is approximately equal to that at $q = 0$. From approximately $q = 1.0$ Å^{-1} to $q = 1.5$ Å^{-1} it has the opposite sign to that at $q = 0$ and by 2 Å^{-1} it is no longer measurable. A preliminary analysis of the variation with q suggests that the data are sensitive to the short-range three-body terms as well as the triple-dipole term.

A fuller account of this project will be published elsewhere.

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[1]J. A. Barker, R. O. Watts, J. K. Lee, T. P. Schafer, and Y. T. Lee, J. Chem. Phys. 61, 3081 (1974).

[2]P. A. Egelstaff, A. Teitsma, R. McPherson, S. S. Wang, and S. D. Wang, to be published.

[3]B. M. Axilrod and E. Teller, J. Chem. Phys. 11, 299 (1943).

[4]N. J. Trappeniers, T. Wassenaar, and G. J. Wolkers, Physica (Utrecht) 32, 1503 (1966).

Calculation of *s-p* Coherence in Hydrogen after Electron Capture

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The electron capture by protons in a gas target is calculated in the Oppenheimer-Brinkman-Kramers approximation. The complete density matrix for the $n = 2$ hydrogen level is established. Electric-field–induced s-p coherence quantum beats are investigated. Good agreement with the experimental data is achieved.

Recently, Sellin *et al.*[1] found "a high degree of excitation coherence in electron capture to mixed-parity $n = 2$ states by fast protons in gases." This result was essentially derived from the intensities I^+ (I^-) of Lyα-Stark quantum beats in an external electric field F^+ (F^-) parallel (antiparallel) to the beam axis, exploiting the fact that the difference signal $(I^+ - I^-)$ is directly proportional to the coherence density-matrix element and odd in the electric field[2] while the sum signal $(I^+ + I^-)$ depends only on the incoherent part of the excitation (terms with even powers in the field).

In this Letter we briefly report on a theoretical interpretation of the new data given in Ref. 1. It is based on the, to our knowledge, first calculation of the complete ($n = 2$) density matrix including the coherence part. For the sake of simplicity we restrict ourselves to an extended Oppenheimer-Brinkman-Kramers (OBK) approximation.

In the following treatment we divide the complete interaction process into three steps: (a) the electron capture at small projectile-target distances ($\lesssim R_0$), (b) the post-collision interaction at large distances ($> R_0$), and (c) the final evolution of the density matrix under the influence of the applied electric field.

(a) Because of the short interaction time in the energetic ion-atom collision ($\tau_c \approx 10^{-16}$ s), the capture process is essentially spin independent and is not influenced by external fields. Charge transfer from the helium ground state (binding energy ϵ_i) to $n = 2$ hydrogenic orbital angular momentum states (binding energy ϵ_f) can therefore be calculated by ignoring the fine and hyperfine structures of the projectile. The impact-parameter–dependent capture amplitude at projectile velocity v in OBK approximation is given by[3]

$$A_{lm}(\vec{b}) = -i\int_{-\infty}^{R_0/v} dt \langle \varphi_{lm} | -|\vec{r}_p|^{-1} | \varphi_{\mathrm{He}(1s)} \rangle . \qquad (1)$$

Atomic units are used and the beam direction is chosen as quantization axis. The distance between the active electron and the projectile is denoted by $|\vec{r}_p|$. The wave functions used in Eq. (1) must include the translation factors that account for the projectile-target relative motion. Because of the exponential decay of the overlap integral at large distances, the upper bound in Eq. (1) can be pushed to $R_0/v = \infty$. Since in the OBK approximation the s-p coherence phase turns out to be rather insensitive to the details of the atomic-target orbital, we approximate the He ground state by a hydrogenlike wave function scaled to charge Z_i.

After the charge transfer, the unnormalized excited-state ($n = 2$) density matrix

$$\sigma_{lm,l'm'} = \int d^2b A_{lm}(\vec{b}) A_{l'm'}^*(\vec{b}) \qquad (2)$$

contains only five nonvanishing elements. (This

follows from axial symmetry and hermiticity). Four of them are real ($\sigma_{00,00}=\sigma_s$, $\sigma_{10,10}=\sigma_{p0}$, and $\sigma_{1\pm1,1\pm1}=\sigma_{p,\pm1}$). The diagonal part of Eq. (2) describes the usual OBK charge-transfer cross sections. The only nonvanishing off-diagonal element with $m_l=m_l'$, namely, $\sigma_{00,10}=\sigma_{sp}$, represents the capture-induced s-p coherence. For the latter we find

$$\sigma_{sp}=i(16Z_i^5\gamma\pi/3D^{12})(1-3/7D^2) \quad (3)$$

with $D=(\frac{1}{4}+\gamma^2)^{1/2}$ and $\gamma=(\epsilon_f-\epsilon_i)/v+\frac{1}{2}v$. Notice, that σ_{sp} is purely imaginary, independently of the velocity and the binding energy of both initial and final state. This is a direct consequence of the opposite parity of the s and the p state. The sign of σ_{sp} is, however, velocity dependent. It is positive for $v\gg1$ corresponding to a fixed phase of $+\frac{1}{2}\pi$.

(b) After electron capture, the excited hydrogenic state is perturbed by the post-collision interaction $H_I=-\vec{d}\cdot\vec{F}_I$ of the atomic dipole $\vec{d}$ with electric field $\vec{F}_I$ of the residual target ion. At large distances the electric field is essentially parallel to the beam direction. Fine-structure effects are negligible in this regime in view of typical interaction times $\tau_{post}\approx10^{-14}$ s. In the linear-Stark-effect regime the equation of motion for the density matrix can be solved in the decoupled spin-orbital angular momentum representation[4] yielding, for example, the coherence density-matrix element

$$\sigma_{sp}(R)=\cos^2(\tfrac{1}{2}\varphi)\sigma_{sp}+\sin^2(\tfrac{1}{2}\varphi)\sigma_{sp}^*-\tfrac{1}{2}i\sin\varphi(\sigma_s-\sigma_{p0}), \quad (4)$$

where $\varphi=(1/v)\int_{R_0}^{R}\Delta E_S\,dR$. The upper bound $R=v\,\tau_{post}$ of the integral may be taken as $R=\infty$ since the Stark splitting energy ΔE_S decreases as R^{-2}. The not uniquely defined lower bound was set equal to the mean radius of the excited state $R_0=\langle r\rangle_{n=2}=5.5$ a.u. as a plausible choice. It follows from Eq. (4) that the OBK coherence phase [Eq. (3)] is not affected by the post-collision interaction. However, the absolute value $|\sigma_{sp}|$ decreases, if $\sigma_s>\sigma_{p0}$ (a condition, that was shown to hold both experimentally[5] and theoretically).

(c) For detection times $t\gg\tau_{post}$ the density matrix evolves under the combined influence of the applied electric field and the spin-orbit interaction. The observed angular-dependent intensities $I^{\pm}(t)$ are given by $\sigma^{\pm}(t)$ averaged over the unresolved fine structure. For a 90° detection geometry and by again exploiting the axial symmetry, one finds that

$$I^{\pm}(t)\propto\langle p_{1/2,1/2}|\sigma^{\pm}(t)|p_{1/2,1/2}\rangle+\tfrac{5}{4}\langle p_{3/2,1/2}|\sigma^{\pm}(t)|p_{3/2,1/2}\rangle$$
$$+\tfrac{3}{4}\langle p_{3/2,3/2}|\sigma^{\pm}(t)|p_{3/2,3/2}\rangle-(1/\sqrt{2})\langle p_{1/2,1/2}|\sigma^{\pm}(t)|p_{3/2,1/2}\rangle\cos\omega_{fs}t, \quad (5)$$

where the state vectors are labeled by $|1_{j,m_j}\rangle$. The time evolution of $\sigma^{\pm}(t)$ is governed by the energy matrix

$$H=\begin{pmatrix}\omega_{fs}-i\Gamma_p/2 & 0 & -\sqrt{2}V^{\pm}\\ 0 & -\omega_0/2-i\Gamma_p/2 & V^{\pm}\\ -\sqrt{2}V^{\pm} & V^{\pm} & \omega_0/2\end{pmatrix}. \quad (6)$$

In writing the matrix elements a $p_{3/2,1/2}$, $p_{1/2,1/2}$, $s_{1/2,1/2}$ subspace representation was used. The exponential decay of the p state enters via Γ_p, its zero-field decay constant. The linear Stark effect manifests itself in the matrix element $V^{\pm}=\langle s_{1/2}|(-\vec{d}\cdot\vec{F}^{\pm})|p_{1/2}\rangle=\sqrt{3}F^{\pm}$. The energy matrix (6) also includes the Lamb shift ω_0, the fine-structure splitting ω_{fs} and $\varpi_{fs}=\omega_{fs}-\omega_0/2$. The density matrix emerging from step (b), exemplified by Eq. (4), serves as the initial conditions for the third process since $1/\omega_{fs}\gg\tau_{post}$. The following calculations make use of the fact that at the experimental field strength (525 V/cm) the Stark matrix elements $V^{\pm}$ between the $p_{1/2}$ and $s_{1/2}$ state are roughly of the same magnitude as ω_0. Therefore, the energy matrix (6) must be diagonalized in the $s_{1/2}$-$p_{1/2}$ subspace, while the influence of the $p_{3/2}$ state ($\approx10\omega_0$ away in energy) can be treated as a perturbation. Including corrections to first order in V/ω_{fs}, this method reproduces the experimental beat frequency $\varpi=15.32\pm0.05$ GHz within 1%. We are therefore confident that the final expression for the difference signal amplitude (which also contains $p_{3/2}$ state first-order contributions proportional to

V/ω_{fs}):

$$I^+ - I^- \propto \frac{V}{\omega}\,\mathrm{Re}\sigma_{sp}\left\{\exp(-\Gamma_1 t)\left[\frac{1+\omega_0/\omega}{2}\left(1+\frac{2V^2\omega_0}{\omega^2\overline{\omega}_{fs}}\right)+\frac{2V^2}{\omega\overline{\omega}_{fs}}\right]\right.$$
$$-\exp(-\Gamma_2 t)\left[\frac{1-\omega_0/\omega}{2}\left(1+\frac{2V^2\omega_0}{\omega^2\overline{\omega}_{fs}}\right)-\frac{2V^2}{\omega\overline{\omega}_{fs}}\right]-\frac{5\omega}{2\overline{\omega}_{fs}}\exp(-\Gamma_p t)$$
$$\left.-\exp(-\Gamma t)\left(\frac{\omega_0}{\omega}+\frac{4V^2}{\omega\overline{\omega}_{fs}}-\frac{8V^4}{\omega^3\overline{\omega}_{fs}}\right)\cos\overline{\omega}t\right\}-\frac{V}{\omega}\,\mathrm{Im}\sigma_{sp}\exp(-\Gamma t)\sin\overline{\omega}t. \quad (7)$$

is a rather good approximation to the physical situation. We have introduced the abbreviations $\omega=(\omega_0^2+4V^2)^{1/2}$, $\Gamma_1=\Gamma(1+\omega_0/\overline{\omega})$, $\Gamma_2=\Gamma(1-\omega_0/\overline{\omega})$, and $\Gamma=\Gamma_p/2$. Fortunately, increasing lifetimes of levels with increasing quantum numbers n prevent cascade effects from contributing appreciably to the Lyα signal. Small lifetime corrections and the effect of hyperfine splitting have also been neglected. The oscillating part in Eq. (7) contains, of course, Eck's[2] expression. Notice, however, the additional constant shift terms which are proportional to $\mathrm{Re}\sigma_{sp}$ and thus odd under reversal of the field. They deserve attention in connection with collision-induced dipole charge distribution in excited hydrogen states. For comparison with experiment the effects due to the finite length of the observation window and the finite length of the gas cell must be considered. Both effects lead to a reduction of the amplitude of the oscillating terms in the sum—and difference—signals. Since the fine-structure quantum-beat contribution to Lyα intensity [Eq. (5)] is washed out by averaging, the corresponding terms are omitted (the gas-cell length in Ref. 1, ≈ 0.05 cm, is greater than the fine-structure beat wavelength at $v=2.73$ a.u.).

In Fig. 1 we compare our theoretical calculations with the data of Sellin *et al.*[1] for (I^+-I^-) and I^+ of Lyα radiation after charge transfer between helium and a 186-keV proton beam at an applied field of 525 V/cm. Apart from a common trivial normalization factor for the curves, there remains only one adjustable parameter of the model. This is the effective beat-attenuation factor A, which incorporates the combined effect of the geometric reduction (experimental value, ≈ 0.75) as discussed above, the attenuation due to the field inhomogeneity near the gas-cell apertures,[1] and the attenuation due to the hyperfine splitting. Both curves I^+ and (I^+-I^-) agree quite well with the experimental data, if one chooses $A=0.55$.

We now turn to the offset of the He difference beat signal to positive values observed in Ref. 1 and suggested to be mainly caused by the experimental setup. According to expression (7), an alternative explanation seems possible. A combined phase and amplitude shift could result from a coherence phase different from $\frac{1}{2}\pi$ (i.e., $\mathrm{Re}\sigma_{sp}\neq 0$). Physically, this would correspond to an initial dipole moment associated with the excited hydrogen state, an effect reported earlier by Gaupp, Andrä, and Macek.[6] Further inspection of the signs for the various coefficients in Eq. (7) shows, however, that a simultaneous amplitude shift to positive values and a phase delay with respect to the negative-going sine oscillation is ruled out. Therefore the corrections of Ref. 1, also used in our fit, cannot, at present, be replaced by a more convincing explanation.

The good agreement between experimental and theoretical beat intensities in Fig. 1 could suggest that all OBK density-matrix elements are very

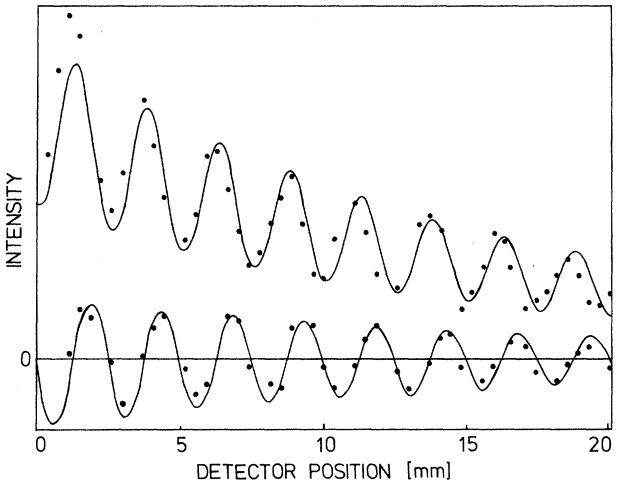

FIG. 1. Lyα intensity vs. distance downstream from the He gas cell at field strength 525 V/cm and proton velocity $v=2.73$ a.u.. Upper curve, calculated intensity I^+ for electric field parallel to the beam. Lower curve, signal difference between fields parallel and antiparallel to the beam (I^+-I^-). Experimental data are from Ref. 1 (after correction; see text).

accurate. This conclusion is only partially valid. First of all, over a wide range of velocities ($v = 1$ to 5 a.u.) we find high degrees of relative s-p coherence $|\sigma_{sp}|/(\sigma_s\sigma_p)^{1/2}$ up to 0.98. Qualitatively, this agrees with experimental values $\lesssim 0.8$ of Ref. 6 in the case of beam-foil interaction. Similarly, the phase of σ_{sp} is confirmed by the phase of the difference beat signal. Because of the complicated dependence of the modulation depth of I^+ and the amplitude ratio $(I^+ - I^-)/I^+$ on the relative cross sections σ_{pm}/σ_s, however, the fit in Fig. 1 does not allow a reliable estimate of these relative cross sections. For example, we have also found good fits to the data by inserting Shakeshaft's proton-hydrogen capture cross sections, determined within a seventy-coupled-states calculation,[7] but still using the OBK results for the degree and the phase of atomic coherence. Therefore, the agreement between experiment and theory should not be overestimated if one considers a relative cross sections in different substates. Further investigations concerning orientation and alignment[8] of the excited hydrogenic state after electron capture are in progress.

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[1]I. A. Sellin, L. Liljeby, S. Mannervik, and S. Hultberg, Phys. Rev. Lett. 42, 570 (1979).
[2]T. G. Eck, Phys. Rev. Lett. 31, 270 (1973).
[3]M. R. C. McDowell and J. P. Coleman, *Introduction to the Theory of Ion-Atom Collisions* (North-Holland, Amsterdam, 1970), p. 155.
[4]E. Kupfer and H. Winter, Z. Phys. A285, 3 (1978).
[5]M. J. Alguard and C. W. Drake, Phys. Rev. A 8, 27 (1973).
[6]A. Gaupp, H. J. Andrä, and J. Macek, Phys. Rev. Lett. 32, 268 (1974).
[7]R. Shakeshaft, Phys. Rev. A 18, 1930 (1978).
[8]U. Fano and J. Macek, Rev. Mod. Phys. 45, 553 (1973).

Absorption Line-Narrowing Experiment between Atoms of Two Different Species

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An experiment analogous to a three-level absorption line-narrowing experiment is performed between atoms of two different species (krypton and xenon) connected by a resonant excitation transfer. Results show that the hole burned in the velocity distribution of krypton atoms is transferred to xenon ones. This effect is interpreted by introducing a cross-collision kernel. Analysis of the experimental line shapes is consistent with a Gaussian shape of this kernel, whose width is found to be pressure independent.

Conventional absorption line-narrowing (ALN) experiments[1] involve a three-level system in atoms of a given species. A transition $a \leftrightarrow b$ is saturated by a first, intense laser beam, while the coupled transition $b \leftrightarrow c$ is probed by a second, weak laser beam [Fig. 1(a)]. The experiment which we describe in this paper is similar in principle to such an ALN experiment. However, it presents a fundamental difference: The saturated and probed transitions belong to atoms of *different species* (krypton and xenon, respectively). Here, the levels a of Kr and a' of Xe, connected by a resonant excitation transfer [Fig. 1(b)], play the role of the common level a of the usual three-level system. The remarkable feature that we have observed is that the *line-narrowing* effect remains. In other words, the hole burned in the longitudinal velocity distribution of the level a of Kr atoms gives rise through the excitation-exchange process to a hole in the level a' of Xe atoms. It is to be noted that the radiation fields cannot induce coherence between the levels b and c' of the pseudo three-level system of Fig. 1(b). This results in a simpler theoretical expression of the signal than in the case of the true three-level system of Fig. 1(a).

Our experiment can also be compared to a two-level saturated absorption experiment in the presence of foreign perturber atoms. In such an experiment, the same "active" atoms experience both the saturating and the probe fields. In a recent work of this type,[2] the modification of the