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COMMENT

Collective field effects in electron–atom scattering in a low-frequency laser field

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Abstract. Several recent papers have examined the theory of electron–atom scattering in a low-frequency laser field. Only one of these papers obtained rough agreement with the size of the experimental cross section through the identification of a novel physical process: collective field effects. The purpose of this paper is to demonstrate that an error in the formulation led to an overestimate of the interaction potential by five orders of magnitude. The experimental results cannot be explained by collective field effects.

Recent experiments by Wallbanks and Holmes (1993, 1994) and Bader (1986) have triggered significant interest in the scattering of electrons by atoms in a low-frequency laser field. These experiments were performed in a geometry such that the momentum transferred to the electron during the scattering process was perpendicular to the laser polarization. In this geometry, the cross section for absorbing or emitting photons is greatly reduced from cross sections where a component of the momentum transfer is parallel to the laser polarization. This reduction arises, crudely speaking, because the laser field cannot do work on the electron in this geometry. The theoretical interest in the problem results from the measured cross section being orders of magnitude larger than expected.

To a large extent, the expectations are based on the Kroll–Watson approximation. Therefore, it is natural to probe the assumptions in this approximation for an explanation of the experimental results. Geltman (1995) probed quantum mechanically and Rabadán *et al* (1994) semiclassically the assumption that the laser does not affect the shell structure of the atom. The dominant effect is that the laser induces a dipole moment on the atom which can interact with the electron through a long-range z/r^3 potential. The cross section from this interaction was too small to explain the experimental results. Collins and Csanak (1995), Chen and Robicheaux (1996) and Geltman (1996) probed the assumption that the Kroll–Watson approximation was valid when the momentum transfer is perpendicular to the laser polarization. In the first two works, essentially exact close-coupling methods were employed but the theoretical cross section was found to be too small by factors of roughly 5–10. Geltman employed perturbation theory to show that the Kroll–Watson approximation was not valid for weak lasers and by implication not valid for strong lasers; the large laser fields in the experiments precluded a direct comparison although the form of the differential cross section was reproduced. Chen and Robicheaux also found that the exchange interaction and Feshbach resonances in the electron–atom scattering could not explain the experimental results. Madsen and Taulbjerg (1995) generalized the Kroll–Watson approximation so that the approximations were valid for the experimental parameters; their

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differential cross section did not have the experimental shape and was orders of magnitude too small for most of the angles measured. The only possible explanation in this group of papers was the suggestion of Madsen and Taulbjerg of an unexplained uncertainty in the experimental momentum transfer direction of 20° (Chen and Robicheaux state 'We find the shifted angle would need to be as large as 20° to account for the difference between experiment and theory...').

A recent paper by Varro and Ehlotzky (1995) claimed that the experimental results could be explained by the interaction of the electron with a collective potential obtained from adding the potentials from all of the atoms in the laser beam. In contrast, Chen and Robicheaux state 'The probability of double scattering under this (experimental) condition is negligible'. It is the purpose of this paper to indicate the error in the derivation of Varro and Ehlotzky. When the correct interaction potential is utilized, the collective field potential does not have any effect on the scattering cross section. The rough agreement they obtained with experiment was completely fortuitous. This paper will present arguments at three levels (intuitive physical picture, perturbation theory, exact numerical time-dependent calculation) against their potential and for the replacement of their potential with one identical in form but $\sim 10^5$ times smaller.

The derivation of the collective potential is based on the interaction of an electron with one atom in a laser field. To find the collective potential, sum the potentials from all of the atoms in the laser with the atoms randomly distributed. Varro and Ehlotzky obtained the wrong potential for an electron interacting with one atom. For simplicity, the following derivation will be for the atom at the origin and atomic units will be used throughout this paper. The main interest is in the asymptotic form of the potential when the electron is far from the atom. Therefore in the following the scattering electron will be considered distinguishable from the atomic electrons. In all that follows, electrons $1-N$ will be atomic electrons (with a properly antisymmetrized wavefunction) and electron $N+1$ will be the scattering electron.

The starting point of Varro and Ehlotzky's potential is the examination of the Hamiltonian in the acceleration gauge for all of the electrons:

$$H = \sum_{j=1}^{N+1} \frac{p_j^2}{2} - \frac{N}{|\boldsymbol{r}_j + \boldsymbol{\alpha}(t)|} + \sum_{i<j}^{N+1} \frac{1}{|\boldsymbol{r}_j - \boldsymbol{r}_i|} \tag{1}$$

where $\boldsymbol{\alpha}(t) = -\alpha_0 \hat{z} \sin(\omega t)$ with $\alpha_0 = A_0/\omega c = E_0/\omega^2$ (A_0 is the amplitude of the vector potential and E_0 is the amplitude of the electric field). For the frequencies and field strengths in the experiments $\alpha_0 \simeq 1.7$ au. Varro and Ehlotzky found the potential to lowest order in the field strength by expanding the potentials with $\boldsymbol{r}_{N+1}$ in inverse powers of r_{N+1}

$$V(\boldsymbol{r}_{N+1}, t) \simeq \langle \psi_{\text{atom}} | \sum_{j=1}^{N} (\boldsymbol{\alpha}(t) + \boldsymbol{r}_j) | \psi_{\text{atom}} \rangle \cdot \boldsymbol{r}_{N+1}/r_{N+1}^3 \tag{2}$$

where ψ_{atom} is the solution of only the atomic part of the Hamiltonian in (1):

$$H = \sum_{j=1}^{N} \frac{p_j^2}{2} - \frac{N}{|\boldsymbol{r}_j + \boldsymbol{\alpha}(t)|} + \sum_{i<j}^{N} \frac{1}{|\boldsymbol{r}_j - \boldsymbol{r}_i|}\,. \tag{3}$$

This step is correct. They next incorrectly applied perturbation theory by using the unshifted ground-state wavefunction for ψ_{atom} ($= \psi_g(\boldsymbol{r}_1, \boldsymbol{r}_2, \ldots, \boldsymbol{r}_N) \exp(-\mathrm{i} E_g t)$) because they only want the potential to lowest order in the field strength. Because $\langle \psi_g | \boldsymbol{r}_j | \psi_g \rangle = 0$, this gives a potential

$$V(\boldsymbol{r}_{N+1}, t) = \boldsymbol{\alpha}(t) \cdot \boldsymbol{r}_{N+1}/r_{N+1}^3. \tag{4}$$

This application of perturbation theory is incorrect because the perturbation on the atom is not perturbative. The change in the atomic potential from the laser, $\Delta V = \sum_j N/r_j - N/|\boldsymbol{r}_j + \boldsymbol{\alpha}|$, is enormous because α is swinging back and forth with an amplitude of nearly 2 au which is larger than the size of the atomic shells.

Refutation by appeal to a physical picture. The potential obtained by Varro and Ehlotzky is unreasonable because the atomic wavefunction they employ is unphysical. In the experiments, $\omega = 0.0043$ au giving a period of $\tau = 1460$ au. In the Hamiltonian, equation (3), the nucleus is slowly swinging back and forth with an amplitude of $\sim$1.7 au. Why do the electrons stay centred at the origin? They do not stay centred, of course. They track the nucleus giving an atomic wavefunction $\psi_{\text{atom}} = \psi_{\text{g}}(\boldsymbol{r}_1 + \boldsymbol{\alpha}(t), \boldsymbol{r}_2 + \boldsymbol{\alpha}(t), \ldots)\exp(-\mathrm{i}E_{\text{g}}t)$. Using this wavefunction gives $V(\boldsymbol{r}_{N+1}, t) = 0$ which is not exactly correct but gives more accurate results than the potential in (4). To obtain the correct asymptotic potential a more accurate atomic wavefunction needs to be found.

Refutation by correct application of perturbation theory. The form of the asymptotic potential can be obtained from the correct application of perturbation theory and by the correct choice of gauge for the electrons. The correct gauge is prescribed by the form of the laser interaction: $E_0 z$ in the length gauge, $E_0 p_z/\omega$ in the velocity gauge, and $E_0 z/(r^3\omega^2)$ in the acceleration gauge. For the scattering electron, r is very large so the acceleration gauge is best. For the atomic electrons, distances and momenta are $\sim$1 au and thus the length gauge is the correct choice since $1/\omega \sim 230$ is a large number. This gives a Hamiltonian

$$H = \sum_{j=1}^{N}\left(\frac{p_j^2}{2} - \frac{N}{r_j} + E(t)z_j\right) + \sum_{i<j}^{N}\frac{1}{|\boldsymbol{r}_j - \boldsymbol{r}_i|} + \frac{p_{N+1}^2}{2} + \bar{V}(\boldsymbol{r}_{N+1}, t) \tag{5}$$

where $E(t) = A_0\omega\sin(\omega t)/c$ and

$$\bar{V}(\boldsymbol{r}_{N+1}, t) = -\frac{N}{|\boldsymbol{r}_{N+1} + \boldsymbol{\alpha}(t)|} + \sum_{j=1}^{N}\frac{1}{|\boldsymbol{r}_{N+1} + \boldsymbol{\alpha}(t) - \boldsymbol{r}_j|}\,. \tag{6}$$

The atomic wavefunction can be obtained by a perturbative expansion of the wavefunction in powers of $E_0 = A_0\omega/c$ (unlike the expansion parameter of Varro and Ehlotzky, $A_0/\omega c$, this expansion parameter is small, $A_0\omega/c \sim 3\times 10^{-5}$). The wavefunction accurate to first order in $A_0\omega/c$ is

$$\psi_{\text{atom}} = \mathrm{e}^{-\mathrm{i}E_{\text{g}}t}\left\{\psi_{\text{g}} + \sum_n \psi_n[B_n\sin(\omega t) + C_n\cos(\omega t)]\right\} \tag{7}$$

where the coefficients are given by

$$B_n = \frac{A_0\omega}{c}\langle\psi_n|\sum_{j=1}^{N}(z_j - R_z)|\psi_{\text{g}}\rangle\Big/\left(E_{\text{g}} - E_n - \frac{\omega^2}{E_{\text{g}} - E_n}\right)$$

$$\simeq \frac{A_0\omega}{c}\langle\psi_n|\sum_{j=1}^{N}(z_j - R_z)|\psi_{\text{g}}\rangle/(E_{\text{g}} - E_n) \tag{8a}$$

$$C_n = -\mathrm{i}\omega B_n/(E_{\text{g}} - E_n) \simeq 0 \tag{8b}$$

where the small size of the laser frequency compared to the differences in atomic energies has been used. This wavefunction may be used to obtain the asymptotic form of the potential

$$V(\boldsymbol{r}_{N+1}, t) = \langle\psi_{\text{atom}}|\bar{V}(\boldsymbol{r}_{N+1}, t)|\psi_{\text{atom}}\rangle = -\alpha_{\text{d}}E(t)z_{N+1}/r_{N+1}^3 \tag{9}$$

where α_{d} is the static dipole polarizability of the atom. For He, $\alpha_{\text{d}} = 1.32$ au. This potential is not new and was used by Geltman (1995) and Rabadán *et al* (1994). Note that

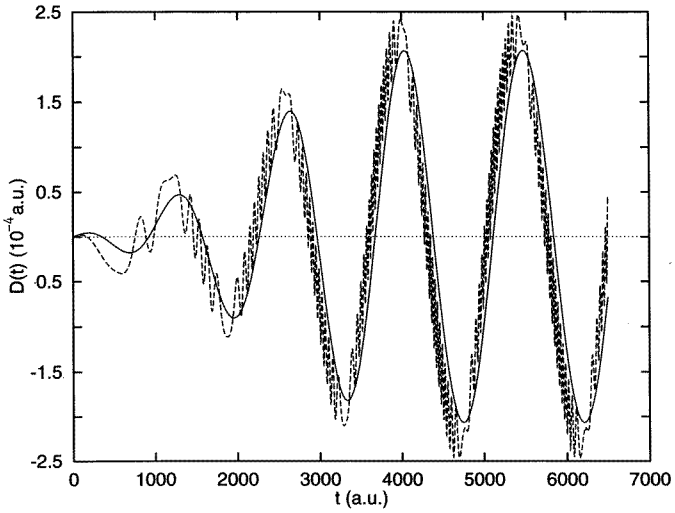

Figure 1. Plot of $D(t)$, equation (11), from direct time propagation of the atomic wavefunction (broken curve) and correct application of perturbation theory (full curve). The fast oscillation in the numerical $D(t)$ arises from the fast laser rise time. Varro and Ehlotzky's value would be 10^4 times larger.

this potential is of the same form as (4) but smaller by a factor of $\alpha_d\omega^2/N \sim 10^{-5}$. Since the form of the potential is the same, all of the resulting formulae in Varro and Ehlotzky may be used but with the smaller factor in the potential. The result is that collective field effects play no role in the experiments.

Refutation by exact numerical solution. Finally, the status of computers and numerical techniques allows an exact calculation of the atomic wavefunction. The form of the asymptotic potential does not depend on the type of atom so calculations will be performed for a soft-core hydrogen atom in cylindrical coordinates. The time-dependent potential in the acceleration gauge was chosen to be $V(\boldsymbol{r}, t) = -1/\sqrt{\rho^2 + \Delta\rho^2 + (z + \alpha(t))^2}$ with $\alpha(t)$ chosen to match the experimental field strength and frequency. The wavefunction was solved for on a grid in ρ and z with $\Delta\rho = 0.2$ au and $\Delta z = 0.2$ au (100 grid points in ρ and 200 grid points in z). The kinetic energy operator was approximated by a three point difference in each direction. The time propagation was carried out with the second-order leapfrog algorithm

$$\begin{aligned}\psi(t + \Delta t) &= \psi(t - \Delta t) - 2\mathrm{i}\Delta t H(t)\psi(t)\\ \psi(t + 2\Delta t) &= \psi(t) - 2\mathrm{i}\Delta t H(t + \Delta t)\psi(t + \Delta t)\end{aligned} \tag{10}$$

which has the useful property of exactly unitary propagation as long as $1/\Delta t$ is larger than the largest eigenvalue of the discretized Hamiltonian. The largest value of Δt that could be used with the Hamiltonian was $\Delta t = 0.01$ au.

The form of the asymptotic potential depends on

$$D(t) \equiv \langle\psi_{\mathrm{atom}}|z + \alpha(t)|\psi_{\mathrm{atom}}\rangle. \tag{11}$$

Varro and Ehlotzky's value for this parameter is $D(t) = \alpha(t)$, whereas the correct form should be $D(t) = -\alpha_d\, \mathrm{d}^2\alpha(t)/\mathrm{d}t^2$. The static polarizability of the softcore atom was obtained by fitting the numerical ground-state energy in an electric field F to $E_g(F) = E_g(0) + \alpha_d F^2/2$ giving a value of $\alpha_d = 6.61$ au. The difficulty with the direct numerical

solution of ψ_{atom} is that the laser period $\tau = 1460$ au which means 146 000 time steps are needed to propagate the wavefunction for one period. Three calculations were performed with different rise times of the laser. A rise time of zero laser periods gave a $D(t)$ with a fast oscillation with amplitude 0.01 au. With a rise time of one laser period, $D(t)$ tracked the expected value of $-\alpha_{\text{d}}\,\mathrm{d}^2\alpha(t)/\mathrm{d}t^2$ but with a fast oscillation of amplitude 2.5×10^{-4} au superimposed. In the third calculation, the $\alpha(t) = -\alpha_0 \sin(\omega t)\sin^2(\omega t/12)$ for $t \leqslant 3\tau = 4380$ au then $\alpha(t) = -\alpha_0 \sin(\omega t)$ for $t \geqslant 3\tau$ where $\alpha_0 = 1.69$ au and $\omega = 0.0043$ au. The laser is turned on over a time of three laser periods. The numerical result for $D(t)$ is plotted in figure 1 as a broken curve while the perturbation theory value $D(t) = -\alpha_{\text{d}}\,\mathrm{d}^2\alpha(t)/\mathrm{d}t^2$ is plotted as a full curve. Outside of the small amplitude, fast oscillation from the fast laser rise time, the two results are in wonderful agreement. Note, the result given by Varro and Ehlotzky is 10 000 times larger than the functions plotted in figure 1.

In conclusion, an error in the derivation of Varro and Ehlotzky caused an overestimate of the collective interaction potential by a factor of 10^5 for He. Collective field effects do not play a role in the experiments measuring electron–atom scattering in a laser.

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