Multiple-Scattering Approach to Coherent Excitation in Electron-Capture Collisions

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The complete density matrix for the $n=3$ level of hydrogen coherently excited by electron capture, $H^+ + He \rightarrow H(n=3) + He^+$, at medium to high energies is calculated with the first Born approximation and for the first time with a multiple-scattering theory, namely the continuum distorted-wave approximation. Final-state interactions are also explicitly incorporated to account for the long-range coupling effects between the degenerate states of the projectile. Comparison is made with recent experimental data and a quasiclassical interpretation of coherences in terms of the orbital parameters of the captured electron is proposed.

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Recently, Havener *et al.*[1] have reported the experimental determination of the $n=3$ density matrix formed by electron capture in $H^+ + He \rightarrow H(n=3) + He^+$ at collision energies between 40 and 80 keV. These measurements extend the previous work of Sellin *et al.*[2] on the $n=2$ density matrix to higher n and lower collision velocities with a somewhat different experimental technique. As a most striking result Havener *et al.* have found clear evidence of a large, positive dipole moment of the excited electron, $\langle d_z \rangle > 0$, along the beam ($\hat{z}$) axis in contrast to the predictions of the first Born [Oppenheimer-Brinkman-Kramers (OBK)] approximation.[3] Indeed, the OBK approximation gives vanishing expectation values for all observables odd under parity and time-reversal (PT) transformation. Therefore, the observation of a nonvanishing dipole moment clearly indicates the failure of the first Born approximation and points to the necessity of including multiple-scattering contributions in a calculation of the density matrix. Investigation of coherent excitation can thus reveal detailed information on the influence of higher-order perturbations and provides, as we shall see, a sensitive test for various scattering models.

The Born series for charge-exchange possesses the remarkable feature that the leading contribution to the cross section is provided at asymptotically high velocity by the second-order term[4] (double scattering). For finite velocity, however, both first and second Born approximations[5] fail to reproduce the experimental capture cross sections. In order to remedy this situation, various approximation schemes[6] have been devised during recent years to accommodate higher-order (multiple-scattering)[7] terms of the Born series.

We have calculated the axially symmetric $n=3$ density matrix without detection of the scattering angle of the outgoing projectile,

$$\sigma_{nlm,nl'm'} = \frac{\delta_{m,m'}}{(2\pi)^2 v} \int d^3k \, t_{nlm}(\vec{k}) t^*_{nl'm'}(\vec{k}) \delta\left(\vec{k}\cdot\vec{v} + \frac{v^2}{2} + \Delta\epsilon\right), \tag{1}$$

with four different approximations as discussed below. In Eq. (1) $t_{nlm}(\vec{k})$ denotes the transition matrix element[7] from a $1s$ initial target state to a hydrogenic final projectile state (nlm) as a function of the momentum transfer $\vec{k}$. The difference in the binding energies is denoted by $\Delta\epsilon = \epsilon_f - \epsilon_i$. For the $n=3$ level the axial symmetry allows altogether fourteen independent nonvanishing parameters: six diagonal elements corresponding to the usual substate cross sections and four (complex) off-diagonal elements describing coherences between different substates. These off-diagonal elements provide detailed information on the relative phases of the scattering amplitudes not accessible in conventional determinations of the cross sections.

In this Letter we confine ourselves to a systematic study of the expectation values of the z component of the Runge-Lenz vector $\vec{A}$ and of $\vec{L}\times\vec{A}$, i.e., $\langle A_z \rangle$ and $\langle (\vec{L}\times\vec{A})_z \rangle$, where $\vec{A}$ is given (in a.u.) by

$$\vec{A} = (n/Z_p)[\tfrac{1}{2}(\vec{p}\times\vec{L} - \vec{L}\times\vec{p}) - Z_p \vec{r}/r], \tag{2}$$

with $\vec{p}$ and $\vec{L}$ being the linear and angular momenta, respectively, and Z_p the charge of the projectile. These two multipoles[8] of rank 1 can be expressed in terms of the density matrix elements as

$$\langle A_z\rangle = (4/\mathrm{Tr}_{n-3}\sigma)\mathrm{Re}(\tfrac{2}{3}^{1/2}\sigma_{s,p_0}+\tfrac{1}{3}\sqrt{3}\sigma_{p_0,d_0}+\sigma_{p_1,d_1}), \quad (3)$$

$$\langle(\vec{L}\times\vec{A})_z\rangle = (-4/\mathrm{Tr}_{n-3}\sigma)\mathrm{Im}(\tfrac{2}{3}^{1/2}\sigma_{s,p_0}+\tfrac{2}{3}\sqrt{3}\sigma_{p_0,d_0}+\sqrt{2}\sigma_{p_1,d_1}). \quad (4)$$

This choice is dictated by the observation that $\vec{L}$ and $\vec{A}$ are the generators of the O(4) symmetry group[9] of the hydrogen atom. In a classical picture, $\vec{A}$ points from the projectile nucleus to the "perihelion" of the electronic orbit with magnitude proportional to the eccentricity, whereas $\vec{L}\times\vec{A}$ points along the orbital velocity at the perihelion. Moreover the matrix elements of the dipole operator $\vec{d}$ are related[9] to $\vec{A}$ for each n subspace by

$$\vec{d} = -\vec{r} = \tfrac{3}{2}n\vec{A}. \quad (5)$$

We have calculated $\langle A_z\rangle$ and $\langle(\vec{L}\times\vec{A})_z\rangle$ with four different approximations of increasing complexity and accuracy: (a) As a standard for comparison, we first evaluate the OBK approximation containing only single-scattering contribution. (b) An improvement of the OBK density matrix can be achieved by including the final-state mixing of the nearly degenerate hydrogenic levels due to the Coulomb field of the residual target ion. Previous studies[3,10] indeed indicate that coherences in degenerate manifolds are extremely sensitive to final-state interactions and that low-order perturbation theories including only impulsive short-range interactions are inadequate to describe the coherence effects. A simplified analytical treatment, called the post-collision interaction (PCI)[3] model is possible in which the evolution of the density matrix under the influence of the linear Stark effect,

$$U_{\mathrm{PCI}} = \exp[(i/v)\int_{R_0}^{\infty} dR\ \vec{d}\cdot\vec{F}(R)], \quad (6)$$

is determined after the projectile has left the close collision region where capture is expected to take place (i.e., $R > R_0 = \langle r\rangle_n$). $\vec{F}(R)$ describes the electric field produced by the ionized target in the vicinity of the projectile and becomes essentially parallel to $\vec{v}$ at large internuclear separations such that $\vec{d}\cdot\vec{F} \simeq d_z F$. We will refer to this combined model as the OBK-PCI approximation. (c) To include multiple-scattering contributions to the capture process, we use the continuum distorted-wave[11] (CDW) approximation. Despite some known deficiencies[6] the CDW approximation has major credentials, (i) as it treats distortion of the entrance and exit channel on equal footing and therefore is well suited for nearly symmetric collision systems, (ii) as it gives reasonable agreement with experimental data for integrated cross sections, and finally (iii) since the capture amplitude is available in analytic form for arbitrary final states.[7] (d) In analogy to the OBK-PCI model, we have also investigated the influence of the long-range Stark mixing upon the CDW density matrix. The resulting CDW-PCI model then incorporates both multiple-scattering contributions during the close collision and the Stark mixing of coherently excited projectile states in the outgoing channel.

We approximate the He($1s$) state by a hydrogenic orbital with an effective charge $Z_T = 1.35$ chosen to reproduce the experimental binding energy. We have found a weak dependence of $\langle A_z\rangle$ and $\langle(\vec{L}\times\vec{A})_z\rangle$ upon variations of the target wave function. Comparison with preliminary results of a calculation using a Hartree-Fock He ground-state wave function shows deviations of about 10% above the matching velocity $v = 1.35$.

Comparison of the various scattering models with experimental data for $\langle A_z\rangle$ [Fig. 1(a)] and

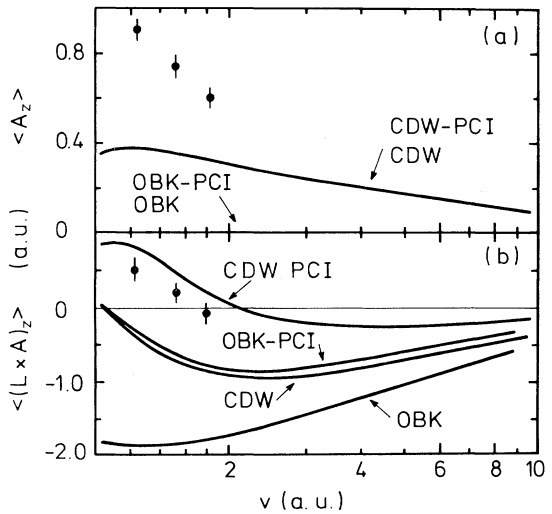

FIG. 1. (a) Expectation value of the z component of the Runge-Lenz vector $\langle A_z\rangle$ following charge transfer $p + \mathrm{He} \rightarrow \mathrm{H}(n=3) + \mathrm{He}^+$ as a function of projectile velocity. Dots with error bars, experimental data of Havener *et al.* (Ref. 1); solid line, CDW and CDW-PCI. (b) Expectation value of the z component $\langle(\vec{L}\times\vec{A})_z\rangle$ as a function of the projectile velocity in different approximations.

$\langle(\vec{L}\times\vec{A})_z\rangle$ [Fig. 1(b)] clearly exhibits the extreme sensitivity of the coherence parameters upon the details of the underlying approximation. As noted above, in the Born approximation, $\langle A_z\rangle$ vanishes for all energies. This remains true if Stark mixing (OBK-PCI) is included since both the OBK and the evolution operator $U_{\rm PCI}$ preserve PT symmetry.[12] In these approximations, all PT-odd variables are identically zero. The CDW method yields a nonvanishing $\langle A_z\rangle$ (or dipole moment). The direction of displacement of the electron as well as the energy dependence agrees reasonably well with the data. The captured electron lags behind the projectile, i.e., $\langle A_z\rangle > 0$. The absolute magnitude, however, is too small by a factor of 3 at 40 keV and a factor 1.8 at 80 keV. The trend of the experimental findings suggests that the agreement should improve at higher velocities. It is no surprise that only a qualitative agreement is obtained since the CDW approximation is intrinsically a high-energy approximation. Extrapolation down to the matching velocity ($v=1.35$) should therefore be considered with caution. One notes also that CDW and CDW-PCI models give identical results for $\langle A_z\rangle$ as a consequence of the fact that $U_{\rm PCI}$ commutes with A_z [Eqs. (5) and (6)]. One sees therefore that a finite value of $\langle A_z\rangle$ is a direct measure of the presence of second- and higher-order terms in the perturbation expansion of the capture amplitude. The quantity $\langle A_z\rangle$ is, on the other hand, insensitive to the PCI effects.

A different situation occurs for $\langle(\vec{L}\times\vec{A})_z\rangle$ an observable even under PT transformation. All four approximations give nonzero results which are sensitive to *both* multiple-scattering contributions and PCI effects. The OBK-PCI reduces the OBK value to such an extent that it comes close to the CDW result. The PCI correction to the CDW approximation has a dramatic effect upon $\langle(\vec{L}\times\vec{A})_z\rangle$ which gives rise to a change of sign at velocities $v\simeq 2$ a.u. The CDW-PCI model reproduces quite well the experimental features which show a zero near $v\simeq 1.6$ a.u. The comparison indicates the superiority of the CDW-PCI calculation and points to the necessity of incorporating PCI effects into a perturbation calculation of coherent excitation by electron capture.

The expectation values, $\langle A_z\rangle$ and $\langle(\vec{L}\times\vec{A})_z\rangle$, may now be used to extract a simplified "classical" but instructive orbital picture of the coherently excited manifold. Let us consider for a moment an angle-differential scattering process with detection of the scattering angle of the outgoing projectile in the x-z collision plane. By reflection symmetry, only two components of each vector, $\langle A_x\rangle, \langle A_z\rangle$ and $\langle(\vec{L}\times\vec{A})_x\rangle, \langle(\vec{L}\times\vec{A})_z\rangle$, can have nonvanishing expectation values. This corresponds, in the classical picture, to a Kepler ellipse in the x-z plane with an angular momentum pointing in either the $+y$ or the $-y$ direction depending on the rotational sense (Fig. 2). Classically, three out of five parameters suffice to determine completely the tilt of the major axis and the rotational sense. In an angle-integral experiment all transverse components vanish because of rotational symmetry leaving a "classical" orbit incompletely defined. We can nevertheless attempt to interpret the experimental data in terms of a classical orbit: We use the integral density matrix to describe $\langle A_z\rangle$ and $\langle(\vec{L}\times\vec{A})_z\rangle$ and the corresponding *differential* density matrix [proportional to the integrand in Eq. (1)] to obtain information on the expectation values of the transverse components.

The picture that emerges in the CDW-PCI model is the following. We find $\langle A_x\rangle$ to be negative at intermediate energies which suggests a Runge-Lenz vector tilted towards the beam axis [Fig. 2(a)]. The change of sign in $\langle(\vec{L}\times\vec{A})_z\rangle$, however, implies a change of sign of the angular momentum $\langle L_y\rangle$ as a function of the energy. One expects, therefore, a counterclockwise rotation for lower velocities and a clockwise rotation at higher velocities in accordance with the classical density-gradient model.[13] Without the use of knowledge from the differential density matrix, an alternative interpretation can be

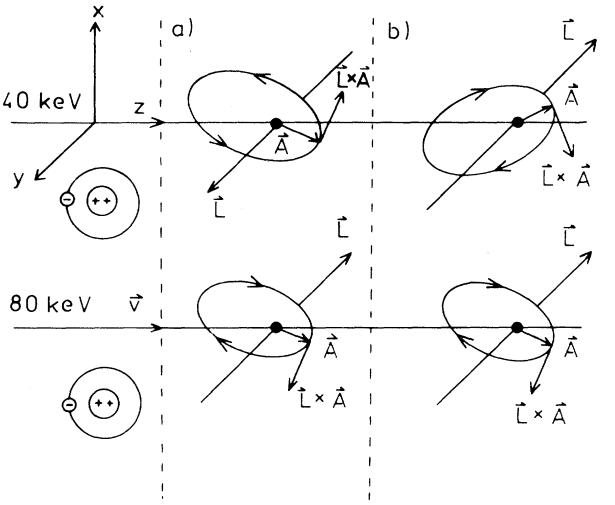

FIG. 2. "Classical" orbits of the coherently excited electron (see text); (a) model suggested by the CDW-PCI approximation; (b) possible alternative also consistent with the data of Ref. 1. All orbits lie in the x-z plane.

formulated where the sense of rotation remains fixed while the Runge-Lenz vector rotates [Fig. 2(b)]. Without further experimental and theoretical analysis, one cannot at present distinguish between these two (consistent) possibilities. It should be noted that even in the case of differential scattering the correspondence of the expectation values of $\vec{A}$, $\vec{L}\times\vec{A}$, and $\vec{L}$ to those of a single classical ellipse is, in general, not unique[12] because the quantum statistical expectation values are independent from each other in contrast to the classical case.

In conclusion, we have isolated two coherent multipoles, $\langle A_z\rangle$ and $\langle(\vec{L}\times\vec{A})_z\rangle$, which provide a sensitive measure of multiple-scattering contributions to the charge-exchange process. Moreover, the interpretation of coherent excitation in terms of a classical orbital picture of the captured electron sheds new light on an until-now poorly understood phenomenon. A more detailed study of additional multipoles of the complete density matrix is in progress. Future investigations should focus also on an improvement of the primary capture amplitude, e.g., by adapting a strong-potential Born approximation[6] to nearly symmetric systems.

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[1]C. Havener, W. Westerveld, J. Risley, N. Tolk, and J. C. Tully, Phys. Rev. Lett. **48**, 926 (1982); C. Havener, Ph.D. thesis, North Carolina State University, 1983 (unpublished).

[2]I. Sellin, L. Liljeby, S. Mannervik, and S. Hultberg, Phys. Rev. Lett. **42**, 570 (1979).

[3]J. Burgdörfer, Phys. Rev. A **24**, 1756 (1981).

[4]R. Shakeshaft and L. Spruch, Rev. Mod. Phys. **51**, 369 (1979).

[5]P. R. Simony and J. H. McGuire, J. Phys. B **14**, L737 (1981).

[6]J. Briggs, K. Taulbjerg, and J. Macek, Comments At. Mol. Phys. **12**, 1 (1982), and references therein.

[7]L. J. Dubé, J. Phys. B **17**, 641 (1984).

[8]J. Burgdörfer, Z. Phys. A **309**, 285 (1983).

[9]M. Englefield, *Group Theory and the Coulomb Problem* (Wiley-Interscience, New York, 1972).

[10]Y. Band, Phys. Rev. A **19**, 1906 (1979).

[11]I. M. Cheshire, Proc. Phys. Soc. London **84**, 89 (1964).

[12]L. J. Dubé and J. Burgdörfer, to be published.

[13]H. Schröder and E. Kupfer, Z. Phys. A **279**, 13 (1976).