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LETTER TO THE EDITOR

Coherent electron beam excitation of a model hydrogen atom

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Abstract. Excitation of a model hydrogen atom by a coherent electron beam is calculated by direct solution of the time-dependent Schrödinger equation on a three-dimensional lattice. The Coulomb interaction between the electrons is described by $v(r_1, r_2) = \frac{1}{r_>}$ and all angular momenta are set to zero. The coherent beam consists of moving wavepackets for each of two free electrons which arrive at the third bound electron at different times. As recently predicted by Robicheaux and Noordam (2000 *Phys. Rev. Lett.* **84** 3735), the probability for a weak excitation should vary as a function of arrival time. The time-dependent calculations for the model hydrogen atom show that the 1s $\rightarrow$ 2s excitation probability oscillates about an axis which is between one and two times the excitation probability from only one wavepacket. The frequency of the probability oscillation is found to be equal to the 1s $\rightarrow$ 2s transition frequency. Large destructive interference of the 1s $\rightarrow$ 2s cross section is illustrated by contour maps of the 2s projected probability as a function of wavepacket arrival time.

Accurate cross sections for inelastic scattering processes involving the interactions of electrons with atoms and their ions are important for the microscopic modelling of many laboratory and astrophysical plasmas. However, as recently pointed out by Robicheaux and Noordam [1], the very concept of a fixed inelastic scattering cross section for an atomic target fails when the incident electron beam possesses some degree of longitudinal coherence. The development of 'atom lasers' [2, 3] and the 'pulsed electron gun' [4, 5] holds promise for the experimental observation of strong constructive and destructive interference effects in atomic collisions involving coherent matter beams.

In this letter, the excitation of a model hydrogen atom by a coherent electron beam is calculated by direct solution of the time-dependent Schrödinger equation on a three-dimensional numerical lattice. The model for the hydrogen atom, in which all angular momenta are set to zero, is due to Temkin [6] and Poet [7]. This particular model is very popular for testing new theoretical ideas in scattering theory [8–14], due to its numerical simplicity and its close qualitative approximation to actual electron scattering from hydrogen. The coherent beam consists of moving wavepackets for each of two free electrons which arrive at the third bound electron at different times. This three-dimensional scattering problem is solved by direct solution of the time-dependent Schrödinger equation using numerical lattice methods developed previously for the electron-impact double ionization of a model helium atom [15]. In the following paragraphs we first formulate the time-dependent wavepacket method for coherent beam scattering, then present results illustrating coherent effects in excitation probabilities and cross sections, and finally give a brief summary.

The $(N + 1)$-dimensional wavefunction, $\Phi(r_0, r_1, \ldots, r_N, t)$, is a solution to the time-dependent Schrödinger equation given by:

$$\mathrm{i}\frac{\partial \Phi(r_0, r_1, \ldots, r_N, t)}{\partial t} = H(r_0, r_1, \ldots, r_N)\Phi(r_0, r_1, \ldots, r_N, t), \tag{1}$$

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where

$$H(r_0, r_1, \ldots, r_N) = \sum_{i=0}^{N}(-\frac{1}{2}\frac{\partial^2}{\partial r_i^2} - \frac{1}{r_i}) + \sum_{i<j}\frac{1}{\max(r_i, r_j)}, \tag{2}$$

and N is the number of incident free electrons. In the following paragraphs we will present results for $N = 1$ and $N = 2$. Unless otherwise indicated, atomic units are used in all equations and results. The initial condition for the solution of the time-dependent Schrödinger equation is given by:

$$\Phi(r_0, r_1, \ldots, r_N, t = 0) = P_{1s}(r_0)G_1(r_1)\ldots G_N(r_N), \tag{3}$$

where the incoming radial wavepacket is given by:

$$G_{i'}(r_i) = \frac{1}{(\pi w^2)^{\frac{1}{4}}}\mathrm{e}^{-\frac{(r_i - s_{i'})^2}{2w^2}}\mathrm{e}^{-\mathrm{i}kr_i}, \tag{4}$$

w is the width of the packet, $s_{i'}$ is the localization radius, and $E = \frac{k^2}{2}$ is the incident energy. The $1s \rightarrow ns$ excitation probability is given by either:

$$\mathcal{P}_{1s\rightarrow ns} = N\sum_{k_1}\ldots\sum_{k_N}\left|\int_0^\infty \mathrm{d}r_0\int_0^\infty \mathrm{d}r_1\ldots\int_0^\infty \mathrm{d}r_N \right.$$
$$\left.\times P_{ns}(r_0)P_{k_1s}(r_1)\ldots P_{k_Ns}(r_N)\Psi(r_0, r_1, \ldots, r_N, t = T)\right|^2, \tag{5}$$

or approximately by:

$$\mathcal{P}_{1s\rightarrow ns} = N\int_0^\infty \mathrm{d}r_1\ldots\int_0^\infty \mathrm{d}r_N\left|\int_0^\infty \mathrm{d}r_0 P_{ns}(r_0)\Psi(r_0, r_1, \ldots, r_N, t = T)\right|^2. \tag{6}$$

The wavefunction $\Psi(r_0, r_1, \ldots, r_N, t)$ at a time $t = T$ following the collision is obtained by propagating the product wavefunction $\Phi(r_0, r_1, \ldots, r_N, t)$ of equation (3) using Schrödinger's equation on an $(N+1)$-dimensional spatial lattice and then fully antisymmetrizing the resultant spatial function. The bound, $P_{ns}(r)$, and continuum, $P_{ks}(r)$, orbitals needed in the above equations are obtained by diagonalization of the single particle Hamiltonian:

$$h(r) = -\frac{1}{2}\frac{\partial^2}{\partial r^2} - \frac{1}{r}, \tag{7}$$

on a one-dimensional spatial lattice. Finally, the $1s \rightarrow ns$ excitation cross section is given by:

$$\sigma_{1s\rightarrow ns} = \frac{\mathcal{P}_{1s\rightarrow ns}}{\mathcal{F}}, \tag{8}$$

where the incident flux is given by:

$$\mathcal{F} = \frac{Nk^2}{\pi}. \tag{9}$$

For electron scattering from hydrogen in the Temkin–Poet model, we choose a numerical spatial lattice with uniform mesh spacing $\Delta r = 0.4$ and a box size of $R = 48.0$. We first consider scattering by one free electron ($N = 1$) on a two-dimensional lattice of 120×120 points. Initially, the incoming radial wavepacket is centred at $s = 20.0$ with $w = 4.0$ and an incident energy of $E = 50.0$ eV. The ground state of hydrogen on the corresponding one-dimensional lattice has an energy of -13.1 eV. The probability density for the product wavefunction, $|\Phi(r_0, r_1, t)|^2$, is shown in figure 1 at times $t = 0.0$ and $t = 25.0$. The highest probability density of figure 1(a) is found at $r_0 = 1.0$ and $r_1 = 20.0$, corresponding to the most probable position of the 1s target electron and the initial position of the free-electron

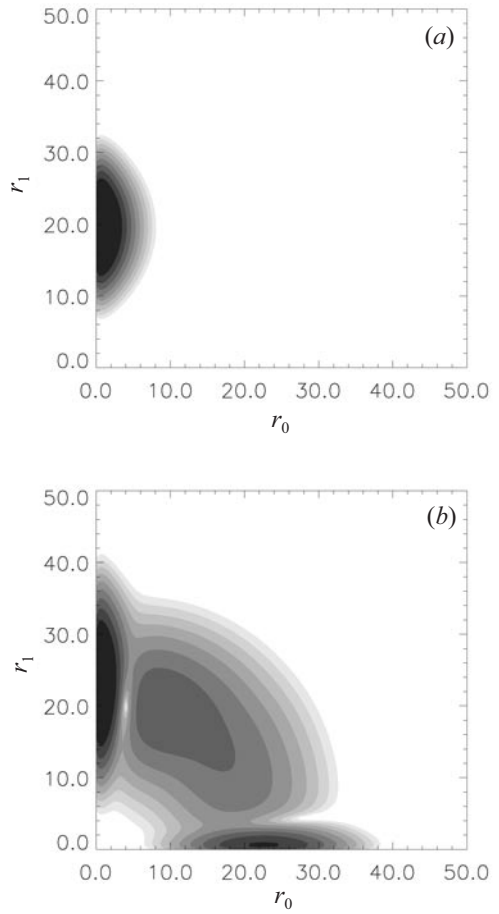

Figure 1. Scattering by one free-electron wavepacket at an incident energy of 50 eV. Probability density, $|\Phi(r_0, r_1, t)|^2$, at (*a*) $t = 0.0$ and (*b*) $t = 25.0$ (r_0 and r_1 are radial distances in atomic units).

wavepacket. The large peaks in the probability density of figure 1(*b*) along the r_0 and r_1 axes are due to elastic scattering and a small amount of target excitation. The probability density centred along $r_0 = r_1$ is due to target ionization. The 1s $\rightarrow$ 2s excitation probability is found from equation (5) to be $\mathcal{P}_{1s\rightarrow 2s} = 2.83 \times 10^{-2}$, which yields a cross section from equations (8) and (9) of $\sigma_{1s\rightarrow 2s} = 0.68$ Mb, where 1.0 Mb = 1.0×10^{-18} cm^2. The spin-averaged triplet cross section is given by $\langle\sigma_{1s\rightarrow 2s}\rangle = \frac{3}{4}\sigma_{1s\rightarrow 2s} = 0.51$ Mb. Although not needed for our purposes in this letter, our representation of the ground state energy of hydrogen and the triplet scattering probabilities and cross sections improves rapidly as the mesh spacing is decreased to $\Delta r = 0.1$.

We next consider scattering by two free electrons ($N = 2$) on a three-dimensional lattice of $120\times120\times120$ points. Initially, one incoming radial wavepacket is centred at $s_1 = 16.0$ and the other incoming radial wavepacket is centred in succession at $s_2 = 28.0$, 29.0, 30.0, 32.0, 36.0 and 38.0. For each wavepacket $w = 4.0$ and the incident energy is $E = 50$ eV. The 1s $\rightarrow$ 2s excitation probabilities as a function of wavepacket separation, $\Delta s = s_2 - s_1$, are shown in figure 2. The 1s $\rightarrow$ 2s excitation probability for $\Delta s = 20.0$ is found to be $\mathcal{P}_{1s\rightarrow 2s} = 1.56 \times 10^{-2}$, which yields a cross section of $\sigma_{1s\rightarrow 2s} = 0.19$ Mb. To consider scattering by two free electrons with larger spatial separation, we increased the size of the three-dimensional lattice to $145 \times 145 \times 145$ points. Initially, one incoming radial wavepacket

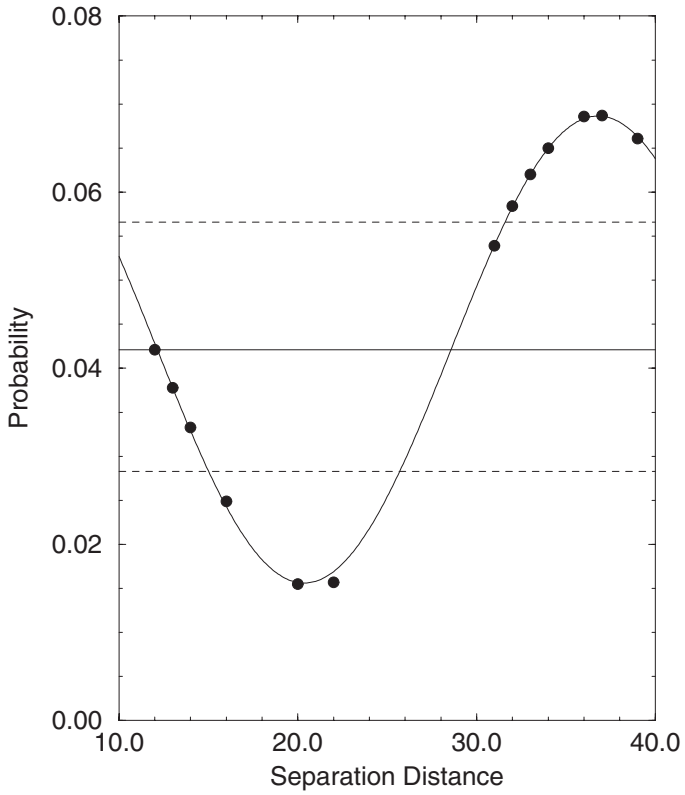

Figure 2. Probabilities for 1s → 2s excitation by two free-electron wavepackets as a function of wavepacket separation. Full circles, specific wavepacket calculations; solid curve, sine function fit; solid line, mean axis; lower dashed line, probability for scattering by one free electron; upper dashed line, twice the probability for scattering by one free electron (separation distance in atomic units).

is centred at $s_1 = 10.0$ and the other incoming radial wavepacket is centred in succession at $s_2 = 40.0, 41.0, 42.0, 43.0, 45.0, 46.0$ and 48.0. For each wavepacket $w = 4.0$ and the incident energy is $E = 50.0$ eV. The 1s → 2s excitation probabilities as a function of wavepacket separation are again shown in figure 2. The 1s → 2s excitation probability for $\Delta s = 36.0$ is found to be $\mathcal{P}_{1s\to 2s} = 6.90\times 10^{-2}$, which yields a cross section $\sigma_{1s\to 2s} = 0.83$ Mb.

As seen in figure 2, the time-dependent wavepacket calculations produce 1s → 2s excitation probabilities as a function of wavepacket separation that oscillate about an axis which is between one and two times the excitation probability from only one wavepacket. The probability on axis is $\mathcal{P}_{1s\to 2s} = 4.21 \times 10^{-2}$, where $\mathcal{P}_{1s\to 2s} = 2.83 \times 10^{-2}$ for one wavepacket. Fitting to a sine function, the wavelength of the oscillation is found to be $\lambda = 32.9$. For $E = 50$ eV incident electrons, this translates to an oscillation frequency $\omega = \frac{2\pi k}{\lambda} = 0.37$, which is precisely the transition frequency difference between the 1s and 2s bound states in the model hydrogen atom. The excitation probability should continue to oscillate for larger and larger wavepacket separations, as long as the external environment has no effect on the target hydrogen atom.

Although the 1s → 2s excitation probabilities found in the time-dependent wavepacket calculations are at the few per cent level, we are not in the 'weak' scattering limit, which forms

the basis for the coherent scattering predictions of Robicheaux and Noordam [1]. In the 'weak' scattering limit, the excitation probability for scattering by two free electrons should oscillate about an axis which is two times the excitation probability from only one wavepacket. Our conjecture is that the shortfall seen in figure 2 is due to 'strong' scattering effects, i.e. multiple scattering of the target electron and scattering between the free-electron wavepackets. To check our conjecture, we repeated the one- and two-electron wavepacket calculations on the same 120^3 and 145^3 point lattices at an incident energy of $E = 70$ eV. For these calculations, however, the interaction between all electrons, as found in equation (2), was reduced by the substitution:

$$\sum_{i<j} \frac{1}{\max(r_i, r_j)} \rightarrow \sum_{i<j} \frac{\alpha}{\max(r_i, r_j)}, \tag{10}$$

where $\alpha = 0.50$ and $\alpha = 0.25$. For half the normal electron–electron interaction, we found a probability on axis of $\mathcal{P}_{1s\rightarrow 2s} = 1.49 \times 10^{-2}$, where $\mathcal{P}_{1s\rightarrow 2s} = 8.5 \times 10^{-3}$ for one wavepacket. For quarter the normal electron–electron interaction, we found a probability on axis of $\mathcal{P}_{1s\rightarrow 2s} = 4.1 \times 10^{-3}$, where $\mathcal{P}_{1s\rightarrow 2s} = 2.1 \times 10^{-3}$ for one wavepacket. Thus, the time-dependent wavepacket calculations are converging towards the 'weak' scattering limit as we reduce the electron–electron interaction in the model hydrogen atom. We note that 'normal' coherent scattering in the three-dimensional Temkin–Poet model for hydrogen is much stronger than that expected in a full nine-dimensional calculation which includes non-zero angular momenta. Thus, a reduced electron–electron interaction in the model is closer to that found in real electron–atom collisions.

To illustrate the destructive interference caused by the scattering of two free electrons on the target, we examine in greater detail the full electron–electron interaction calculations at an incident energy of $E = 50$ eV and a wavepacket separation of $\Delta s = 20.0$. The probability density for the product wavefunction projected onto the 1s orbital, $|\int_0^\infty \mathrm{d}r_0 P_{1s}(r_0)\Phi(r_0, r_1, r_2, t)|^2$, is shown in figure 3 at six different times from $t = 0.0$ to $t = 25.0$. The highest probability density of figure 3(a) is found at $r_1 = 12.0$ and $r_2 = 32.0$, corresponding to the initial positions of the two free-electron wavepackets. The peak of the probability density follows the classical path of $(r_1(t), r_2(t)) = (|12.0 - kt|, |32.0 - kt|)$, where $k = 1.92$ for an incident energy of $E = 50.0$ eV. Thus, at $t = 6.25$ the density peak is at (0.0, 20.0), while at $t = 16.7$ the density peak is at (20.0, 0.0). The classical path is a 'two cushion shot' in billiards, which can be seen in the progression from frames (a) to (f) in figure 3. The ripples in the probability density at times near $t = 6.25$ or $t = 16.7$ are due to wave interference between the incoming and outgoing components of a single wavepacket as it reflects at the origin. The probability density for the product wavefunction projected onto the 2s orbital, $|\int_0^\infty \mathrm{d}r_0 P_{2s}(r_0)\Phi(r_0, r_1, r_2, t)|^2$, is shown in figure 4 at six different times from $t = 0.0$ to $t = 25.0$. There is no probability density in figure 4(a) since the initial target electron is in the 1s orbital. The maximum probability densities are found in figure 4(c) at $t = 10.0$ and figure 4(d) at $t = 15.0$ following the arrival of the first free electron, but before the arrival of the second free electron. In figure 4(f) at $t = 25.0$ the probability density is reduced following the arrival of the second free electron due to destructive interference between the wavepackets. Furthermore, the featureless ball of probability density found in figures 4(c) and (d) has been replaced by a double lobe in figure 4(f) with a clear interference minimum through its centre.

In summary, the excitation of a model hydrogen atom by a coherent electron beam is studied by direct solution of the time-dependent Schrödinger equation on a three-dimensional lattice. For two incident electron wavepackets, the 1s $\rightarrow$ 2s excitation probability is found to oscillate about an axis which is between one and two times the excitation probability for

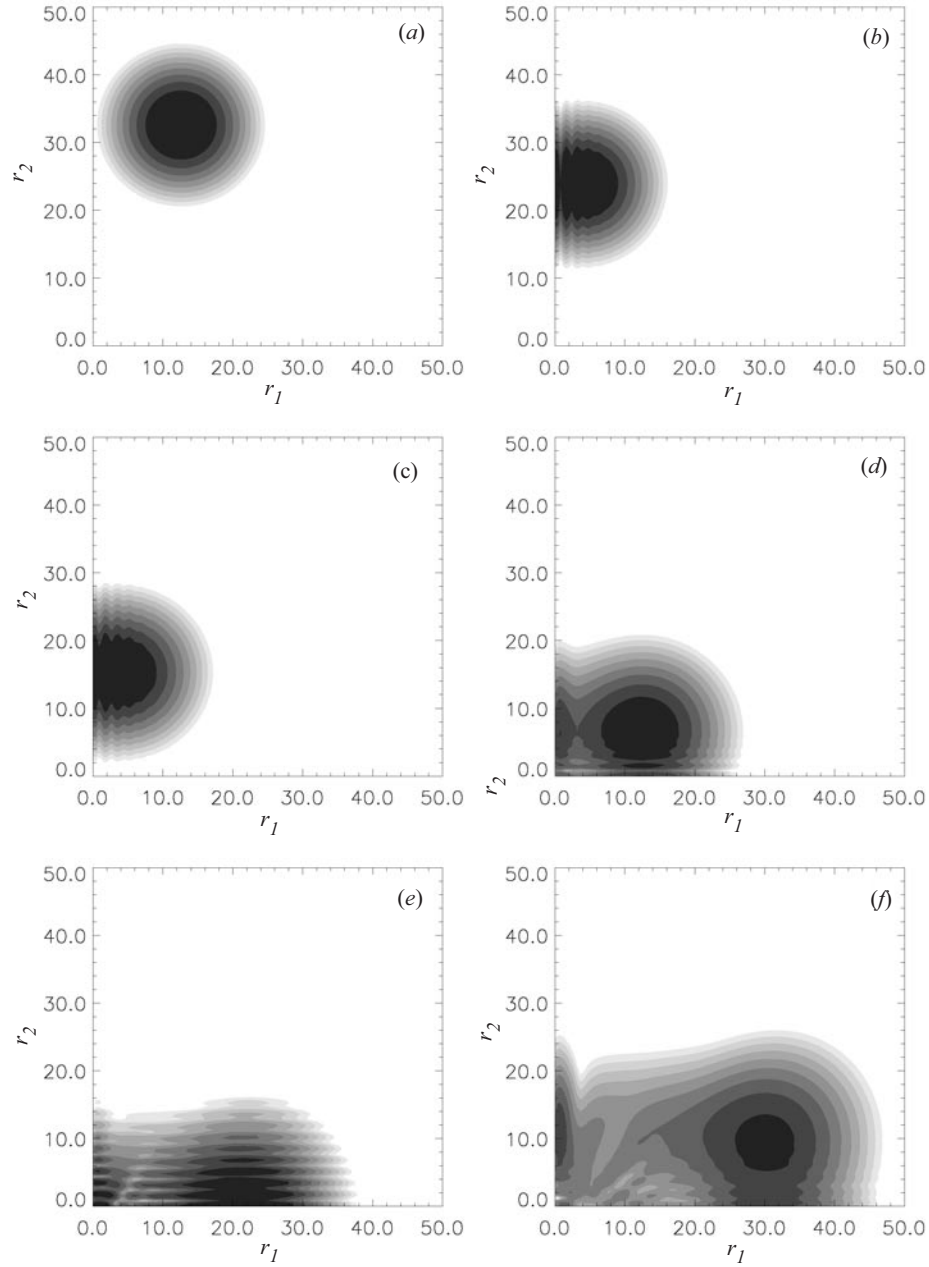

Figure 3. Scattering by two free-electron wavepackets at an incident energy of 50 eV. Probability density, $|\int_0^\infty \mathrm{d}r_0 P_{1s}(r_0)\Phi(r_0, r_1, r_2, t)|^2$, at (*a*) $t = 0.0$, (*b*) $t = 5.0$, (*c*) $t = 10.0$, (*d*) $t = 15.0$, (*e*) $t = 20.0$ and (*f*) $t = 25.0$ (r_1 and r_2 are radial distances in atomic units).

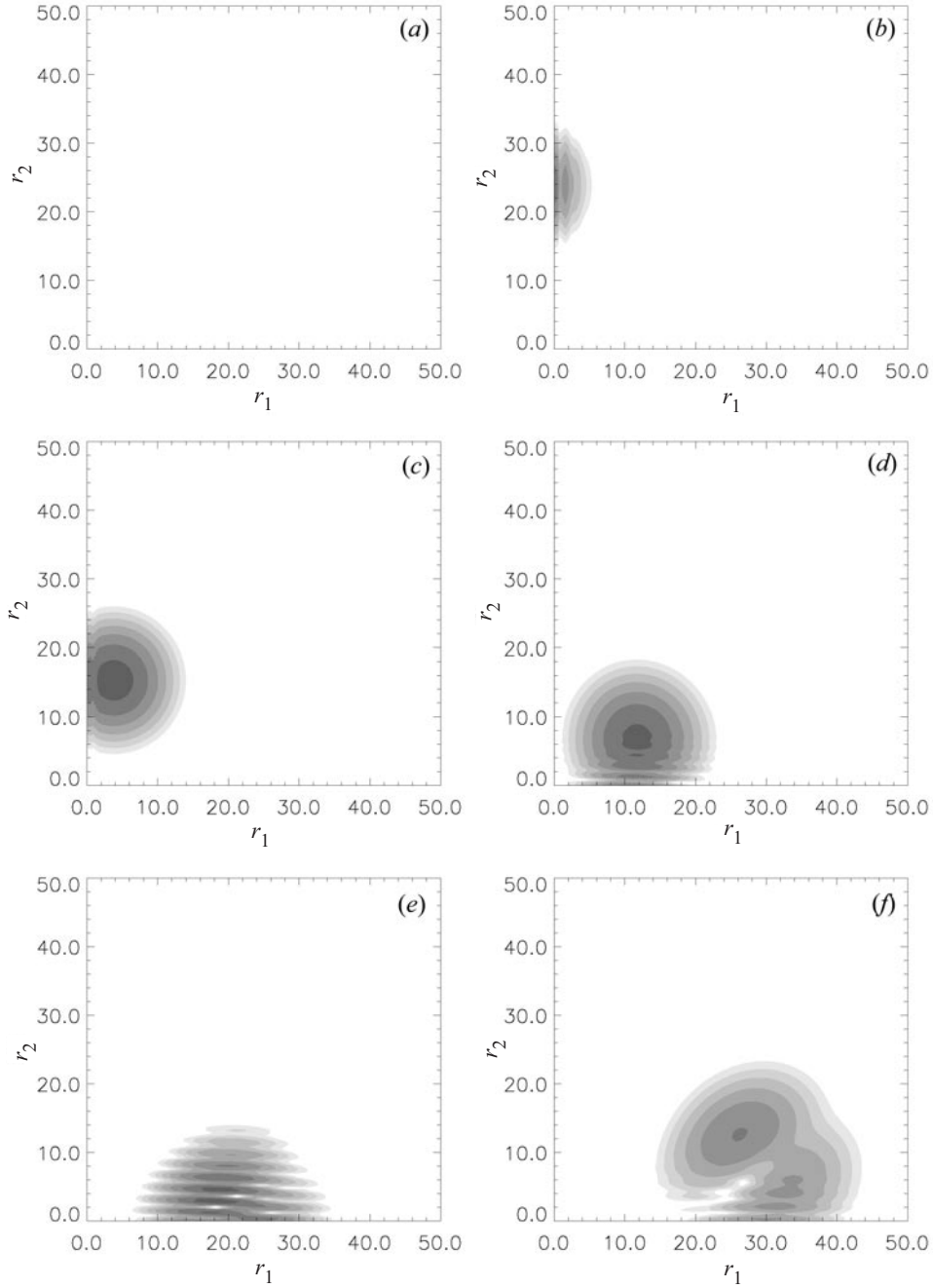

Figure 4. Scattering by two free-electron wavepackets at an incident energy of 50 eV. Probability density, $|\int_0^\infty \mathrm{d}r_0 P_{2s}(r_0)\Phi(r_0, r_1, r_2, t)|^2$, at (*a*) $t = 0.0$, (*b*) $t = 5.0$, (*c*) $t = 10.0$, (*d*) $t = 15.0$, (*e*) $t = 20.0$ and (*f*) $t = 25.0$ (r_1 and r_2 are radial distances in atomic units).

one incident electron wavepacket. When the electron–electron interaction is reduced in the model Hamiltonian, the two wavepacket excitation probability is found to oscillate about an axis which is about two times the excitation probability for one wavepacket, in agreement with 'weak' scattering theory [1]. In addition, the frequency of the 1s $\rightarrow$ 2s excitation probability as a function of wavepacket arrival time is found to precisely equal the 1s $\rightarrow$ 2s transition frequency. In conclusion, we find time-dependent scattering theory to be a clear and powerful tool for studying coherence effects in electron–atom collisions. In future, the development of coherent matter beams may lead to experimental observation of these same interference effects in various atomic collision cross sections.

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References

[1] Robicheaux F and Noordam L D 2000 *Phys. Rev. Lett.* **84** 3735–9
[2] Mewes M O *et al* 1997 *Phys. Rev. Lett.* **78** 582–5
[3] Bloch I, Hansch T W and Esslinger T 1999 *Phys. Rev. Lett.* **82** 3008–11
[4] Robicheaux F, Lankhuijzen G M and Noordam L D 1998 *J. Opt. Soc. Am.* B **15** 1–5
[5] Rella C W *et al* 1999 *J. Opt. Soc. Am.* B **16** 182–7
[6] Temkin A 1962 *Phys. Rev.* **126** 130–42
[7] Poet R 1978 *J. Phys. B: At. Mol. Phys.* **11** 3081–94
[8] Callaway J and Oza D H 1984 *Phys. Rev.* A **29** 2416–20
[9] Bray I and Stelbovics A T 1992 *Phys. Rev. Lett.* **69** 53–6
[10] Watanabe S, Hosoda Y and Kato D 1993 *J. Phys. B: At. Mol. Opt. Phys.* **26** L495–501
[11] Ihra W, Draeger M, Handke G and Friedrich H 1995 *Phys. Rev.* A **52** 3752–62
[12] Bartschat K and Bray I 1996 *Phys. Rev.* A **54** R1002–5
[13] Baertschy M, Rescigno T N, Isaacs W A and McCurdy C W 1999 *Phys. Rev.* A **60** R13–16
[14] Jones S and Stelbovics A T 2000 *Phys. Rev. Lett.* **84** 1878–81
[15] Pindzola M S, Mitnik D and Robicheaux F 1999 *Phys. Rev.* A **59** 4390–8