Quantum-Mechanical Phase Jumps in Collision-Induced Coherent Excitation

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(Received 13 February 1979)

We present experimental and theoretical excitation probabilities $\mathcal{P}$ and coherence parameters λ and χ for $Be^+(2s \rightarrow 2p)$ excitation in 2–15-keV Be^+-Ne collisions with impact parameters near 1.6 a.u. The phase difference χ between the quantum-mechanical amplitudes for $M_L = 0$ and $M_L = 1$ substate excitation exhibits a rapid change near 4 keV, where λ, which measures the relative size of those amplitudes, approaches its maximum value of unity. Impact-parameter calculations of $\mathcal{P}$, λ, and χ predict the trends found experimentally.

Measurements of coherence parameters for excited states formed in atomic collisions provide very detailed tests of collision theories.[1] Efforts have concentrated on p states, where the results are normally reported in terms of λ, the excitation probability of the $M_L = 0$ substate relative to the total probability $\mathcal{P}$, and χ, the phase angle between the $M_L = 0$ and $M_L = 1$ substate excitation amplitudes. Combined with the total excitation probability $\mathcal{P}$, λ, and χ determine completely the complex quantum-mechanical excitation amplitudes, apart from an arbitrary phase factor.

In recent experimental and theoretical works,[2-4] we have obtained detailed information about the excitation mechanisms responsible for projectile resonance-line emission and polarization for quasi-one-electron systems, e.g., alkali-atom–rare-gas collisions. These systems are often of a very clean nature, with the rare-gas atom acting as a spinless and excitationless particle. Currently, we are extending our studies to include coherence parameters and have selected the Be^+-Ne system as a suitable test case. This Letter reports (i) experimental excitation probabilities $\mathcal{P}$ together with λ and χ parameters for $Be^+(2s \rightarrow 2p)$ excitation for 2–15-keV impact energy at a fixed, reduced scattering angle of 4 keV deg, corresponding to an impact parameter[4] near 1.6 a.u., and (ii) theoretical results of a three-state impact-parameter calculation displaying for the first time detailed $(\mathcal{P}, \lambda, \chi)$ values for an ion-atom collision. In a narrow region around 4 keV, the phase χ shows a 180° change associated with a very small value of the $M_L = 1$ amplitude. The main experimental trends are reproduced by theory.

The coherence properties of the p state excited in the collision and expressed through the λ and χ parameters manifest themselves through the polarization of the light emitted in the decay of the excited state subsequent to the collision.[5-6] Using the polarized-photon, scattered-ion coincidence technique,[5] we have measured the relative Stokes vector $\vec{P} = (P_1, P_2, P_3)$ for the light emitted perpendicular to the scattering plane. Details of the experimental arrangement have been given elsewhere.[7] A minor change in the apparatus allowed a 90° rotation of the collision plane around the beam axis so that the Stokes vector in the collision plane also could be measured. $\vec{P}$ is defined by $P_1 = [I(0°) - I(90°)]/I$, $P_2 = [I(45°) - (135°)]/I$, and $P_3 = [I(\mathrm{RHC}) - I(\mathrm{LHC})]/I$, where I is the total light intensity, RHC and LHC denote right- and left-handed circular polarization, and $I(\alpha)$ is the linearly polarized intensity component measured at an angle α to the beam direction. The connection between the measured $\vec{P}$ and the parameters λ and χ is discussed below where it is shown how depolarization due to internal forces and cascades is taken into account.

In a fully coherent excitation of a p state in a fast collision, for which spin-dependent forces can be neglected, the orbital part of the final state can be represented by the ket (in $|LM_L\rangle$ notation)

$$|\psi\rangle = a_0|10\rangle + a_1|11\rangle + a_{-1}|1,-1\rangle,$$

where $a_1 = -a_{-1}$ because of reflection symmetry in the collision plane. For this case, $\mathcal{P} = |a_0|^2 + 2|a_1|^2$, $\lambda = |a_0|^2/\mathcal{P}$, and χ is defined by $a_0 = |a_0|$

and $a_1 = |a_1|e^{i\chi}$. Then[6]

$$\vec{P} = \left(2\lambda - 1,\ -2[\lambda(1-\lambda)]^{1/2}\cos\chi,\ 2[\lambda(1-\lambda)]^{1/2}\sin\chi\right). \tag{1}$$

The degree of polarization $|\vec{P}|$ is unity in this case.

In the present experiment we excite the $2\,^2P$ term of $^9\mathrm{Be}^+(I=\frac{3}{2})$. The fine and hyperfine structure (FS and HFS), which develops after the collision, introduces a mixing of the $|LM_L\rangle$ states, which results in a decrease of the degree of polarization observed. However, HFS can be neglected in this case since it is small compared to the natural linewidth.[3] FS is taken into account by replacing $\vec{P}$ by $\vec{P}' = (\frac{7}{3}P_1, \frac{7}{3}P_2, P_3)$ in Eq. (1).[8] The FS-corrected degree of polarization $|\vec{P}'|$ is unity.

Since energy-loss analysis of the scattered particles was not performed, a further complication is caused by decays from ^2P levels populated through cascades.[4] These cascades introduce a further reduction in the observed degree of polarization. While cascade contributions from ^2S levels are unpolarized, this is not necessarily the case for cascades from ^2D levels. Nevertheless, in the following we assume and test experimentally that the total effect of cascades is a virtually unpolarized background of intensity I_B, added to the intensity I_D from directly excited ^2P levels. Under this assumption, it can be shown that (i) $|\vec{P}'| = I_D/(I_D+I_B)$ and (ii) Eq. (1) is valid with $\vec{P}$ replaced by the unit vector $\vec{P}'/|\vec{P}'|$,

$$\vec{P}'/|\vec{P}'| = \left(2\lambda - 1,\ -2[\lambda(1-\lambda)]^{1/2}\cos\chi,\ 2[\lambda(1-\lambda)]^{1/2}\sin\chi\right). \tag{2}$$

Simultaneous neon excitation would blur the picture even further, but these channels are not significantly populated.[4]

The 90° rotation of the collision plane allows determination of the Stokes vector (P_4, P_5, P_6) of the light emitted in the collision plane, perpendicular to the beam. If the light were fully coherent, this vector would be $(1,0,0)$. The FS effect yields a Stokes vector $(\frac{7}{3}P_4, 0, 0) = [7\lambda/(4+3\lambda), 0, 0]$. The cascade effect modifies this vector further so that approximately $\frac{7}{3}P_4 = |\vec{P}'|\,[7\lambda/(4+3\lambda)]$.

Figure 1 shows measured values of P_1, P_2, P_3, and P_4. The degree of polarization $|\vec{P}'|$ varies from 1.0 at 2 keV to 0.5 at 15 keV with a minimum of 0.3 near 4 keV. Calculated values of P_4 based on the assumption of unpolarized cascade contribution are indicated by crosses. The good agreement with the measured P_4 values supports the basic assumption.

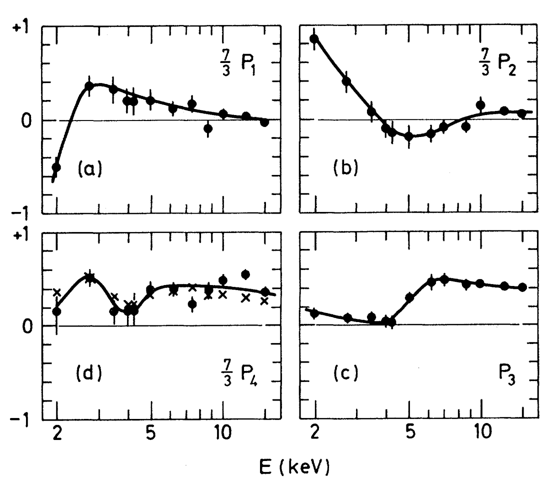

FIG. 1. Measured polarizations P_1, P_2, P_3, and P_4 vs impact energy for fixed impact parameter $b \simeq 1.6$ a.u.

Figures 2(a)–2(c) show the energy dependence of experimentally determined excitation probabilities $\mathcal{P}$ normalized to previous energy-loss measurements[4] and corresponding values of λ and χ obtained using Eq. (2). We notice that λ is close to unity near the 4-keV minimum in $\mathcal{P}$. The phase angle χ decreases from a value of about 90° at high energy to a value near 0° at 4 keV. There is here a rapid change of χ of about 180°, but, within the error bars, it cannot be distinguished whether the change is +180° or −180°, or whether the jump is discontinuous or just very steep [curves labeled 1–3 in Fig. 2(c)]. The reason for the jump becomes clear if one translates the λ and χ parameters to the amplitudes a_0 and a_1. Figure 3 shows the energy dependence of the complex number a_1, measured in units of a_0 for clarity. The phase χ of a_1 changes rapidly near 4 keV, where a_1 passes very close to the origin. The phase jumps, curves 1–3 in Fig. 2(c), correspond to the paths labeled 1–3 in the inset of Fig. 3. From the experiment it is not possible to judge whether a_1 passes just to the right of (1), or to the left of (2), or straight through (3) the origin. Nevertheless, Fig. 3 shows clearly why χ changes rapidly by about 180° in regions where λ is close to unity ($a_1 \simeq 0$).

In recent theoretical work,[2] we have obtained interpretations of experimental emission cross

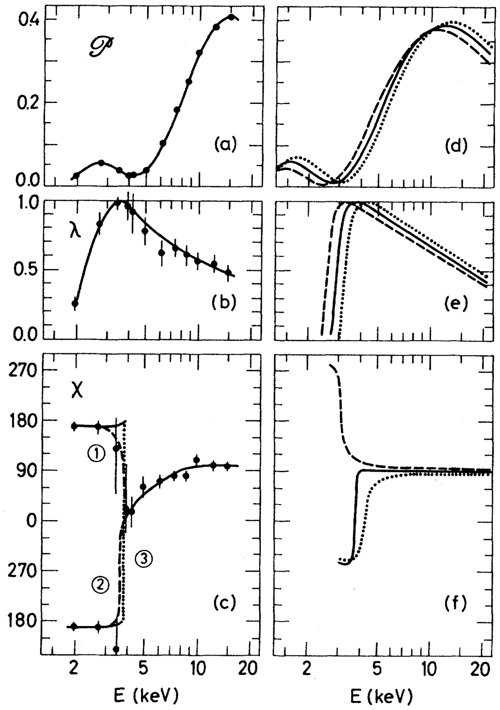

FIG. 2. (a)–(c) Experimental emission probability $\mathcal{P}$ and coherence parameters λ and χ. (d)–(f) Theoretical $\mathcal{P}$, λ, and χ curves for $2s \rightarrow 2p$ excitation evaluated at $b = 1.5$ a.u. (dotted line), 1.6 a.u. (solid line), and 1.7 a.u. (dashed line) ($\mathcal{P}$ has been multiplied by 0.4).

sections and polarizations of impact radiation for a series of quasi-one-electron systems, e.g., (Be^+, Mg^+)-(He, Ne). We have solved for the probability amplitudes of a one-center atomic expansion the rectilinear impact-parameter equations in close coupling, based upon the direct electrostatic interaction V_{HF} of the projectile valence electron and the Hartree-Fock, frozen-core ground-state charge distribution of the target. For the Be^+-Ne system we have performed a three-state $(2s, 2p_0, 2p_{\pm 1})$ model calculation with the electron-neon potential V_{HF} scaled to reproduce the proper magnitude of the maximum of the $2s$-$2p$ total cross section ($\simeq 4.2$ Å^2 at $E_{lab} \approx 12-13$ keV). Scaling of the simple coulombic potential V_{HF} for the e-Ne interaction is a necessary modification to account for the neglect of the repulsive Pauli exclusion forces.[9] The scaling chosen ($V_{HF} \rightarrow \frac{1}{3} V_{HF}$) also leads to cross-section predictions that are within a factor of 2 of the experimental values[3] over the entire energy range $1.5 < E_{lab} < 75$ keV and predicts the characteristic local minimum in polarization at $E_{lab} \simeq 3$ keV (Ref. 3) previously discussed for other

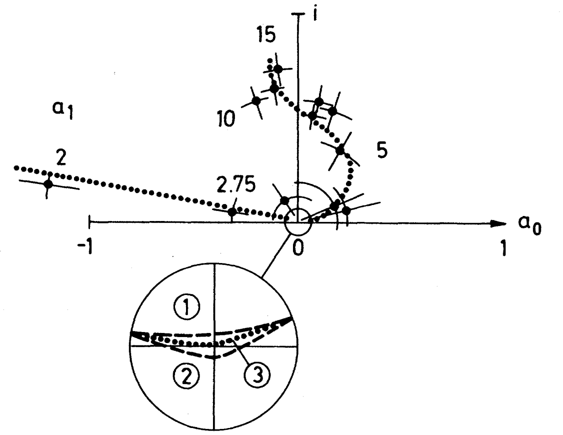

FIG. 3. The complex amplitude a_1 plotted in units of a_0. The numbers next to the experimental points indicate the projectile energy in keV. The inset shows possible passages of the origin, corresponding to the curves in Fig. 2(c).

systems.[2]

The corresponding theoretical results for $\mathcal{P}$, λ, and χ are shown in Figs. 2(d)–2(f) for impact parameters $b = 1.5$, 1.6, and 1.7 a.u. Even though the absolute values of $\mathcal{P}$ are too large, the energy variation of $\mathcal{P}$ compares quite well to that of the experimental result, Fig. 2(a). Also, theory predicts the energy dependence of λ with a maximum close to unity near 4 keV and, associated with that, a rapid change of the phase angle χ by about 180°. The three theoretical curves for $\chi(b, E)$ display changes that correspond closely to the three passage possibilities shown in Fig. 3, although details of the theoretical results differ from the experimental curves. We notice in particular that the predictions of decaying λ, and χ converging towards 90° in the high-energy limit are confirmed experimentally. These results are general asymptotic predictions of the close-coupling calculation and not related to the constant $\chi = 90°$ result of the particular molecular curve-crossing model previously reported by Jaecks *et al.*[5] For lower energies, theory displays a minimum value of λ close to zero and, associated with it, one more phase jump for χ of about 180° [not shown in Figs. 2(e) and 2(f) for clarity].

The extremum structure of λ may be interpreted theoretically in terms of a Massey criterion,[9] causing an out-of-phase cancellation of the excitation amplitudes for the two magnetic substates. This result does not depend upon the particular model potential considered but is a consequence of the odd and even time symmetry of the transition-matrix elements associated with the $2s$-$2p_0$ and $2s$-$2p_1$ transitions, respectively.

In conclusion, the observed correlation between an extremum value of λ and a phase change of χ of + or $-180°$ is explained and shown to be consistent with the current picture of excitation in collisions of quasi-one-electron systems. The phenomenon is, however, of a more general nature and should be observable in other ion-atom, atom-atom, and electron-atom collisions. Earlier experimental[10] and theoretical[11] results suggest that this is actually the case. However, the previously reported data points are so widely spaced that the rapid phase change has not been evident.

This work was supported in part by NATO travel grants.

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Observation of Electromagnetic Angular Momentum within Magnetite

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(Received 27 November 1978)

A hollow cylinder of magnetite at 4 K has been subjected to the simultaneous action of constant axial magnetic field $\vec{B}$ and low-frequency radial electric field $\vec{E}$. The reaction torque, which appears in the magnetite as the electromagnetic angular momentum changes, has been observed for the first time. Preliminary results favor the Livens' reaction-force density $-\partial(\epsilon_0\vec{E}\times\vec{B})/\partial t$ and contradict the alternative $-\partial(\epsilon_0\mu_0\vec{E}\times\vec{H})/\partial t$ of Einstein and Laub which was followed by Abraham and is widely accepted by modern authors.

The problem of specifying the total electromagnetic force density in material media has been under debate for over a century. Terms involving electromagnetic fields and their gradients have been understood and experimentally confirmed to such an extent that at present, no controversy seems to remain relating to them.[1] Thus there is now general agreement on the form of the tridimensional part of the electromagnetic energy-momentum tensor, the four-divergence of which is the force density. The final question, namely, whether the reaction-force density due to the rate of change of electromagnetic momentum is $-\partial(\vec{D}\times\vec{B})/\partial t$ (Minkowski), $-\partial(\epsilon_0\mu_0\vec{E}\times\vec{H})/\partial t$ (Einstein-Laub, or $-\partial(\epsilon_0\vec{E}\times\vec{B})/\partial t$ (Livens) has recently been under experimental investigation and the first alternative has already been conclusively ruled out at low frequencies.[2] In summary, we find that the remaining choice for the force density in isotropic materials without free charges or conduction currents is either

$$\vec{F}=(\vec{M}\cdot\nabla)\mu_0\vec{H}+(\vec{P}\cdot\nabla)\vec{E}+\tfrac{1}{2}\nabla(\rho_e+\rho_m)+\frac{\partial\vec{P}}{\partial t}\times\mu_0\vec{H}+\epsilon_0\mu_0\vec{E}\times\frac{\partial\vec{M}}{\partial t} \tag{1}$$