PHYSICAL REVIEW LETTERS

VOLUME 25 | 10 AUGUST 1970 | NUMBER 6

ZERO-FIELD QUANTUM BEATS SUBSEQUENT TO BEAM-FOIL EXCITATION*

H. J. Andrä†
Department of Physics, University of Arizona, Tucson, Arizona 85721
(Received 4 May 1970)

The creation of M_L alignment due to the foil excitation process is demonstrated with the observation of zero-field quantum beats in He 3^3P-2^3S 3880-Å, hydrogen H_α, and hydrogen H_β emission.

In this Letter we report the first observation of zero-field quantum beats subsequent to coherent beam-foil excitation of hydrogen and helium. In order to explain intensity fluctuations previously observed in He II emission,[1] it had been suggested that the impulsive foil excitation[2] ($\sim 10^{-14}$ sec) may create a coherent superposition of atomic wave functions[3]:

$$\psi(t) = \sum_{JM_J} a_{JM_J} u_{JM_J} \exp\left[-\left(\frac{\gamma_J}{2} + i\frac{E_J}{\hbar}\right)t\right],$$

where u_{JM_J} is a time-independent atomic eigenfunction, and the a_{JM_J} are expansion coefficients. The emission, due to the subsequent decay to some lower level, can then show time-dependent intensity oscillations for each polarization.[3] However, these early observations can readily be explained with the coherent mixing of fine structure (fs) levels of high excited states by the small motional electric field due to running the experiment in the earth's magnetic field.[4] Despite this, the suggested concept of coherent excitation remains of great interest for further applications of the beam-foil technique. Even in the presence of the earth's magnetic field, an experimental test can easily be carried out involving only low excited states where the field influence in negligible.

H^+ or $^4He^+$ ions were accelerated to 133 and 475 keV, respectively, and sent through a thin carbon foil (8 $\mu g/cm^2$). Of the emerging partly excited beam, a 1-mm section (equivalent to a time resolution of 0.25 nsec) was observed with a photon-counting system perpendicular to the beam axis through appropriate interference and polarization filters. The polarization is defined parallel and perpendicular to the beam direction, which is also used as quantization axis throughout this Letter. The intrinsic polarizing effects in the detection system were measured to be less than 0.5%.

In the ^{4}He I 3^3P-2^3S 3889-Å emission, quantum beats with a frequency of 655 ± 6 MHz were observed, corresponding to the fs separation $^3P_1$-$^3P_2$ of 658.55 MHz.[5] They can be described by $I(t) = A\exp(-\gamma t)[1 + B\cos(E_{12}t/\hbar)]$, where γ is the decay constant and E_{12} is the energy separation of the levels involved. The beat amplitude B and its phase (sign of B) change with the polarization direction detected. $B_\parallel = +0.070 \pm 0.005$, $B_\perp = -0.040 \pm 0.005$ [see Figs. 1(a) and 1(b)]. A residual beat amplitude equal to that of the sum of both polarization directions is found for a measurement without polarization filter. $B_\triangle = +0.030 \pm 0.003$ [see Fig. 1(c)]. However, only relative intensities were recorded so that the initial intensity constant A and the resulting overall polarization of the emitted light were not measured.

A calculation based on the coherent magnetic field mixing of initially incoherent $|JM_J\rangle$ wave functions gives a theoretical beat amplitude in the earth's magnetic field (0.3 G) of less than 0.5% for the frequency of interest.[6] Consequently the observed beats with an amplitude of 7%

must then be due to an initial coherent superposition of states. This initial wave function cannot be determined completely in the JM_J representation by the experimental results. However, based on the necessary condition of impulsive excitation[7] and on the assumption that spin forces can be neglected in the atom-foil interaction, the number of unknowns can be reduced. Only the relative initial population of the M_L states at $t=0$, with the spin randomly distributed, has to be determined. This relates the foil-excitation mechanism to electron or ion impact excitation.[8] There it is known that the different M_L states appear incoherently excited in a noncoincidence experiment. Accordingly, the foil excitation creates a completely incoherent mixture of initial $|M_L M_S\rangle$ states with cross sections $\sigma_0=\sigma_0(M_L=0)$, $\sigma_1=\sigma_{+1}(M_L=+1)=\sigma_{-1}(M_L=-1)$, and $\sigma_2=\sigma_{+2}(M_L=+2)=\sigma_{-2}(M_L=-2)$. Due to the spin-orbit interaction, one obtains for $t>0$

$$\psi(t,|M_L M_S\rangle)=\sum_{JM_J} C(LM_L SM_S|LSJM_J)u_{JM_J} \times\exp\left[-\left(\frac{\gamma_J}{2}+i\frac{E_J}{\hbar}\right)t\right],$$

where the $|M_L M_S\rangle$ indicate the initially excited wave functions out of which the coherent mixture of $|JM_J\rangle$ states develop. The C's are Clebsch-Gordan coefficients.

Applied to the He quantum beats, one can then determine the ratio of the cross sections which best fits the data: $\sigma_1/\sigma_0=0.8\pm0.1$ at 475 keV/atom. Several attempts to possibly fit the data assuming initial coherence between different $|M_L M_S\rangle$ states failed completely. This and the observation of beats with a nonpolarizing detector at 90° to the quantization axis confirm the concept of initial foil excitation of incoherent $|M_L M_S\rangle$ states.

The effect of possibly present electric fields (surface charging, contact potentials, and motional electric field) on the "zero-field" beats in the neutral-He emission can be neglected completely. (Only second-order Stark effect!) The situation changes drastically when these measurements are carried out on hydrogen. Already in a small electric field, the linear Stark effect may cause coherent mixing of the nearly l-degenerate fs levels, which may exceed the effect from the pure excitation coherence. However, considering the experimental geometry one can exclude at once surface charging as well as contact potentials from responsibility for the observed beats in the hydrogen emission. They could not produce a field uniform enough to explain the regularity of the beats. Only the motional electric field has then to be considered in more detail as a perturbing influence on zero-field measurements with hydrogen. An estimate has been made with a Stark mixing calculation[6,9] based on initial incoherent $|JM_J\rangle$ wave functions which shows that up to $n=4$ no beat amplitude of the level pairs of interest exceeds 2% in a field of 2 V/cm (maximum motional electric field in this experiment).[9] This is less than observed [Figs. 1(d)-1(g)]. An additional argument that the recorded beats are indeed due to excitation coherence can be based on the beat phases. It can be shown that they would not change with polarization when the coherence is field induced.

Another difficulty arises from the l degeneracy. Except for $n=2$, it is not possible to determine the beat amplitude of a given $j(=l+\frac{1}{2})$-$j(=l-\frac{1}{2})$ level pair because of the unknown background due to the decay of the other l states of

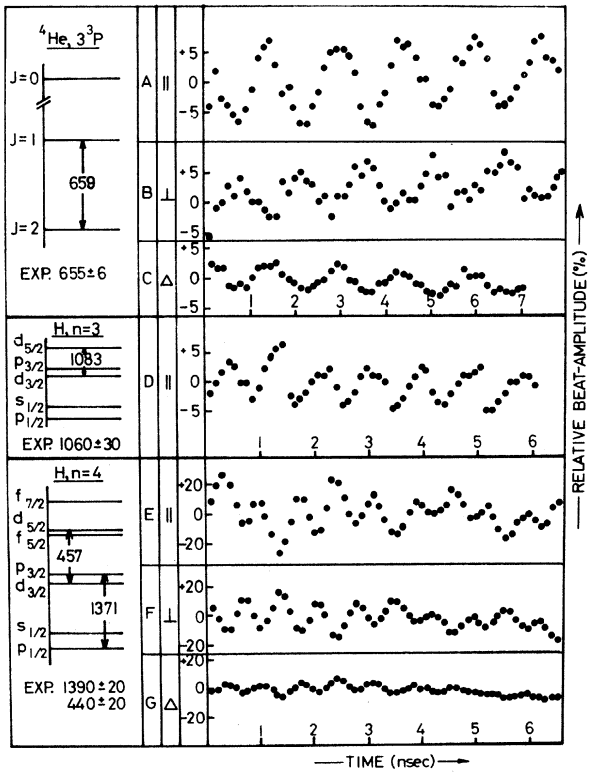

FIG. 1. Schematic level diagrams and observed quantum beats of ^{4}He 3^3P at 475 keV/atom; H, $n=3$, and H, $n=4$ at 133 keV/atom. Note the different amplitude scale for H, $n=4$ and the different time scale for He. All frequencies are given in MHz. The detected polarization is indicated by $\|$ for parallel and $\perp$ for perpendicular to the beam axis. Δ is assigned to measurements without polarization filter.

the same n. Therefore only comparative results can be given for the M_L cross sections based on the information carried by the beat phases. Because of this restriction, it is not necessary to include the hyperfine interaction, which has a strong influence on the beat amplitudes but not on their phases.[8]

The observed beats in the H_α emission have an amplitude with negative phase: $B_\parallel = -0.030 \pm 0.005$ [Fig. 1(d)]. They correspond to the $d_{3/2}$-$d_{5/2}$ fs separation (1083.27 MHz)[10] with a measured value of $\nu_{dd} = 1063 \pm 30$ MHz, which is the only resolvable frequency with the experimentally set time resolution for $n = 3$. The change of the phase with polarization has been checked. From its sign one deduces $\sigma_2/(\sigma_0 + \sigma_1) > 1$ at 133 keV/atom.

In the H_β emission, a superposition of beat frequencies is observed; the data can be decomposed into frequencies $\nu_{pp} = 1390 \pm 20$ MHz and $\nu_{dd} = 440 \pm 20$ MHz. They correspond to the fs separations $p_{1/2}$-$p_{3/2}$ (1371.07 MHz) and $d_{3/2}$-$d_{5/2}$ (457.03 MHz), respectively. From the beat phases one deduces for the two p states $\sigma_1/\sigma_0 > 1$ and for the two d states $\sigma_2/(\sigma_0 + \sigma_1) < 1$ at 133 keV/atom.

An additional slight modulation of the $p_{1/2}$-$p_{3/2}$ beats was observed. Its explanation could be based on the hfs interaction or on the possible coherent excitation of different l states (s-d). However, further rigorously field-free experiments, especially extending to longer delay times, are necessary in order to obtain more precise data for a nonspeculative Fourier-analytical interpretation.

These considerations show that the assumptions employed for the successful description of the experimental results need refinement and that the initial wave functions may not be unique. In this context also the measurement of the energy dependence of the M_L cross-section ratios in comparison with those found in electron-impact collisions[8] is of great interest for the further understanding of the foil-excitation mechanism.

However, with the observation of the zero-field beats, it has been established that the foil excitation produces aligned excited states. This alone opens a wide field for application of the beam-foil technique to Hanle effect, level crossing, quantum beat, and magnetic resonance experiments, especially on ionized atoms, which were not easily accessible previously. So far, experiments of the first three types have been carried out successfully and will be reported elsewhere in more detail.

I would like to express my appreciation to Professor S. Bashkin for the hospitality I enjoyed in his group, and to Dr. T. Hadeishi for his great interest in this work. I also wish to thank Professor A. I. Sherwood for many encouraging discussions.

---

*Work supported in part by the National Aeronautics and Space Administration, the Office of Naval Research, and the Aerospace Research Laboratories, Office of Aerospace Research, U. S. Air Force Office of Scientific Research, under Contract No. F33615-70-C-1007.

†Present address: I. Physikalisches Institut der Freien Universität Berlin, 1000 Berlin 33, Germany.

[1]S. Bashkin and G. Beauchemin, Can. J. Phys. 44, 1603 (1966).

[2]L. Kay, Phys. Lett. 5, 36 (1963); S. Bashkin, Nucl. Instrum. Methods 28, 88 (1964); S. Bashkin, in *Beam-Foil Spectroscopy*, edited by S. Bashkin (Gordon and Breach, New York, 1968).

[3]J. Macek, Phys. Rev. Lett. 23, 1 (1969).

[4]W. S. Bickel and S. Bashkin, Phys. Rev. 162, 12 (1967).

[5]J. P. Descoubes, C. R. Acad. Sci. 259, 327 (1964).

[6]H. J. Andrä, to be published. For a first introduction see the similar formalism in Ref. 9.

[7]P. A. Franken, Phys. Rev. 121, 508 (1961).

[8]I. Percival and M. Seaton, Phil. Trans. Roy. Soc. London, Ser. A 25, 113 (1958), and Colloq. Int. Centre Nat. Rech. Sci., No. 162, 21 (1967).

[9]I. A. Sellin, C. D. Moak, P. M. Griffin, and J. A. Biggerstaff, Phys. Rev. 184, 56 (1969).

[10]All hydrogen fs separations are taken from J. D. Garcia and J. E. Mack, J. Opt. Soc. Am. 55, 654 (1965).