

Experiments on free–free electron scattering, specifically the absorption or emission of 1.17 eV photons from a Nd:YAG laser field by an unbound electron when it is scattered by an atom or molecule, are reviewed. For large scattering angles such experiments are well described by a simple analytical theory that is independent of the properties of the target. At small scattering angles this theory breaks down for targets with a high dipole polarizability α, and an additional term needs to be incorporated in the scattering amplitude. This term is proportional to the dipole polarizability, and hence introduces the properties of the target into the free–free cross section—i.e., the laser field 'dresses' the atom. A progress report is given of free–free experiments designed to look for such 'dressed atom' effects during the electron-impact excitation of argon in the presence of a laser field; the lowest excited states of argon have $\alpha \approx 300$ atomic units.

Keywords: electron scattering, free–free, laser field

(Some figures may appear in colour only in the online journal)

1. Introduction

The absorption or emission of radiation by charged particles during collisions with atoms and molecules has long been known to be important in astrophysical and plasma phenomena. As long ago as 1930, Pannekoek [1] considered the effect of 'free–free' transitions, i.e., the absorption (or emission) by unbound electrons of energy during collisions in the presence of radiation. He found that this type of transition is important for determining the infrared opacities of certain types of stars [2]. Chandrasekhar and Breen [3] found that existing discrepancies in the long wavelength absorption of the solar atmosphere could be explained by free–free cross sections, for electrons on negative hydrogen ions, that were an order of magnitude larger than those previously used. Free–free transitions are also known to dominate the radiation transport in certain types of air plasmas, such as cascade arcs and shock tubes [4], and they are also important in the heating of plasmas by radiation [5]. More recently, it has been proposed to use laser-assisted collision processes to manipulate ionization and capture in ion-atom collisions [6]. Free–free experiments carried out under controlled laboratory conditions, such as those described below, can therefore provide detailed information on these important processes, even though the targets investigated are different.

The first experiments involving electron scattering in a laser field were on the so called 'free–free' transitions where the *elastic* scattering process is involved as a mechanism for allowing both energy and momentum conservation when the incident electron absorbs or emits a photon in the field of the atom [7]. Weingartshofer *et al* [8] observed the absorption and emission of up to three 0.117 eV photons from a continuous CO_2 laser during e–Ar elastic scattering. Wallbank and Holmes [9–11] carried out similar experiments using a pulsed CO_2 laser and a helium target. They reported the absorption and emission of up to five 0.117 eV photons and found intensities much greater than theoretical predictions. The first experiment on electron-impact excitation in the presence of a laser field (simultaneous electron-photon excitation (SEPE)) was carried out by Mason and Newell [12]. Luan *et al* [13] investigated SEPE in He using a Nd:YAG laser which produces photons of energy 1.17 eV, ten times the CO_2 laser energy. They found disagreement between theory and experiment. In an (e, 2e)

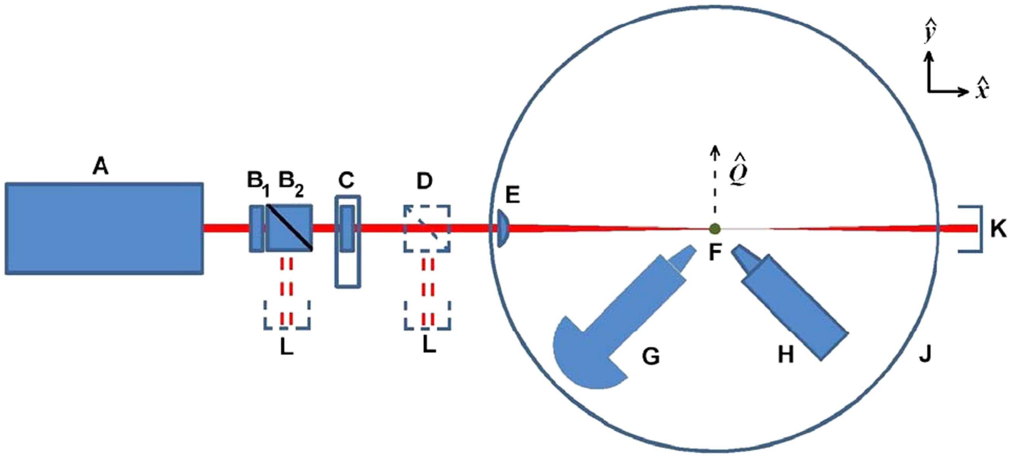

Figure 1. Electron scattering apparatus used for free–free experiments. A: Nd:YAG laser, B–E: optical components, F: He nozzle, G: scattered-electron detector, H: electron gun, J: vacuum chamber, K: beam dump, L: power meter (for set-up), $\hat{Q}$: momentum transfer direction. Reprinted figure with permission from [18]. Copyright (2014) by the American Physical Society.

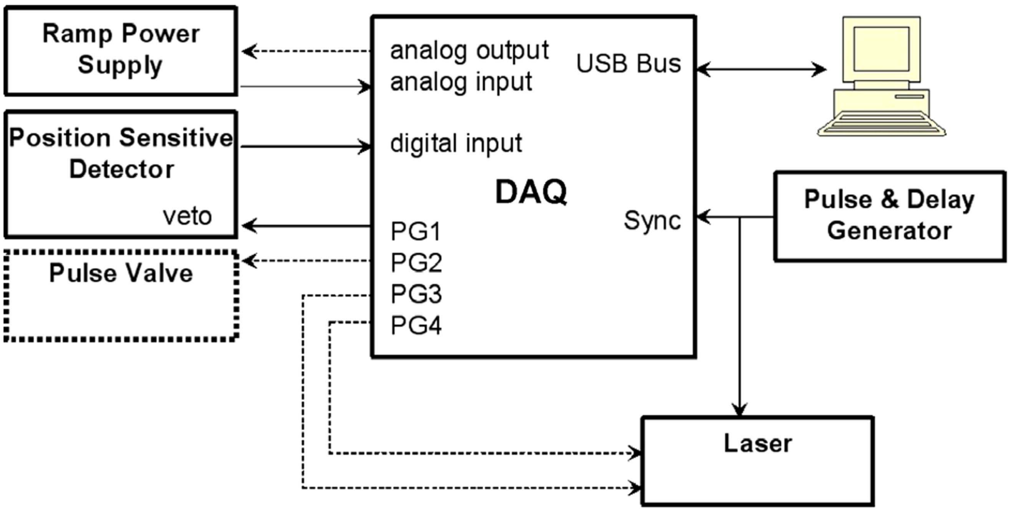

Figure 2. Data acquisition system (DAQ).

experiment, Höhr *et al* [14] investigated He ionization by 1 keV electrons in the presence of a laser field of 1.17 eV photons from a Nd:YAG laser. Their results were in poor agreement with quantum calculations but could—oddly enough—be explained with a simple classical model. The above experiments indicate that laser-assisted electron collisions are not yet well understood.

2. Experimental tests of the Kroll–Watson approximation (KWA)

As stated above, the 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 [15, 16]. 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,

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

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 are carrying out experiments of this type using a Continuum Powerlite 9030 laser, photon energy 1.17 eV, with a repetition rate of 30 Hz, and pulse duration ~8 ns and a nominal energy 1.6 J per pulse. The apparatus is shown schematically in figure 1. The spectrometer and the laser have been interfaced with a data acquisition system (DAQ) developed specially for our experiments; see figure 2. At the heart of the DAQ is a Parallax Inc. Propeller™ chip that contains eight separate processors that can operate independently while being synchronized by a single system clock [17]. Data is acquired in a time spectrum, of 12.5 ns time bins, that spans the laser firing. The laser-assisted signal is found from the laser-on signal minus the laser-off signal. The data-collection system also records the relative laser power, via both a power meter and a thermocouple on the laser beam dump, and the energy-selection voltage on the scattered-electron optics.

Our first three free–free experiments tested the KWA [19], a semi-classical theory used fairly successfully to interpret most free–free experiments, but never before tested for a laser field of 1.17 eV photons. The KWA relates the free–free cross section $d\sigma^{(n)}_{KWA}/d\Omega$, for absorption ($n < 0$) or emission ($n > 0$) of n photons, to the field-free elastic scattering cross section

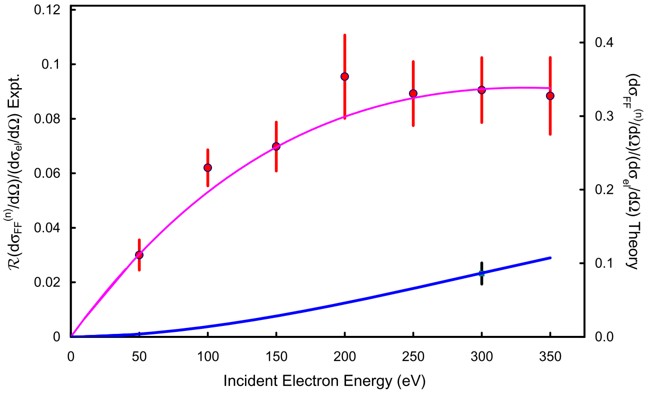

Figure 3. Free–free transitions for He at incident electron energies 50 to 350 eV. The solid circles are the experimental data for one-photon emission, and the solid square is for two-photon emission. The solid lines are one-photon and two-photon KWA calculations fitted to our experiment at 300 eV. Reprinted figure with permission from [20]. Copyright (2011) by the American Physical Society.

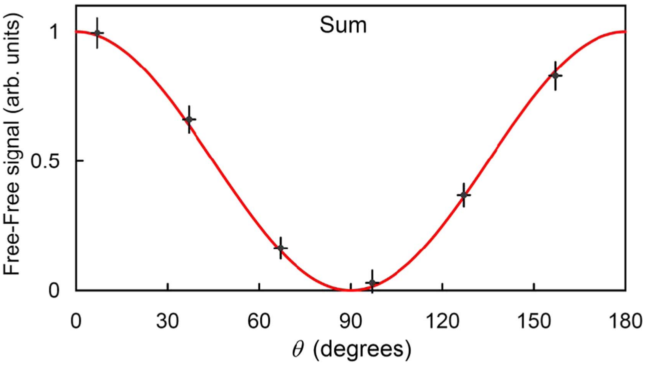

Figure 4. The free–free signal for He as a function of laser polarization direction. Reprinted figure with permission from [18]. Copyright (2014) by the American Physical Society.

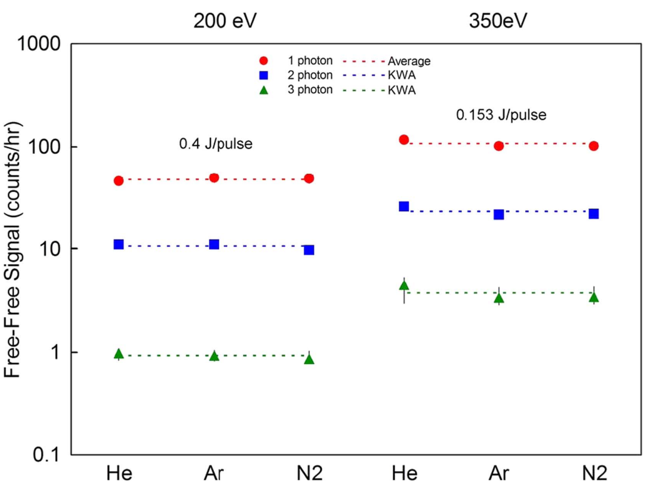

Figure 5. The free–free signal for different targets: He, Ar, and N_2. Reprinted figure with permission from [21]. Copyright (2016) by the American Physical Society.

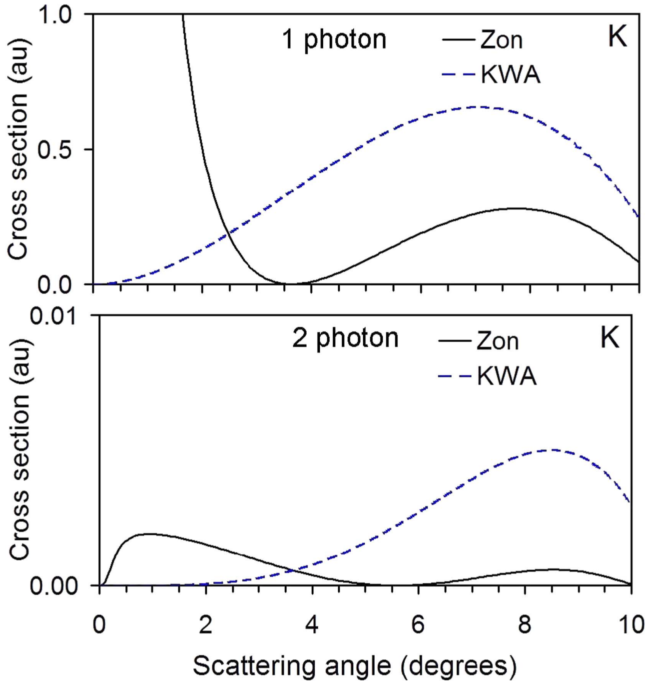

Figure 6. Calculated angular distributions for the absorption of one and two 1.17 eV photons during the elastic scattering of 350 eV electrons by potassium ($\alpha = 290$). Blue broken line: Kroll–Watson approximation for undressed atoms. Black solid lines: Zon's model for dressed atoms.

$d\sigma_{el}/d\Omega$, by [19]

$$\frac{d\sigma_{KWA}^{(n)}}{d\Omega} = \frac{k_f}{k_i} J_n^2(x) \frac{d\sigma_{el}}{d\Omega},$$

$$\text{with} \quad x = -0.022\lambda^2 I^{1/2} E_i^{1/2} \frac{\hat{\epsilon} \cdot \vec{Q}}{k_i}. \tag{2}$$

Here $\vec{k}_i$ and $\vec{k}_f$ are the initial and final electron momenta, and J_n is a Bessel function of the first kind of order n, λ is the wavelength of the radiation in μm, I is its intensity in GW cm^{-2}, $\hat{\epsilon}$ is the polarization direction, E_i is the incident electron energy in eV, and $\vec{Q} = \vec{k}_f - \vec{k}_i$ is the momentum transfer. In fact what is measured in an experiment is the ratio of the laser-on cross section (*FF*) to the laser-off cross section,

$$(d\sigma_{FF}^{(n)}/d\Omega)/(d\sigma_{el}/d\Omega) = \frac{k_f}{k_i} J_n^2(x), \tag{3}$$

which provides a direct test of the KWA.

Our first experiment [20] measured single-photon emission at a range of incident electron energies from 50 to 350 eV on He. The results are shown in figure 3, where it can be seen that our data is perfectly consistent with the KWA.

Our next test of the KWA, for 1.17 eV photons, measured the free–free signal as a function of the linear polarization direction with respect to the momentum transfer direction [18]. This is of interest because the experiments of Wallbank and Holmes [11], using 0.117 eV photons, 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. We measured the free–free signal (single-photon

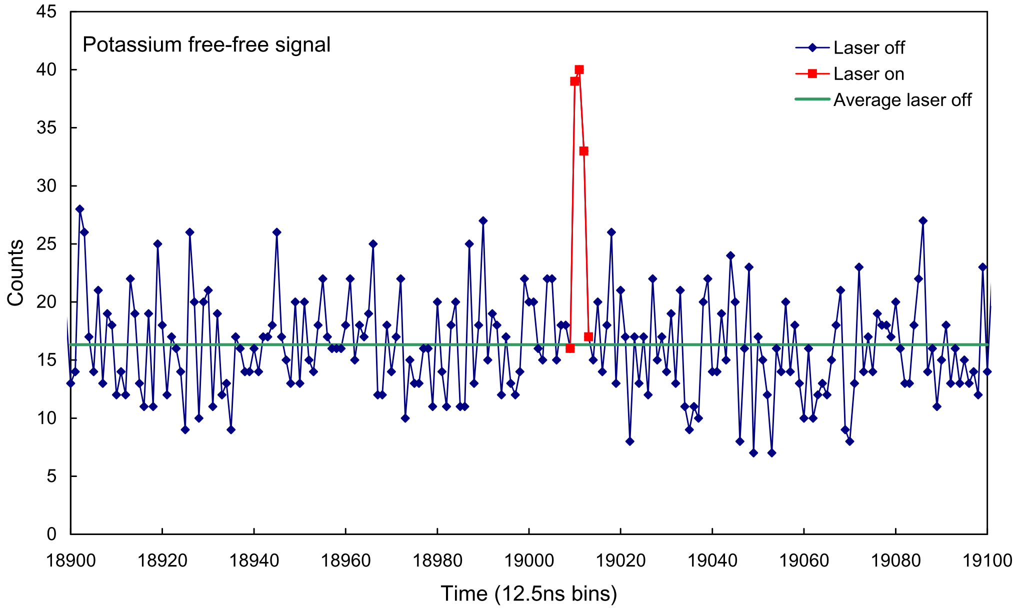

Figure 7. Potassium free–free signal in the timing spectrum. The red and blue data points are for electrons that reach the detector while the laser is on and off, respectively.

absorption) for electrons of energy 30, 60, 120, and 200 eV incident on He in the presence of a laser field with polarization θ varied over the full 180°. The KWA predicts a free–free signal $\propto\cos^2\theta$ at all these energies for our laser intensity. For all incident energies our results were consistent with the KWA, and we found no evidence of a non zero free–free signal when the laser polarization was perpendicular to the momentum transfer direction. Figure 4 shows the sum of the data at all energies. The curve is pure $\cos^2\theta$, and it can be seen that there is no evidence of a non zero minimum. Covering the full 180° range gives a more rigorous test of the KWA than a single measurement at 90°—in fact our measurements are the first ever comprehensive investigation, for any photon energy, of the free–free signal as a function of polarization direction.

Our third experiment investigated free–free scattering from different targets [21]. Both our earlier experiments were carried out using helium as a target, and both investigated only single-photon processes. This experiment extended our test of the KWA for 1.17 eV photons by measuring the free–free signal for 1, 2, and 3 photon absorption for He, Ar, and N_2 targets. These three targets span a large mass range with $M_{He} = 4$ u, $M_{N_2} = 28$ u, and $M_{Ar} = 40$ u, and lowest electronic excitation energies in eV of about 6 (N_2), 12 (Ar), 21 (He). One of the key assumptions 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 of the target, and the laser intensity is sufficiently small that multi-photon excitation processes can be ignored. A more interesting requirement for the KWA to be true is that only first order 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. Figure 5 shows our results for incident electron energies 200 and 300 eV on the three targets. At each energy, the count rate in the elastic peak was adjusted to be the same for each of the targets, and measurements of the free–free signal were made at 1.17 eV, 2.34 eV, and 3.51 eV above the elastic peak, corresponding to 1, 2, and 3 photon absorption. All the results are in good agreement with the KWA, and there is no evidence of any target effects. This is particularly surprising for N_2, in view of its low-lying vibrational and rotational structure. To our knowledge, this is the first direct comparison of free–free scattering for different targets.

3. Deviations from the KWA: dressed atoms

In fact deviations from the KWA are predicted at small angle scattering—the experiments described above were for scattering angles at 90° or greater. The deviations result from the distortion of the target by the electric field of the laser; this effect of laser radiation 'dressing' an atom during an electron–atom collision is currently of great interest. It was first treated theoretically by Gersten and Mittleman [22] in 1976, and it has taken nearly 40 years for its experimental observation. 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* [23]. Byron and Joachain [24] investigated the case where the atom is 'dressed' by the electric field of the laser, and therefore the KWA is not expected to be correct, particularly at very small scattering angles. They evaluated the effect of a hydrogen atom dressed by 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 [25] in the context of Bremsstrahlung. They concluded that the effects in helium ($\alpha = 1.4$ [atomic units] [26]) would be negligible, and suggested the heavier noble gases as possible candidates.

Morimoto *et al* carried out experiments in Xe, for which $\alpha = 28$ [26]. They measured the angular distribution of

Configuration	Term	J	Level (eV)
$3s^23p^6$	^{1}S	0	0.0000000
$3s^23p^5(^2P°_{3/2})4s$	$^2[3/2]°$	2	11.54835433
		1	11.62359262
$3s^23p^5(^2P°_{1/2})4s$	$^2[1/2]°$	0	11.72316029
		1	11.82807106
$3s^23p^5(^2P°_{3/2})4p$	$^2[1/2]$	1	12.90701519
		0	13.27303799
$3s^23p^5(^2P°_{3/2})4p$	$^2[5/2]$	3	13.07571560
		2	13.09487245
$3s^23p^5(^2P°_{3/2})4p$	$^2[3/2]$	1	13.15314376
		2	13.17177759
$3s^23p^5(^2P°_{1/2})4p$	$^2[3/2]$	1	13.28263891
		2	13.30222736
$3s^23p^5(^2P°_{1/2})4p$	$^2[1/2]$	1	13.32785693
		0	13.47988670

Figure 8. Ar energy levels [30].

electrons having absorbed one photon of energy 1.55 eV during elastic scattering. The KWA predicted that the intensity should fall to zero at small scattering angles, but their data showed a rising intensity as the scattering angle was decreased, in qualitative, but not quantitative, agreement with a calculation based on Zon's model [25] that includes the effect of dressing via the polarizability α. This quantitative disagreement was possibly due to experimental complications at the very small scattering angles ($\sim 0.5°$) for which dressing effects began to occur. They also measured the angular distribution for two-photon absorption, and found negligible dressing effects, in agreement with Zon's model prediction.

Zon's model [25] yields a simple analytical formula for the cross section [23], which includes the effect of dressing,

$$\frac{d\sigma_{ZON}^{(n)}}{d\Omega} = \frac{k_f}{k_i}\left| J_n(x) f_{el} - \frac{\alpha m_e^2 \omega^2 x}{2\pi\varepsilon_0 Q^2} J_n'(x)\right|^2, \quad (4)$$

where f_{el} is the field-free scattering amplitude ($d\sigma_{el}/d\Omega = |f_{el}|^2$), m_e is the electron mass, $\omega = 2\pi c/\lambda$ is the frequency of the laser radiation, and J'_n is the first derivative of the Bessel function; the quantity x is the same as in equation (2). The first term is simply the KWA, and the second term represents the dressing of the atom by the laser. This equation predicts that dressing effects are observable from zero scattering angle, up to a scattering angle that increases strongly with α.

One target with a large dipole polarizability is potassium, with $\alpha = 290$ [26]. Using equation (4) we have calculated the expected KWA and Zon cross sections for one- and two-photon processes during the elastic scattering of electrons by potassium, with parameters appropriate for our experimental set-up. The incident electron energy is 350 eV, the photon energy is 1.17 eV (Nd:YAG laser), the laser intensity is 15 GW cm^{-2}, and the field-free elastic scattering cross sections were taken from the NIST database [27]. The results of our calculations are shown in figure 6. The effect of the high value of α for K is to shift the minima in the Zon cross sections to much larger scattering angles, into a region more readily experimentally accessible than in the Xe case. The Zon calculation also predicts dramatic effects in *both* one- and two-photon cross sections, unlike for Xe.

We have carried out exploratory free–free experiments in potassium, using the metal vapour oven previously used on Cd experiments [28]. Figure 7 shows our first ever free–free signal in potassium, for an incident energy of 350 eV, and the kinematics shown in figure 1 with the laser polarization along the momentum transfer direction; this arrangement maximizes the free–free signal. No dressed atom effects are expected for these kinematics; the experiment was designed to test the feasibility of free–free experiments in metal vapours. In order to get a measurable free–free count rate it was necessary to produce an intense potassium beam, as a result of which the exit aperture of the gun eventually blocked up—even without the electron beam collimating apertures (needed to prevent the electron beam from entering the detector at the small scattering angles) required for the observation of dressed atom effects (see figure 6).

We are currently investigating a system that suffers from neither of these disadvantages: free–free *inelastic* scattering in Ar. The dipole polarizability of the ground state of Ar is $\alpha = 11$ [26], whereas for the lowest excited states $\alpha \approx 300$ [29] (i.e., similar to the potassium ground state). Figure 8 shows the ground, and lowest excited states of Ar [30], and figure 9 shows an energy loss spectrum (laser-off) taken with the unmonochromated electron beam used in our experiments.

In the absence of a laser field, electron-impact excitation of the first excited state, at high incident electron energy and small scattering angle, is given by the dipole-allowed transition

$$\mathrm{Ar}(|3p^6\rangle) + e(E_i) \rightarrow \mathrm{Ar}(|3p^5\rangle|4s\rangle) + e(E_f), \quad (5)$$

but when a laser field is present the reaction is

$$\mathrm{Ar}(|3p^6\rangle) + e(E_i) + \mathcal{N}\hbar\omega \rightarrow \mathrm{Ar}(|3p^5\rangle[|4s\rangle + |4p\rangle]) + e(E_f) + \mathcal{N}'\hbar\omega, \quad (6)$$

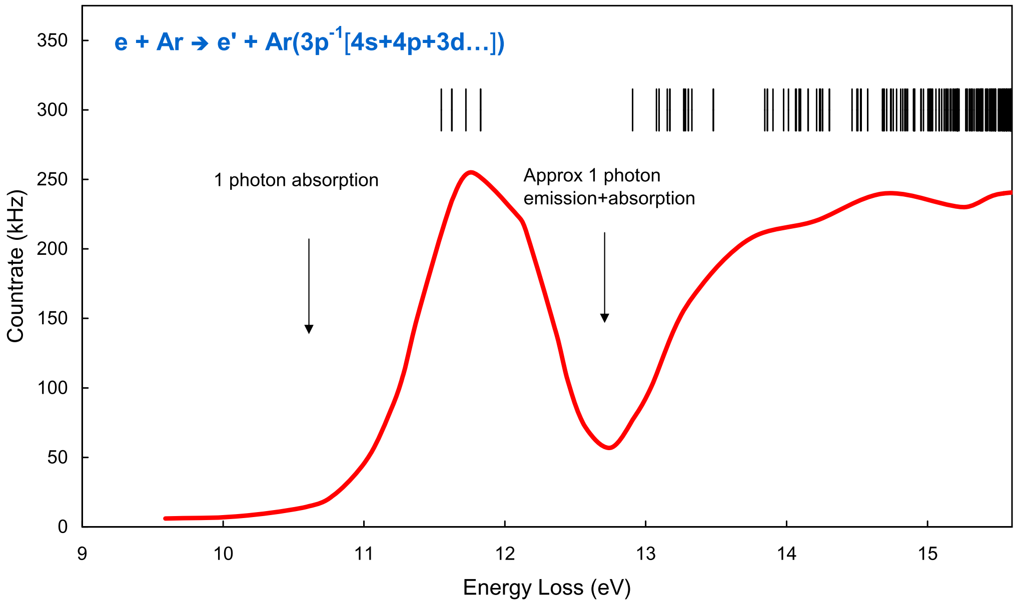

Figure 9. Ar electron-impact energy loss spectrum for 350 eV incident energy electrons scattered through 45°. The positions of excited Ar states are indicated by the vertical bars above the curve.

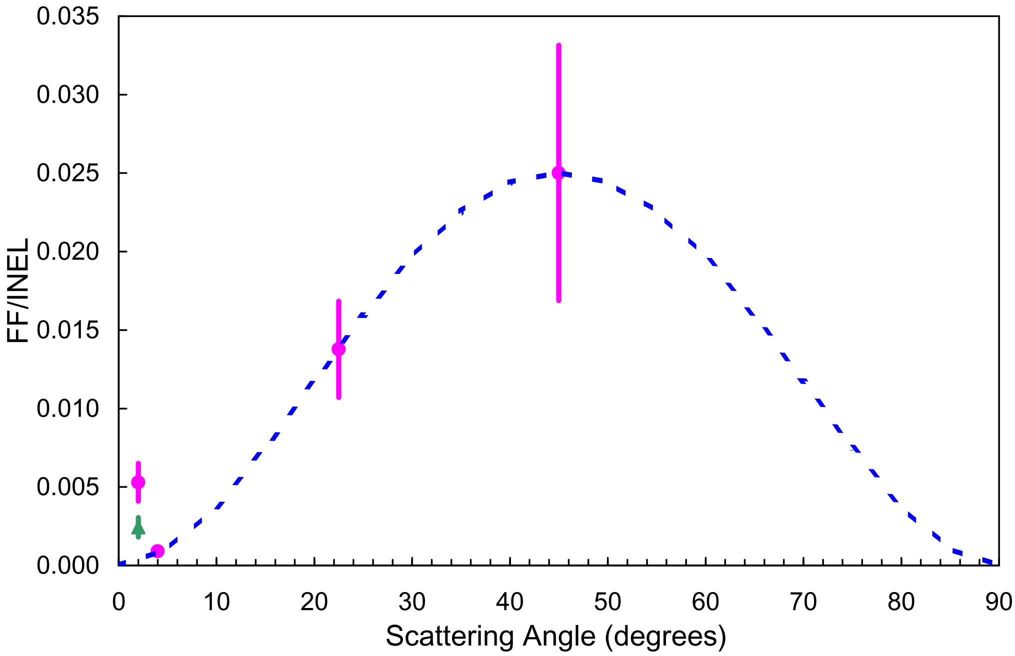

Figure 10. Provisional Ar inelastic free–free angular distribution for 350 eV incident electrons. The circles and triangle are for electron energy loss below and above the energy of the first excited state, indicated by the vertical arrows in figure 9 (see text). The vertical bars are the statistical uncertainties. The dashed line is a KWA calculation normalized to the data at 45°.

due to Stark mixing between opposite parity excited states; it is this mixing that leads to a high dipole polarizability. Thus dressed atom effects are expected to be observed for excitation at small scattering angles.

Figure 10 shows the results of preliminary free–free experiments carried out on the lowest excited state of Ar. A KWA calculation is also shown, normalized to the data at a scattering angle $\theta_{sc} = 45°$. (The experimental geometry is different from the earlier experiments: the gun is positioned 90° anticlockwise from that shown in figure 1; the scattered-electron detector then has an angular range $\theta_{sc} = 0 \rightarrow 90°$, with the laser polarization along 45°.) The experimental points at 4° and 22.5° are consistent with the KWA, but the two data points at 2° are not, and support the possibility of a target dressed by the laser field. The upper, circular, data point at 2° corresponds to a measurement at an energy loss

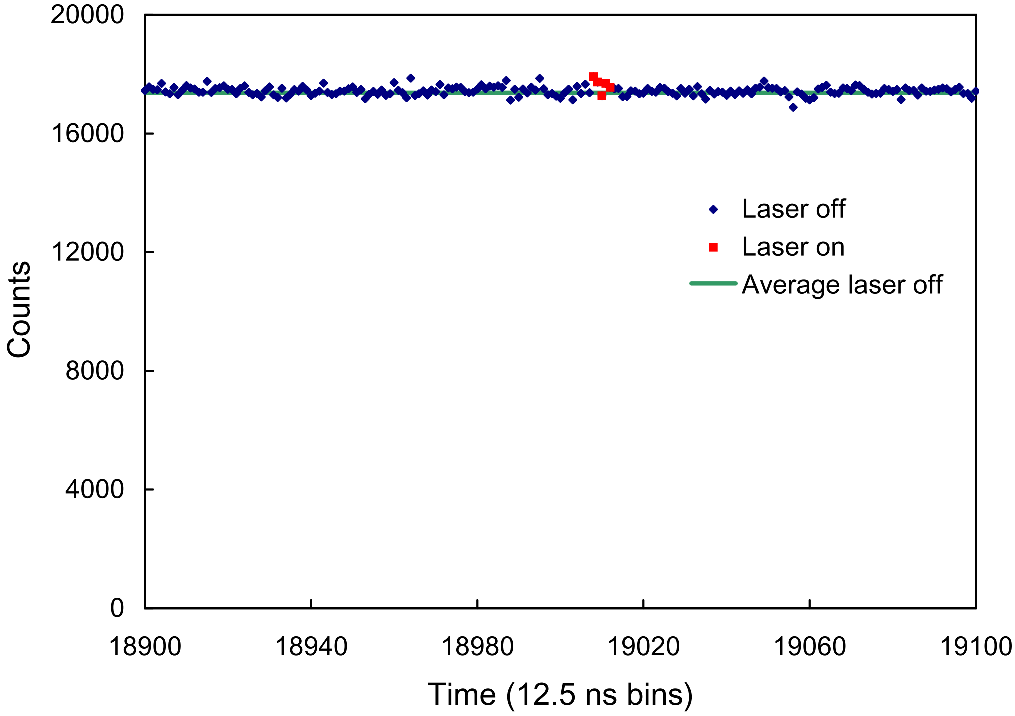

Figure 11. Timing spectrum for the circular 2° data point in figure 10. The spectrum took a week to obtain and the free–free signal (red squares) is at the 4σ level above the average laser-off signal.

one photon energy below the position of the maximum in the energy loss spectrum (i.e., absorption), and is indicated by the left vertical arrow in figure 9. The lower, triangular, data point at 2° is a measurement at an energy loss corresponding to the position of the minimum in the energy loss spectrum indicated by the right vertical arrow in figure 9; it should contain a contribution not only from photon *emission* during $3p \to 4s$ excitation, but also photon *absorption* during higher excitations such as $3p \to 3d$, etc. Zon's model is given in terms of the elastic scattering amplitude; it is unclear how it should be modified to be used for inelastic scattering, so it is not shown in figure 10.

Both these data points necessitated long run times to achieve statistically significant results. Figure 11 shows the timing spectrum for the 2° absorption experiment which lasted one week in order to obtain a free–free signal at the 4.4σ level. (The other 2° experiment is significant at the 4.5σ level; the timing spectra for both types of 2° experiments added together yield a result significant at the 6.27σ level.) These free–free experiments in Ar are continuing; the whole inelastic scattering angular distribution $0 \to 90°$ needs to be measured with adequate statistics, in order to confirm the existence of dressed atom effects.

4. Summary and conclusions

We have carried out experiments which support the existence of dressed atom effects due to the presence of a laser field during the electron-impact excitation of Ar. The effects have been observed at a scattering angle of 2°, as a free–free signal greatly in excess of that predicted by the KWA.

It should be stressed that these are preliminary results that need to be verified before dressed atom effects can be confirmed for the electron-impact excitation of Ar in a laser field.

Acknowledgments

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