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Trajectory-interference effects in ion-atom collisions

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Eikonal expressions for differential scattering amplitudes for electronic s-$p_{\pm 1}$ transitions in heavy-particle collisions are derived. It is shown that differential orientation effects follow from an interplay between phases related to the heavy-particle dynamics and the Coulomb perturbation of the initial and final states. At intermediate energies where semiclassical trajectory calculations give preferred orientation for both capture and excitation we show that in the resulting differential scattering cross sections the orientation tends to vanish for excitation, whereas, it prevails for capture in doubly- and multiply-charged-ion–atom collisions.

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In recent years it has become possible to study the most basic quantities of electronic transitions in atomic collisions by coincidence measurements, also in combination with laser-prepared initial states [1]. The characterization of initial and final states is often given in terms of orientation and alignment parameters, i.e., parameters describing the dynamics and the shape of the charge-cloud distribution [2]. Within the theoretical description it is assumed that for most projectile energies the internuclear motion can be described by a classical trajectory $\mathbf{R}(t)$, often approximated by a straight line $\mathbf{R}(t)=\mathbf{b}+\mathbf{v}t$. Thus, in a number of recent works [3–5], impact-parameter dependent coherence properties have been studied, e.g., exploring preferred orientation (propensity rules) in excitation and capture.

It is the purpose of the present Rapid Communication to show that the impact-parameter dependent coherence properties of many collision systems cannot be used directly to interpret the outcome of the asymptotic scattering, *even if state-resolved quantum-mechanical differential cross sections compare qualitatively well with the impact-parameter dependent transition probabilities.* To this end we present a theoretical analysis of the scattering amplitudes for oriented s-$p^{\text{nat}}_{\pm 1}$ transitions into a solid angle at $\Omega=(\theta,\phi)$ based on the eikonal method [6]. The p^{nat}_m states here refer to a laboratory-fixed natural frame of reference [7] and (θ,ϕ) are the scattering polar angles in the collision frame, cf. Fig. 1. Calculations based on this theory have shown excellent agreement at small scattering angles with recent H^+-Na$(3p)$ electron transfer experiments [1], notably where a classical folding of the b-dependent probabilities based on potentials fails.

The center-of-mass differential cross sections are obtained in terms of the scattering amplitudes $f_{ji}(\Omega)$ as

$$\frac{d\sigma_j}{d\Omega}=|f_{ji}(\theta,\phi)|^2,\quad f_{ji}(\theta,\phi)=-\frac{\mu}{2\pi}\langle\Phi_j|V_j|\Psi_i^+\rangle \tag{1}$$

in channels with small energy defects compared to the collision energy $\frac{1}{2}\mu v^2$. Here μ is the reduced mass, V_j is the perturbing potential in the final channel, Φ_j is the final state, and Ψ_i^+ is the complete scattering state from an initial state Φ_i. Within the eikonal method [6], f_{ji} can be expressed in terms of the heavy-particle trajectories $\mathbf{R}(b,\Phi,Z)$, and quantizing the states in the collision frame (cf. Fig. 1) we obtain

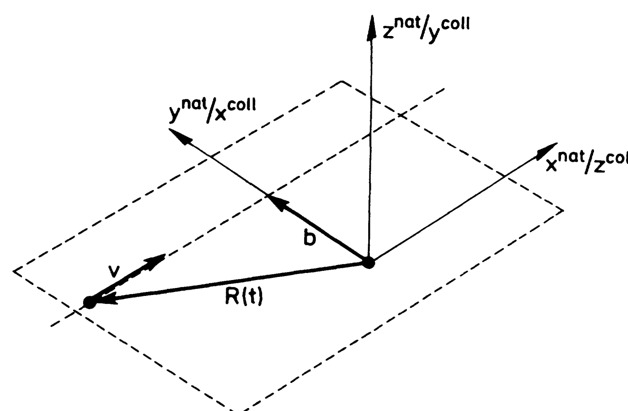

FIG. 1. Planar collision geometry and reference frames used in the scattering description. The "nat" defines the axes of the coordinate system in which $\mathbf{R}=(vt,b,0)$ and the "coll" defines the axes of the coordinate system in which $\mathbf{R}=(b,0,vt)$. A projectile trajectory passing on the left side of the target is shown.

$$f_{ji}(\theta,\phi)=\frac{-ik}{2\pi}\int_0^\infty db\,b\int_0^{2\pi}d\Phi\,e^{-i\eta b\cos(\Phi-\phi)}a_j^{\text{coll}}(b,\Phi)$$
$$=k(-i)^{1+|m_j-m_i|}e^{-i(m_j-m_i)\phi}\int_0^\infty db\,bJ_{|m_j-m_i|}(\eta b)a_j^{\text{coll}}(b)\,. \tag{2}$$

where m_i (m_j) refers to the initial (final) azimuthal quantum number, $k=\mu v$ is the wave number, $\eta=2k\sin(\theta/2)$, and J_n is a Bessel function. In this equation $a_j^{\text{coll}}(b)=a_j^{\text{coll}}(b,\Phi=0)$ is the collision amplitude following from the usual straight-line impact-parameter solution of the collision process in the xz^{coll} plane and modified by the phase of the internuclear core-core interaction. When the final state in question is an oriented state in the natural frame of Fig. 1, it must be expressed by its components in the collision frame because the derivation of Eq. (2) relies on the cylindrical symmetry of the states. Using $p_{\mp}^{\text{nat}}=\mp(1/\sqrt{2})p_0^{\text{coll}}-\frac{1}{2}i(p_-^{\text{coll}}-p_+^{\text{coll}})$, we arrive at the following expression for the scattering amplitudes for s-$p_{\pm}^{\text{nat}}$ transitions,

$$f_{p_{\pm}s}^{\text{nat}}(\theta,\phi)=-ik\int_0^\infty db\,b\left[\mp\frac{J_0(\eta b)}{\sqrt{2}}a_{p_0}^{\text{coll}}(b)-\cos(\phi)J_1(\eta b)a_{p_1}^{\text{coll}}(b)\right]. \tag{3}$$

We easily identify from Eq. (3) the left-right relationship between the scattering amplitudes for the oriented states, $f_{p_{\pm}s}^{\text{nat}}(\theta,\phi)=-f_{p_{\mp}s}^{\text{nat}}(\theta,\pi-\phi)$. The differential orientation parameter, describing the angle-resolved final-state dynamics (sense of electronic rotation), is now derived from the differential cross sections as

$$L_{\perp}(\theta,0)=\frac{|f_{p_+s}^{\text{nat}}|^2-|f_{p_-s}^{\text{nat}}|^2}{|f_{p_+s}^{\text{nat}}|^2+|f_{p_-s}^{\text{nat}}|^2}=-L_{\perp}(\theta,\pi)\,. \tag{4}$$

We note from Eqs. (3) and (4) that for vanishing θ, the first part of the integral dominates due to the behavior of the Bessel functions, so $L_{\perp}(\theta\to 0,0)\to 0$. This is the well-known small-angle scattering limit. In general, and particularly for ion-atom collisions at intermediate collision energies, the outcome of Eq. (4) is nontrivial and determined by the phases and magnitudes of both terms of Eq. (3).

Considering explicitly the core-core interaction [8], the b dependence of the collision amplitudes can be expressed,

$$a_j(b)=|a_j(b)|e^{i[\arg[a_j(b)]+2Z^TZ^P\ln(b)/v]}\,, \tag{5}$$

where Z^P is the projectile-core charge and Z^T is the target-core charge. We note that the contribution of the phase $\arg[a_j(b)]$ may be dominated by the Coulombic part of the perturbing channel potentials as indeed observed in our close-coupling calculations at intermediate and large b, and in that case

$$a_j\approx|a_j|e^{i[2Z^TZ^P-(Z^P+Z)]\ln(b)/v}\,, \tag{6}$$

where the charge $Z=Z^P$ (Z^T) for excitation (capture).

We now introduce the approximation that for $\phi=0$, i.e., left-side scattering in the xz^{coll} plane, the main contribution to the Φ integral of Eq. (2) comes from $\Phi=0$ and $\Phi=\pi$, strictly valid only when $\eta b>1$. Expressing the collision amplitudes of the integrand in terms of the collision amplitudes for the natural-frame states and using $a_{p\pm}^{\text{nat}}(b,\pi)=-a_{p\mp}^{\text{nat}}(b,0)\equiv-a_{p\mp}^{\text{nat}}$, we obtain for the scattering amplitudes for the oriented final $p_{\pm1}^{\text{nat}}$ states

$$f_{p_{\pm1}s}^{\text{nat}}(\theta,0)\sim\int_0^\infty db\,b[|a_{p\pm}^{\text{nat}}|e^{i[(2Z^TZ^P-Z^P-Z)\ln(b)/v-\eta b]}-|a_{p\mp}^{\text{nat}}|e^{i[(2Z^TZ^P-Z^P-Z)\ln(b)/v+\eta b]}]\,. \tag{7}$$

A corresponding, slightly different, expression may be derived from Eq. (3) using the large-argument form of the Bessel functions, i.e., again $\eta b>1$. When $2Z^TZ^P-Z^P-Z>0$ only the first term of the integrand of Eq. (7) (representing left-side trajectories, $\Phi=0$) can become stationary. Thus, when the propensity rule holds we also have $|a_{p-}^{\text{nat}}|\gg|a_{p+}^{\text{nat}}|$ and the stationarity of the first term results in $|f_{p_{-1}s}^{\text{nat}}(\theta,0)|\gg|f_{p_{+1}s}^{\text{nat}}(\theta,0)|$, i.e., the orientation propensity prevails in left-side scattering. However, when $2Z^TZ^P-Z^P-Z=0$ which is the case for excitation processes in ion-atom collision ($Z^T=1$), stationarity is not possible and we predict from Eq. (7) that $|f_{p_{+1}s}^{\text{nat}}(\theta,0)|\approx|f_{p_{-1}s}^{\text{nat}}(\theta,0)|$, i.e., the orientation tends to vanish. We stress that this result, in other words, follows from a coherent interplay between the two terms of Eq. (3) and not as the small-angle limit. Clearly the approximations behind Eq. (7) are not valid as θ approach zero or for small impact parameters. However, we may expect an intermediate region of (θ,b) where this approximate analysis of the eikonal transform can be justified. In the following we investigate these predictions in close-coupling atomic-basis calculations for collision systems where previous coherence studies have been performed within the semiclassical picture [3–5]. For the systems p-H, $\bar{p}$-H, and He^{2+}-H calculations are based on the 14 states spanning the $n=1,2,3$ shells of each of the centers H^+ and He^{2+}.

In Fig. 2(a) we show the excitation probability $P_{2p}(b)$ and $L_{\perp}(b)$ for proton-hydrogen and antiproton-hydrogen collisions for left-side trajectories at $v=1.41$ a.u. (50 keV). Figure 2(b) shows the left-side differential scattering cross sections derived from the quantum-mechanical amplitudes $f_{p_{\pm}s}^{\text{nat}}(\theta,0)$. We first note that the total $2p$ probabilities and $2p$ differential cross sections agree qualitatively: The excitation probability for antiprotons is larger than for protons at smaller impact parameters, giving rise to a larger differential cross section at larger angles. However, the orientation parameters differ drastically in the two figures. In the classical-trajectory picture the orientation parameter for excitation by both projectiles falls rapidly from 0 to -1 as b increases, corresponding to a pure p_{-1}^{nat} state for left-side passages. In the quantum-scattering picture, the orientation parameter for excitation by both projectiles is close to zero in the impor-

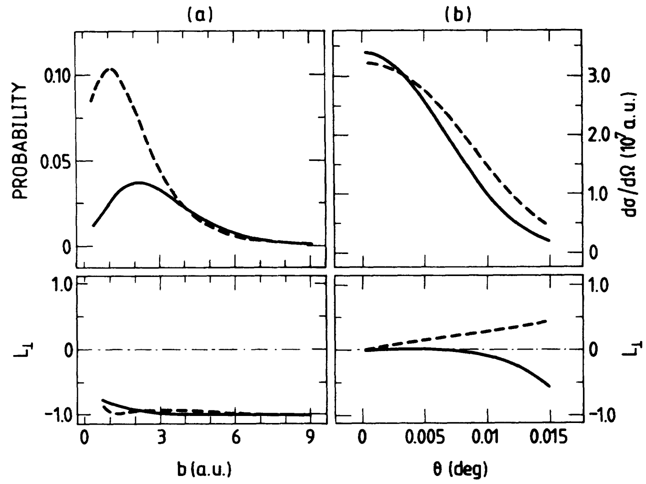

FIG. 2. (a) Probabilities and orientation parameters as functions of impact parameter for $2p$ excitation in proton-hydrogen (solid curve) and antiproton-hydrogen (dashed curve) left-side (cf. Fig. 1) collisions at 50-keV projectile energy. (b) Corresponding $2p$ differential cross sections and differential orientation parameters, Eq. (4), as functions of laboratory scattering angle θ ($\phi=0$), based on Eqs. (1)-(4). The situation corresponds to a detector placed in the xz^{coll} plane on the left side of the target.

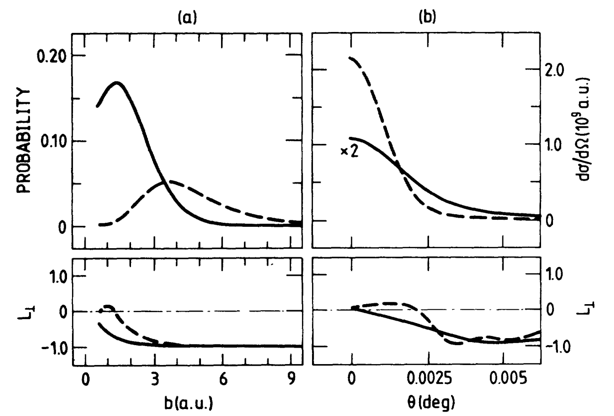

FIG. 3. (a) Probabilities and orientation parameters as functions of impact parameter for $2p$ capture (solid curve) and $2p$ excitation (dashed curve) in He^{2+}-H(1s) collisions at $E=200$ keV ($v=1.41$ a.u.). (b) Corresponding differential cross sections and orientation parameters as function of laboratory scattering angle θ ($\phi=0$).

tant θ range as predicted above since $2Z^TZ^P-Z^P-Z^P=0$ ($Z^T=1, Z^P=\pm 1$). Only at larger angles do we obtain negative $L_\perp(\theta,0)$ values for protons (left-side passage interpretation) and positive $L_\perp(\theta,0)$ values for antiprotons (right-side passage interpretation), in accordance with the expected different effective potentials *at small b,* stemming from the different core-core interactions.

On the other hand, for capture in doubly-charged-ion-atom systems where $2Z^TZ^P-Z^P-Z^T=1$, we expect the orientation propensity to prevail in the differential scattering. This is observed in Fig. 3 where $2p$ capture (solid line) is shown for the $He^{2+}+H(1s) \rightarrow He^+(2p^{\mathrm{nat}}_{\pm 1})+H^+$ channels at the impact velocity $v=1.41$ (a.u.) (200 keV). In this figure we also plot the H(2p) excitation results and indeed we observe that the excitation orientation is close to zero over the same *angle range* where capture orientation is negative and both differential cross sections are large. Note that a typical scattering angle, $\theta=0.002°$, correspond to typical values of ηb around 0.5–2.0. At this angle we obtain $L_\perp=-0.42$ for capture while $L_\perp=+0.14$ for excitation in accordance with the approximate analysis of Eq. (7).

Finally we shall emphasize that the dominating physical mechanisms for singly-charged-ion-atom collisions at lower energies are more difficult to resolve, since strong couplings between various states may induce system-dependent results. For the p-Na(3p) collision system, we have predicted strong differential orientation effects in the collision range 1–5 keV [9] in excellent agreement with parallel experimental investigations [10]. The eikonal method has previously been used in an orientation study of this system by Allan *et al.* [11].

In conclusion we have shown that the predictions of coherence parameters based on trajectory calculations must be performed with care. Interference effects play an important role for the scattering dynamics not only in the limit of vanishing scattering angles, even if correspondence between the quantum and the trajectory pictures can be found in *total* differential quantities.

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[1] J. C. Houver, D. Dowek, C. Richter, and N. Andersen, Phys. Rev. Lett. **68**, 162 (1992).
[2] N. Andersen, J. Gallagher, and I. Hertel, Phys. Rep. **165**, 1 (1988).
[3] W. Fritsch and C. D. Lin, Phys. Rep. **202**, 1 (1991), and Refs. [326-352] therein.
[4] C. D. Lin, R. Shingal, A. Jain, and W. Fritsch, Phys. Rev. A **39**, 4455 (1989).
[5] J. P. Hansen, A. Dubois, and S. E. Nielsen, Phys. Rev. A **44**, 6130 (1991).
[6] R. McCarroll and A. Salin, J. Phys. B **1**, 163 (1968); M. R. Flannery and K. J. McCann, Phys. Rev. A **8**, 2915 (1973).
[7] H. W. Hermann and I. V. Hertel, Comments At. Mol. Phys. **12**, 61 (1982).
[8] R. D. Piacentini and A. Salin, Comput. Phys. Commun. **13**, 57 (1977).
[9] A. Dubois, S. E. Nielsen, and J. P. Hansen, J. Phys. B (to be published).
[10] C. Richter, N. Andersen, J. C. Brenot, D. Dowek, J. C. Houver, J. Salgado, and J. Thomsen, J. Phys. B (to be published).
[11] R. J. Allan, C. Courbin, P. Salas, and P. Wahnon, J. Phys. B **23**, L461 (1990).