[1]T. M. Cormier *et al.*, Phys. Rev. Lett. 40, 924 (1978); T. M. Cormier *et al.*, Phys. Rev. Lett. 38, 940 (1978).

[2]H. Emling *et al.*, Nucl. Phys. A211, 600 (1973).

[3]R. Wieland *et al.*, Phys. Rev. C 8, 37 (1973); R. Wieland, Doctoral Dissertation, Yale University, 1972 (unpublished).

[4]K. A. Erb, D. A. Bromley, and J. Weneser, Comments Nucl. Part. Phys. 8, 111 (1978).

[5]P. P. Singh *et al.*, Phys. Rev. Lett. 28, 1714 (1972).

[6]J. J. Kolata *et al.*, to be published; F. Haas, private communication.

[7]H. J. Fink, W. Scheid, and W. Greiner, Nucl. Phys. A188, 259 (1972).

[8]Y. Abe, Y. Kondo, and T. Matsuse, Prog. Theor. Phys. 59, 1393 (1978); Y. Abe, in *Clustering Aspects of Nuclear Structure and Nuclear Reactions (Winnipeg, 1978)*, AIP Conference Proceedings No. 47, edited by W. T. H. Van Oers *et al.* (American Institute of Physics, New York, 1978); Y. Kondo, Y. Abe, and T. Matsuse, Phys. Rev. C (to be published).

[9]N. Austern and J. S. Blair, Ann. Phys. (N.Y.) 33, 15 (1965).

[10]J. V. Maher *et al.*, Phys. Rev. 188, 1665 (1969).

[11]A. Gobbi *et al.*, Phys. Rev. C 7, 30 (1973).

[12]R. A. Chatwin *et al.*, Phys. Rev. C 1, 795 (1970).

[13]W. Reilly *et al.*, Nuovo Cimento A13, 897 (1973).

[14]A. Arima, G. Scharff-Goldhaber, and K. W. McVoy, Phys. Rev. Lett. 40B, 7 (1972).

[15]F. J. W. Hahne, Nucl. Phys. A104, 545 (1967).

[16]R. H. Bassel, G. R. Satchler, and R. M. Drisko, Nucl. Phys. 89, 419 (1966).

[17]H. Feshbach, J. Phys. (Paris), Colloq. 37, C5-177 (1976).

Observation of Mixed-Parity Electric-Dipole Oscillations in Charge Transfer to the $n=2$ Hydrogen Levels by Fast Protons in Gases

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We find evidence for strong, collision-averaged excitation coherence between $2s$ and $2p_0$ amplitudes in charge transfer by fast protons ($v_i \approx 2$–3 a.u.) undergoing single collisions in He, Ar, and O_2. Quantum beat difference signals in Lyα (Eck beats) induced by reversible electric fields have amplitudes very similar to those seen earlier in C targets, supporting a "last-layer" (gas-layer?) hypothesis.

We have found a high degree of excitation coherence in electron capture to mixed-parity $n=2$ states by fast proton in gases. To our knowledge, there has been no previous experimental observation of such a phenomenon in charge transfer. Theorists working in the very active field[1] of charge transfer seem not to have concentrated on calculating the s-p capture amplitude differences as a test of various competitive theories of charge transfer. We show here that such phase differences are straightforward to measure for single ion-atom collisions, and hope that this paper will serve as a stimulus to calculation.

Since it was first shown in the work of Sellin *et al.*[2] that such mixed-parity excitation coherence is observable in beam-foil excitation, it has also been of interest to inquire whether such beats are induced by an exit surface capture or electric field effect as suggested by Eck[3] (characteristic of a solid-state effect) or whether such collision-averaged, mixed-parity state coherence is a prominent feature of charge transfer and perhaps other single ion-atom collision processes. Comparison of the character of electric dipole coherence in foil versus gas targets is then a promising tool for sorting out intrinsically solid state from binary ion-atom collision coherence phenomena.

In his original paper,[3] Eck proposed a simple technique for separating collision-averaged excitation coherence of H atoms from that induced by the electric fields required to couple levels of opposite parity, which otherwise do not decay to the same final state. The technique depends on applying reversible electric fields $\vec{E}$ parallel and antiparallel to the beam to exploit the fact that the excitation coherence quantum-beat signal is odd under reflection, whereas other signals are not. If there is an initial displacement of electron charge cloud with respect to the proton, or one develops in time because of an inequality in proton and average electron axial velocity, the displacement will be either enhanced or diminished depending on the direction of $\vec{E}$ relative to

that of the charge displacement. Incoherent coupling and quenching effects, light-intensity anisotropies, etc., depend on $|\vec{E}|$, but not on whether $\vec{E}$ is parallel or antiparallel to the quantization axis defined by the common axis ($+z$) of the beam and $\vec{E}$. Eck's first-order perturbation-theory expression for the Lyman-α beat signal is

$$\left(\frac{V}{\omega}\right)^2[\tfrac{1}{3}(\sigma_{p0}+2\sigma_{p1})-\sigma_s]\cos\omega t+\frac{1}{\sqrt{3}}\frac{V\omega_0}{\omega^2}\langle|f_s||f_{p0}|\cos\alpha\rangle\cos\omega t+\frac{1}{\sqrt{3}}\frac{V}{\omega}\langle|f_s||f_{p0}|\sin\alpha\rangle\sin\omega t, \quad (1)$$

plus terms smaller by a factor $\Gamma/2\omega$, all exponentially damped with a damping constant $\Gamma/2$. Here, Γ is the average of perturbed s- and p-state decay rates, ω_0 is the Lamb shift, ω is the perturbed $s_{1/2}$-$p_{1/2}$ level splitting ($\equiv\omega_0$ for $|\vec{E}|=0$), V is the dipole matrix element $(s_{1/2}|e\vec{E}\cdot\vec{r}|p_{1/2})=-\sqrt{3}\,eEa_0/\hbar$, $t=0$ at the time of excitation, and the excitation amplitudes for s- and p-state excitation are $f_s=|f_s|\exp(i\alpha_s)$ and $f_{p0}=|f_{p0}|\exp(i\alpha_p)$, with $\sigma=\langle|f|^2\rangle$ and $\alpha\equiv\alpha_s-\alpha_p$. The angular brackets refer to collision averages. When the perturbation is large, this expression is inaccurate, but can be substantially improved by using a better value for ω obtained by diagonalizing the 3×3 perturbation matrix, incorporating the $p_{3/2}$ state ($\sim10\omega_0$ away in energy). Hence the sum of the signals yields the incoherent oscillations superposed on nonoscillating perturbed $s_{1/2}$, $p_{1/2}$, and $p_{3/2}$ decays, while the difference yields only the coherent excitation terms, containing the s-p phase coherence angle α. If $\bar{\alpha}$ were 0 (or π), for example, the coherent part of the initial wave function $\psi(0)$ would involve $|f_s|u_s\pm|f_{p0}|u_p$, corresponding to a concentration of the electron charge distribution in the backward (+) or forward (−) hemisphere. Here u_s and u_p are the field-free spatial eigenfunctions of the $2s$ and $2p$ states. For $\bar{\alpha}$ near $\pi/2$, $\psi(0)$ would involve $i|f_s|u_s+|f_p|u_p$, corresponding to roughly zero *initial* charge-distribution asymmetry, but one which would reach peak concentration in alternate hemispheres at the subsequent times $t=\pi/2\omega$, $t=3\pi/2\omega$, etc.

The lower curves in Fig. 1 display a direct comparison of difference beat signals for He targets (~10 to 100 mTorr cm) and thin C (~15 μg/cm^2) targets[2] at the same beam energy (186 keV) and field strength (525 V/cm). A uniform normalization was obtained by normalizing the average intensity of the sum signal from the solid-target data[2] to the present He sum signal, facilitating direct comparison of the difference signals on the same vertical scale. The upper curve shows a higher point density signal for $E_{\text{para}}=+525$ V/cm. The excitation coherence beat signal in both He and C is very similar in size relative to the total sum curve intensity (~10% vs ~7%, respectively). As the cell is only $\frac{1}{2}$ mm thick internally, it is difficult to estimate target thickness accurately. It was, however, established by signal-versus-pressure linearity checks that for gases, approximately single-collision charge-transfer conditions prevailed in all cases.

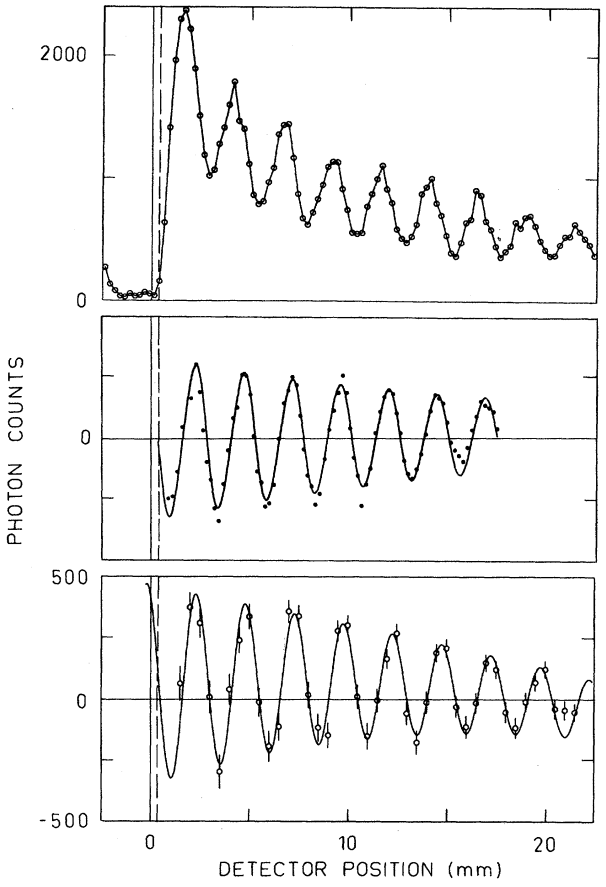

FIG. 1. Signal strengths vs distance downstream at $E=525$ V/cm and 186-keV proton energy. The lower curve shows a difference beat signal ($E_{\text{para}}-E_{\text{antipara}}$) for a helium target. Above it appears a difference signal for the ~15-μg/cm^2 C target of Ref. 2, uniformly normalized. The top curve shows an E_{para} signal in the He target. The solid line marks the geometric center of the gas cell. The dashed line marks both the effective center of the cell arising from the phase delay and the foil position.

The He difference beat signal is offset to positive values. Part of this offset is certainly due to a beam-induced microdischarge problem creating stray light when the downstream plate is negative, but a real offset cannot be completely ruled out at present.

The similarity of amplitudes in gas versus solid targets lends support to the "last-layer" hypothesis often used in beam-foil excitation models.[4] The gas layer adhering to exit surfaces may *cause* the similarity of the single-collision and solid-target beat amplitudes. The distributed length of a gas cell reduces the modulation amplitude through the wave-form averaging brought about by the spread in place (time) of formation. As long as the effective length of the cell is short compared to the beat wavelength (~ 30% in Fig. 1), such amplitude reduction is entirely tolerable (~ 0.8×).

It seems appropriate here to correct a widespread misunderstanding that has propagated through the beam-foil literature for years.[4] Misinterpretation of the intent of Macek's use of the impulse approximation in his early paper[5] on quantum beats in beam-foil light combined with misunderstanding of how the uncertainty principle applies has long delayed application of the time-of-flight quantum-beat method to the gas-target case. As clearly demonstrated in Fig. 1, the short time of passage through a foil target ($\lesssim 10^{-14}$ sec) represents no fundamental advantage whatever in achieving the impulsive excitation needed to observe superposition states. Since the states are produced in single collision, if there is a coherence time, it is instead the much shorter binary-collision time ($\lesssim 10^{-16}$ sec) which is relevant. *One might then expect coherent excitation of any multipole moment of any excited n state, consistent with symmetry and excitation times, for many ion-atom collision processes, and even coherent excitation of different n states.*

The experiment was performed by passing a $\frac{1}{2}$-mm-diam, $\frac{1}{2}$-deg-divergence, 186-keV proton beam through a $\lesssim 0.8$-mm-long gas cell having 0.5-mm apertures, whose 0.5-mm rear wall formed the upstream boundary of a coaxial parallel-plate condenser normal to the beam. Reversible fields were established by applying voltages to a plate 25 mm downstream. Signals were obtained using a BX762 Bendix encapsulated Channeltron in perpendicular viewing geometry, sensitive from 1150 to 1900 Å (peak sensitivity at Lyα). The detector viewed Lyα through a pair of vertical 0.5-mm slits, 1 cm tall, located 5 cm and 16 cm from the horizontal beam. The finite field of view caused wave-form averaging, producing an effective ratio of observed wave-form amplitude to actual of about 0.9×. Signal normalization was accomplished by a Faraday cup mounted behind a 1-mm hole in the downstream plate. The combined cell-length and viewing-length wave-form attenuation factor thus results in difference signals ~ $\frac{4}{3}$ as large as shown in Fig. 1. The solid curve drawn through the He difference oscillations in Fig. 1 results from least-squares fits to the form $A\exp(-\Gamma t)\cos(\omega t-\varphi)$, added to a second-degree-polynomial background needed to account for the stray light previously mentioned. The fitted frequency and damping constants were $\omega = 15.32 \pm 0.06$ GHz and $\Gamma = 0.33 \pm 0.05$ GHz, compared to values of 15.37 and 0.31 GHz derived from an elementary 3×3 Stark matrix diagonalization. The curve for the solid-target data was instead fitted[2] to the coefficients of the difference terms in Eq. (1), using calculated ω and Γ values.

Caution should be used in interpreting the apparent difference in φ between solid and gas targets. Since the field penetrating the gas cell must rise from a small value to full field in a distance ~ 1 mm, during which time the beat frequency evolves from ω_0 to ω, there is an apparent phase shift, which is manifested in the failure of the fitted sum curves to exhibit an extremum at the center of the gas cell. Under the plausible assumption that the phase delay is approximately given by the time delay corresponding to half the effective gas-cell thickness times $(\omega-\omega_0)$, an apparent gas-cell center position can be inferred. This position is indicated by the dashed vertical line in Fig. 1. Referred to this position, the phase of the difference oscillations in He—and in fact also in Ar and O_2—versus C targets are qualitatively similar, and the negative-going difference oscillations cross the base line near the origin in all cases. Again, a connection between ion-atom collision and the "last-layer" hypothesis—perhaps a *gas* layer—may be implied.

Gaupp, Andra, and Macek have made a more complete analysis[5] of extremely similar appearing difference signal data from their later, solid-target experiments performed at overlapping energies. In these data the negative-going difference signal fit also crosses the base line at slightly negative times. In contrast to the conjecture in Ref. 2 that the observed negative-going maximum would correspond to peak concentration in the backward hemisphere, they have calculated

that the electron is ahead of the proton on leaving the foil by $\sim\frac{1}{2}a_0$. Uncertainties in absolute initial phase become relatively unimportant when differences in phase $\Delta\varphi$ from different target gases are compared. We find, for example, that $\varphi_{O_2} - \varphi_{Ar} = -0.1 \pm 0.3$ rad, meaning that within errors the phase angles φ in the form $A\cos(\omega t - \varphi)e^{-\Gamma t}$ are very nearly equal for these two very different target gases. As a function of beam energy, $\varphi_{He} - \varphi_{Ar} = -0.4 \pm 0.2$, 0.0 ± 0.2, and 0.5 ± 0.4 at 110, 190, and 290 keV respectively, exhibiting a velocity dependence of φ so weak as to be nearly within the 1σ (1 standard deviation) statistical fitting errors quoted in this range of beam energies. Within these errors, φ has also been shown to be nearly independent of target gas. It is quite possible these dependences will be stronger at lower, more adiabatic collision velocities.

The present method thus provides an effective tool for sorting out intrinsically solid-state surface effects on atoms excited in foils *vis-à-vis* gases. Additionally, theories of ion-atom charge transfer may be tested by examining the relative phases of opposite-parity capture amplitudes.

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[1]D. N. Tripathy and B. K. Rao, Phys. Rev. A 17, 587 (1978), and numerous references therein; R. A. Mapleton, *Theory of Charge Exchange* (Wiley, New York, 1972).

[2]I. A. Sellin, J. R. Mowat, R. S. Peterson, P. M. Griffin, R. Laubert, and H. H. Haselton, Phys. Rev. Lett. 31, 1335 (1973).

[3]T. G. Eck, Phys. Rev. Lett. 31, 270 (1973).

[4]*Beam-Foil Spectroscopy*, edited by I. Martinson, J. Bromander, and H. Berry (North-Holland, Amsterdam, 1970), p. 344; *Beam-Foil Spectroscopy*, edited by S. Bashkin (North-Holland, Amsterdam, 1973); *Beam-Foil Spectroscopy*, edited by I. A. Sellin and D. J. Pegg (Plenum, New York, 1976).

[5]J. Macek, Phys. Rev. Lett. 23, 1 (1969); J. Macek, Phys. Rev. A 1, 618 (1970); A. Gaupp, H. Andra, and J. Macek, Phys. Rev. Lett. 32, 268 (1974).

Charged Droplets in Cryogenic ^{4}He Vapor

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We have measured the mobility of positive ions in ^{4}He vapor for temperatures between 1.3 and 2.0 K and for saturation ratios between 0.1 and 1.0. We present a model which relates the size of a charged droplet to its mobility and find good quantitative agreement with our data when we calculate the size of the droplet which forms about the ion with classical macroscopic thermodynamic arguments. The radius thus obtained ranges from 7 to 9 Å.

When a positive ion is formed in a vapor the polarization force on the surrounding atoms induces a pressure gradient about the charge. At some distance R, the increased pressure will equal the vapor pressure over a curved surface of that radius and thus a droplet of radius R should form. An equation relating the size of the droplet to the saturation ratio (p/p_{sat}) and temperature was first obtained by Thompson[1] by applying arguments from macroscopic thermodynamics on a microscopic scale.[2,3] When the vapor is ideal and the liquid dielectric constant is nearly unity, the (modified) Thompson equation may be written

$$kT\ln(p/p_{sat}) = 2\sigma/n_l R - \alpha e^2/2R^4, \quad (1)$$

where σ and n_l are the bulk values of surface tension and liquid density, and α is the atomic polarizability. There are at least two reasons why it is important to try to verify this prediction directly. The first is that similar arguments are central to classical nucleation theory but in that case their confirmation must be quite indirect.[2-5] The second is that if such microscopic droplets are shown to exist they will present a valuable opportunity to study the effect of finite size on the thermodynamic properties of a fluid. Helium vapor provides an excellent system in which to study droplet formation because its small surface tension at low pressures favors a large effect. In addition, there is already some tentative evidence from ac mobility measure-

