PHYSICAL REVIEW A VOLUME 51, NUMBER 1 JANUARY 1995

Field-induced dipole effects in laser-assisted elastic electron-atom scattering

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(Received 31 August 1994)

Recent measurements in laser-assisted elastic electron-atom scattering at small angles are not consistent with Kroll-Watson theory. The added effect of the polarization of the atom by the laser field is studied for a typical atomic static field and polarizability. It is found that this effect makes a negligible contribution in the CO_2 laser intensity range used in the measurements.

PACS number(s): 34.80.Qb, 03.65.Nk

Elastic scattering of electrons by atoms in the presence of moderately intense CO_2 laser fields has been under laboratory study since 1977, mainly by the group of Weingartshofer, Wallbank, and collaborators [1–7]. They have observed as high as 11-photon absorption and emission processes in scattering through large angles ($\sim 155°$) with microsecond laser pulses of average intensities of 10^6–10^8 W/cm^2. A comprehensive review by Mason [8] covers all work prior to 1993. The theoretical basis for understanding these results is the Kroll-Watson formula [9], which is expected to be valid for potential scattering and frequencies as low as that of the CO_2 laser (photon energy $=0.117$ eV or 0.00430 a.u.). Since a collision time for a 10-eV electron is about 10^{-15} s, the laser intensity in a μs pulse may be assumed constant over each collision, and one would expect that a pulse intensity average of the Kroll-Watson formula should provide a reasonable description of these experimental results. Actually, such a detailed averaging has not yet been carried out, but the symmetrical pattern of the peaks and their relative magnitudes, as well as their consistency with the associated sum rule, have led to the conclusion that these early results were indeed consistent with the Kroll-Watson theory.

In the most recent measurements, Wallbank and Holmes [6,7] have looked at the free-free spectra of electrons scattered by He at *small angles* and with similar laser pulses, and have found rather dramatic deviations from the predictions of the Kroll-Watson formula. They have observed up to five-photon processes, which gave a fractional change in the laser-assisted signal compared with the no-laser signal for $|n| = 5$ of about 0.1%, while the Kroll-Watson prediction was a factor of $\sim 10^{-16}$. Such a large discrepancy was completely unexpected, and the authors have suggested that it may arise from the neglect (in the Kroll-Watson formula) of atom distortion effects such as the dipole potential which would be induced by the laser field. This unsettled situation is the motivation for the present work, in which we estimate the expected effect of the laser-induced dipole field on the free-free transition process. Other earlier theoretical work [10,11] has been done on the "dressing" of the atom by the field in terms of a more formal treatment. Numerical results were obtained for different values of electron energy, laser frequency, and intensity than those that apply to the recent measurements [6,7]. The application of those methods to the present experimental case would be expected to yield results very similar to those found in the present calculation.

We start with a brief review of Kroll-Watson theory (KW), which predicts the laser-assisted differential cross section

$$\frac{d\sigma^{\rm KW}(n)}{d\Omega}=\frac{k_f(n)}{k_i}J_n^2(\boldsymbol{\alpha}_0\cdot\mathbf{Q})\frac{d\sigma(\epsilon,\mathbf{Q})}{d\Omega}, \qquad (1)$$

where $\boldsymbol{\alpha}_0=\mathbf{E}_0/\omega^2$ for a linearly polarized field of amplitude $\mathbf{E}_0$ and frequency ω, $\mathbf{k}_i$ and $\mathbf{k}_f(n)$ are the initial and final electron wave vectors, $\mathbf{Q}$ is the momentum transfer $\mathbf{k}_f(n)-\mathbf{k}_i$, and energy conservation requires that $k_f^2(n) = k_i^2+2n\omega$. (We use atomic units unless otherwise specified.) The field-free elastic differential cross section on the right-hand side of (1) is evaluated at the shifted effective energy

$$\epsilon=\tfrac{1}{2}k_i^2+n\omega\frac{(\hat{\mathbf{E}}_0\cdot\mathbf{k}_i)}{(\hat{\mathbf{E}}_0\cdot\mathbf{Q})}+\frac{(n\omega)^2}{2(\hat{\mathbf{E}}_0\cdot\mathbf{Q})^2} \qquad (2)$$

and effective scattering angle θ', which corresponds to the momentum transfer $\mathbf{Q}$. The kinetics gives the explicit relation $\cos\theta'=1-Q^2/4\epsilon$. We see that the condition of $\mathbf{E}_0$ being perpendicular to $\mathbf{Q}$ will lead to singularities in ϵ for $|n| \neq 0$, where the strict KW formula should not be used. This singularity is an artifact of an approximation used by Kroll and Watson, and does not appear in more general derivations [12]. The low-frequency (LF) ("soft-photon") formula

$$\frac{d\sigma^{\rm LF}(n)}{d\Omega}=\frac{k_f(n)}{k_i}J_n^2(\alpha_0\cdot\mathbf{Q})\frac{d\sigma(E_i,\mathbf{Q})}{d\Omega}, \qquad (3)$$

where $E_i=k_i^2/2$ agrees with KW for $n=0$ at any frequency, as well as for any n in the $\omega\to 0$ limit. The low-frequency formula is the preferred one in the region where the singularity in ϵ occurs in the Kroll-Watson formula. Here the momentum transfer $\mathbf{Q}$ corresponds to the actual scattering angle θ. For the CO_2 laser frequency we expect good agreement between (1) and (3) everywhere, except in the immediate vicinity of the singularity in ϵ. The above-mentioned sum rule is seen to follow immediately by summing (3)

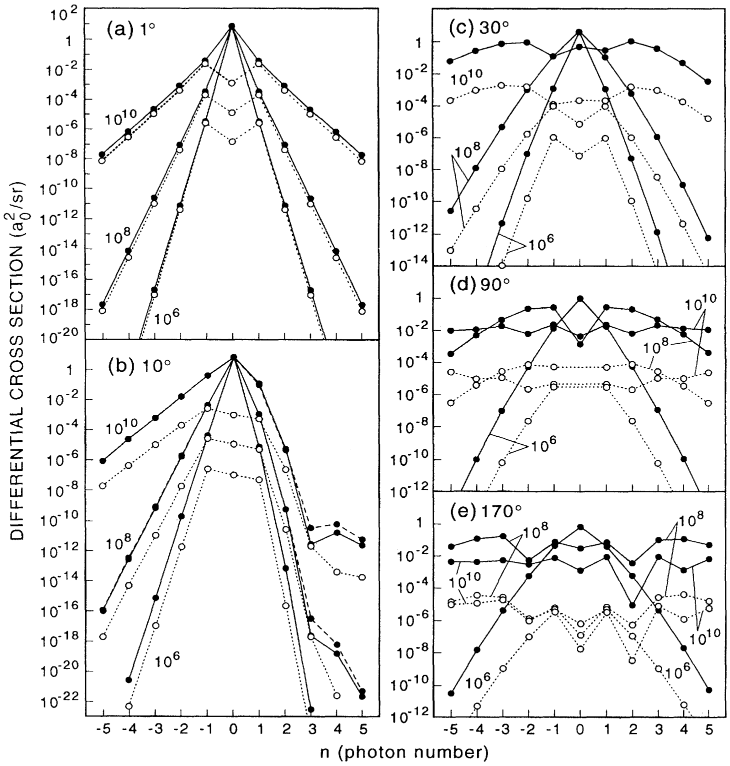

FIG. 1. Calculated laser-assisted electron-atom differential scattering cross sections as a function of photon number (absorbed, $n>0$ or emitted, $n<0$) for $E_i=10$ eV and for the scattering angles (a) 1°, (b) 10°, (c) 30°, (d) 90°, and (e) 170°, at the CO_2 laser intensities 10^6, 10^8, and 10^{10} W/cm^2. The solid points connected by solid lines are the Kroll-Watson $d\sigma^{\rm KW}/d\Omega$. The low-frequency approximation $d\sigma^{\rm LF}/d\Omega$ agrees with $d\sigma^{\rm KW}/d\Omega$ in all cases except for $n=2-5$ for $\theta=10°$, where the $d\sigma^{\rm LF}/d\Omega$ points are connected by a dashed line. The magnitudes of the first-order polarization contributions $|d\sigma_P^{(1)}/d\Omega|$ are given by open points connected by dotted lines. (For $\theta=90°$, $n=0$, $d\sigma_P^{(1)}/d\Omega$ vanishes identically, so no point is shown.) These cross sections are defined only at the integer photon points, and the connecting lines are included purely as a visual aid. Correspondingly, observed spectra would consist of peaks that are the convolution of the product of these cross sections and $\delta(E-E_n)$ with the instrumental energy resolutions.

$$\sum_{n=-\infty}^{\infty} \frac{d\sigma^{\rm LF}(n)}{d\Omega} \xrightarrow[\omega\to 0]{} \frac{d\sigma(E_i,\mathbf{Q})}{d\Omega}, \quad (4)$$

since $k_f(n) \xrightarrow[\omega\to 0]{} k_i$ and $\Sigma_n J_n^2(x)=1$. The large-angle experiments generally confirm that scattering flux in the field-free case is diverted into the n-photon peaks, in agreement with this theoretical expectation.

We will present numerical results for the KW and LF formulas for the case of the assumed typical atomic static potential field,

$$V_{\rm stat}(r)=-e^{-2r}\left(1+\frac{1}{r}\right)-\frac{\alpha_p}{2}(1-e^{-r})^6 \Big/ r^4. \quad (5)$$

We neglect electron exchange, an effect that may be appreciable at these energies. The first term may be recognized as the static field which arises in H(1s), and the second term is the polarization potential that is induced by an incident electron (and has been arbitrarily smoothed out at the origin). We will use the value of $\alpha_p=4.5$ a.u., which is the polarizability of ground-state hydrogen. Although these parameters correspond to the H atom rather than the He atomic target used in the measurements, they are quite adequate for the description needed to try to understand the huge qualitative discrepancy between Kroll-Watson theory and the small-angle measurements. Since the polarizability of He is about 1/3 that of H, our present estimates of field-induced polarization effects may be expected to be on the high side. We evaluate the scattering phase shifts for this potential, which gives us the field-free differential cross sections needed to evaluate the laser-assisted KW and LF cross sections in (1) and (3). These are shown in Fig. 1 for $E_i=10$ eV and for a number of scattering angles and laser intensities, and with the polarization direction taken to be along the direction of electron incidence, as it was taken in the recent experiments [6,7]. The KW singularity in ϵ is approached only in the case of $n>0$ for 10° scattering, and the resulting differences between KW and LF in that region are shown. For all other cases KW and LF are indistinguishable on the logarithmic plot. The largest energy shift for the cases presently studied occurs for $n=3$ and $\theta=10°$, where $\epsilon=37.0$ a.u., an enormous shift from $E_i=0.368$ a.u. The corresponding shift of the KW from the LF cross section as seen in Fig. 1(b) is about a factor of 10. The cross sections for other values of θ shown in Fig. 1 show a greater symmetry in absorption and emission of any particular number of photons. This asymmetry is the result of $\mathbf{E}_0$ or $\boldsymbol{\alpha}_0$ being almost perpendicular to $\mathbf{Q}$, which increases the sensitivity of the magnitude of $\boldsymbol{\alpha}_0\cdot\mathbf{Q}$ to n, and hence the sensitivity of $J_n^2(\boldsymbol{\alpha}_0\cdot\mathbf{Q})$ to the sign of n.

To now make an estimate of the effect of the field-induced polarization, we note that in the same way that the polarization part of the static potential, $\sim\alpha_p/2r^4$, was produced by the incident electron polarizing the H(1s) atom, the laser field $\mathbf{E}_0\sin\omega t$ will also polarize it, producing a time-varying dipole moment, $-\alpha_p\mathbf{E}_0\sin\omega t$, and the additional dipole interaction between the incident electron and atom,

$$V_{\rm pol}(\mathbf{r},t)=\alpha_p\frac{\mathbf{E}_0\cdot\hat{\mathbf{r}}}{r^2}\sin\omega t. \quad (6)$$

As this additional interaction is both nonspherical and time-dependent, we will treat it here only in first order. The first-order S matrix for scattering between Volkov states, which describe a free electron in a laser field, is

$$S_{if}^{(1)}=-i\int_{-\infty}^{\infty}dte^{-i(E_i-E_f)t}\int d\mathbf{r}\ e^{i\mathbf{k}_i\cdot[\mathbf{r}-\mathbf{r}_0(t)]}(V_{\text{stat}}+V_{\text{pol}})\times e^{-i\mathbf{k}_f\cdot[\mathbf{r}-\mathbf{r}_0(t)]}, \tag{7}$$

where $\mathbf{r}_0(t)=\alpha_0\sin\omega t$ corresponds to the same choice of gauge as used by Kroll and Watson. Making the usual Bessel function expansion, $e^{ix\sin(\omega t)}=\Sigma_{n=-\infty}^{\infty}J(x)e^{in\omega t}$, and carrying out the time integral to get the energy-conserving δ functions, we find that the amplitude for an n-photon transition is $T_S^{(1)}(n)+T_P^{(1)}(n)$, where

$$T_S^{(1)}(n)=J_n(\boldsymbol{\alpha}_0\cdot\mathbf{Q})f_{B1}(Q) \tag{8a}$$

and

$$T_P^{(1)}(n)=[J_{n-1}(\boldsymbol{\alpha}_0\cdot\mathbf{Q})-J_{n+1}(\boldsymbol{\alpha}_0\cdot\mathbf{Q})]f_{P1}(\mathbf{Q}). \tag{8b}$$

Here f_{B1} is the usual static-field first-Born amplitude,

$$f_{B1}=-\frac{1}{2\pi}\int d\mathbf{r}\ e^{-i\mathbf{Q}\cdot\mathbf{r}}V_{\text{stat}}(r)=-\frac{2}{Q}\int_0^{\infty}dr\ rV_{\text{stat}}(r)\sin Qr, \tag{9a}$$

and f_{P1} is the first-order polarization field amplitude,

$$f_{P1}=-\frac{i}{2\pi}\int d\mathbf{r}\ e^{-i\mathbf{Q}\cdot\mathbf{r}}\left[\frac{\alpha_p}{2}\frac{\mathbf{E}_0\cdot\hat{\mathbf{r}}}{r^2}\right]=-\alpha_{\text{p}}\frac{\mathbf{E}_0\cdot\hat{\mathbf{Q}}}{Q}. \tag{9b}$$

This latter expression has also been obtained in the work of Byron *et al.* [11]. Both the static-field and polarization amplitudes are real, and hence an interference is possible in principle. In fact, however, over the intensity range considered, 10^6–10^{10} W/cm^2, we find the contribution of the polarization term to be negligible for all angles except for forward scattering, where the two contributions may be of the same order of magnitude.

For the purpose of showing the contribution of the laser-induced polarization term let us write the first-order n-photon cross section as

$$\frac{d\sigma^{(1)}(n)}{d\Omega}=\frac{d\sigma_S^{(1)}(n)}{d\Omega}+\frac{d\sigma_P^{(1)}(n)}{d\Omega}, \tag{10}$$

where

$$\frac{d\sigma_S^{(1)}(n)}{d\Omega}=\frac{k_f(n)}{k_i}J_n^2(\boldsymbol{\alpha}_0\cdot\mathbf{Q})|f_{B1}|^2 \tag{11}$$

is the usual first Born approximation to $d\sigma^{\text{LF}}(n)/d\Omega$, and $d\sigma_P^{(1)}(n)/d\Omega$ is the remaining contribution arising from the laser-induced polarization. Since $d\sigma_P^{(1)}(n)/d\Omega$ contains, and will normally be dominated by, the cross term $\sim T_S^{(1)}(n)\times T_P^{(1)}(n)$, it may be negative, so we show its magnitude only in Fig. 1.

We find that the maximum difference between $d\sigma^{\text{LF}}(n)/d\Omega$ and its first-order approximation, $d\sigma_S^{(1)}(n)/d\Omega$, is a factor 2 for all cases evaluated, and that difference is small (except for forward scattering) compared with the differences between static field contributions (full or first Born) and the laser-induced polarization contributions. The maximum relative magnitudes of $|d\sigma_P^{(1)}(n)/d\Omega|$ occur for the almost forward scattering case ($\theta=1°$), as expected, because of the long range of the dipole force interaction, where its effect could be expected to produce the order of less than a factor 2 correction in the Kroll-Watson cross section. For the case closest to the experimental conditions [6,7], $\theta=10°$ and $I=10^8$ W/cm^2, it can be seen from Fig. 1(b) that the polarization contribution is ≤ 0.1 of the KW cross section for each n. In order to get the ratio $[d\sigma(5)]/d\Omega]/[d\sigma(0)/d\Omega]$ to reach as high a value ($\sim 0.1\%$) as reported in the recent small-angle measurements [6,7], one would require an average laser intensity of at least 10^{11} W/cm^2, which is not the case for the pulses used (average intensity $\sim 10^8$ W/cm^2). On the other hand, if we compare the earlier large-angle data; for example, that contained in Ref. [3] for $\theta=155°$, with our Fig. 1(e) results for $\theta=170°$, we note that an average intensity in the range $10^8>\bar{I}>10^6$ W/cm^2 would allow for a reasonable fit to the data. Such an average intensity is completely consistent with the measured temporal characteristics of the pulse.

We have also evaluated all of these cross sections for two other geometries: $\mathbf{E}_0$ perpendicular to $\mathbf{k}_i$ with (1) $\mathbf{k}_f$ in the $\mathbf{E}_0$-$\mathbf{k}_i$ plane, and (2) $\mathbf{k}_f$ in the plane perpendicular to $\mathbf{E}_0$. In geometry (1) the polarization contributions are of the same order of smallness or less than those in Fig. 1, and in geometry (2) all $n\neq 0$ processes are suppressed because $\alpha_0\cdot\mathbf{Q}$ vanishes. We conclude, on the basis of the present numerical results for a typical static atomic potential and polarizability, that the correction of the Kroll-Watson formula arising from the polarization of the atom target by the laser field is far below what is needed to explain the data [6,7]. The actual peak dipole moment α_pE_0 that is induced by laser intensities in the range 10^6 to 10^{10} W/cm^2 (with $\alpha_p=4.5$ a.u.) is 2.4×10^{-5} to 2.4×10^{-3} a.u., which is a rather small displacement of the center of the electron cloud relative to its normal radius of 1 a.u.

We are thus left with the large differences between experiment and our current theoretical understanding of this process. The dilemma is compounded by the fact that the earlier large-angle measurements are quite consistent with the Kroll-Watson formula, while the newer small-angle data are totally inconsistent with it. A very recent study of this problem using classical mechanics [13] has also come to the conclusion that the effects of laser-induced polarization are negligible. We hope this unsatisfactory situation may be somehow resolved in the not-too-distant future.

I have benefited from correspondence and discussions with Barry Wallbank and Leonard Rosenberg.

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