Hyperfine Quantum Beats in Oriented ^{14}N IV†

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Quantum beats have been observed in both linear- and circular-polarized light using a beam–tilted-foil geometry for the $3s\,^3S$-$3p\,^3P$ transition of ^{14}N IV. Beats between the $F=0$ and $F=1$ hyperfine levels—forbidden in the case of excitation by untilted foils—were observed. A value of $A=695 \pm 15$ MHz was obtained for the hyperfine coupling constant of the upper term.

Much recent attention has been given to the measurement of quantum beats produced by atomic alignment in the beam-foil source, and its use in determining unresolved fine and hyperfine structures. The initial suggestion for such a technique was made by Macek[1] and his prediction was verified by Andrä[2] for ^{1}H I and ^{4}He I fine structure. More recently, extensive measurements have been carried out for the light atoms ($Z \leq 4$).[3] Until now, however, all field-free modulations observed were due to differences in the excitation cross sections to different m_L states; no excitation coherence between different m_L states was necessary or observed. It has been suggested theoretically[4,5] and verified experimentally[6] that if the cylindrical symmetry of the beam-foil source is broken by tilting the foil, atomic orientation can occur leading to the emission of circularly or elliptically polarized light, demonstrating that excitation coherence between different m_L states is produced. In this paper, we demonstrate the feasibility of using this effect to measure hyperfine structure by the observation of quantum beats in circularly polarized light. The work of Ellis[4] showed that in a tilted-foil geometry such beats should occur. Of particular interest is the prediction that $J=0$ to $J=1$ (or $F=0$ to $F=1$) quantum beats which do not occur in linearly polarized or unpolarized light —and which are forbidden entirely for untilted foils—should be observable if circularly polarized light is detected. Hence the observation of $J=0$ to $J=1$ quantum beats can provide a sensitive measure of orientation in a time-resolved atomic excitation, and possibly allows the measurement of fine and hyperfine structure in cases where alignment is lacking. This Letter reports the first measurement of this phenomenon.

The Dynamitron accelerator at Argonne National Laboratory provided beams of a few microamperes of ^{14}N$^+$ at 2.0 MeV which were excited by carbon foils mounted either perpendicular to the beam (0° tilt) or at 45° to the beam. The light emitted perpendicular to both the foil normal and

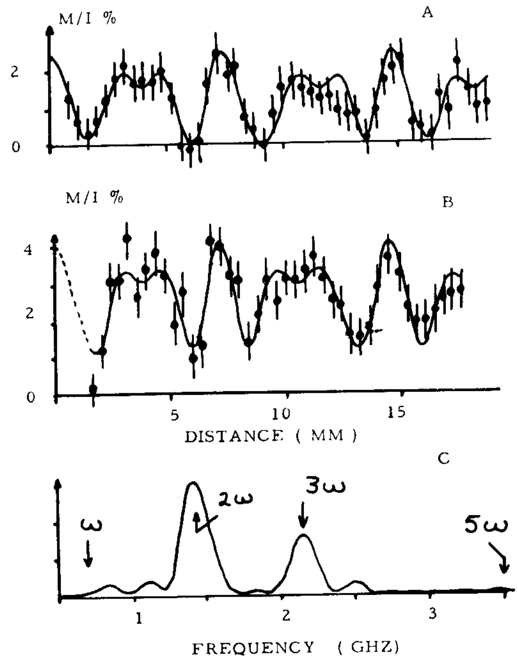

FIG. 1. The Stokes parameter ratio (M/I) for the N IV 3480-Å transition: (a) 0° foil, (b) 45° foil, (c) the Fourier transform of (b). The line in (a) and (b) is a fit based on Eq. (4).

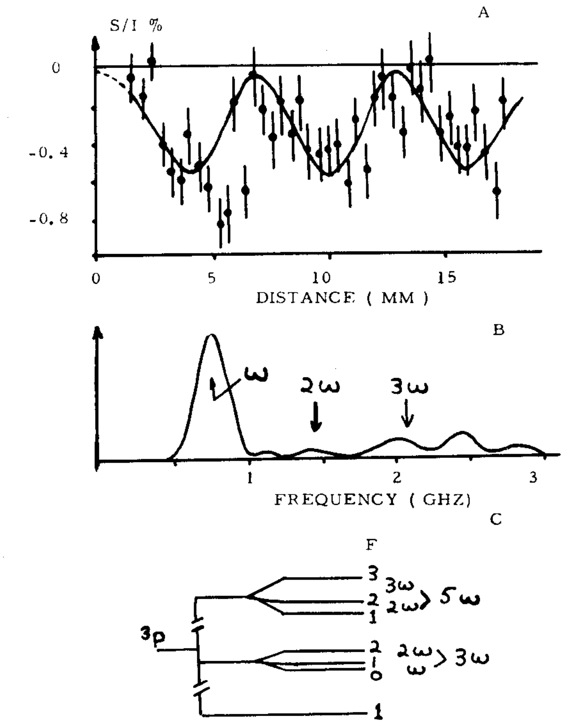

FIG. 2. (a) The Stokes parameter ratio (S/I) for the N IV, 3480-Å transition; (b) the Fourier transform of (a); (c) the energy levels and frequencies of the upper levels $3p\ ^3P_{0,1,2}$, assuming the nuclear magnetic dipole interaction $E(FJ) = \frac{1}{2}A\{F(F+1) - J(J+1) - I(I+1)\}$.

the beam was focused on the entrance slit of a $\frac{3}{4}$-m Czerny-Turner monochromator set to pass the N IV $3s\ ^3S_1$-$3p\ ^3P_{0,1,2}$ transitions at 3478-3485 Å and equipped with a Bailey Centronic 4283 photomultiplier. Between the beam and the monochromator entrance slit was placed a quarter-wave plate, with its fast axis parallel to the beam axis, followed by a focusing lens, a uv linear polarizer, and—finally—a Hanle depolarizer, similar to the experimental arrangement described previously.[6] An on-line computer controlled a stepping motor which rotated the linear polarizer, generally in steps of 90°, while a second stepping motor provided translation of the foil parallel to the beam, typically in steps of $\frac{1}{64}$ in. ≃0.4 mm. The relative Stokes parameter M/I of the emitted light was measured at each foil position by making sets of four measurements (four 90° rotations) with the axis of the linear polarizer alternately parallel and perpendicular to the beam. The relative Stokes parameter S/I was similarly measured using four 90° rotations but with the polarizer axis starting at 45° to the beam axis. For the measurements using the 45° exciter foil, a horizontal-slit beam aperture was inserted ahead of the foil. The slit dimensions of 1×6 mm enabled a reasonable spatial resolution of viewing along the beam to be maintained.

The results of the measurements of the quantum beats in M/I and S/I are shown in Figs. 1 and 2, respectively. ^{14}N has a nuclear spin $I=1$ with a small nuclear magnetic moment giving rise to hyperfine structure which has never been resolved optically. The only previous measurements of nitrogen hyperfine structure are of the N I ground state by Lambert and Pipkin[7] and in N IV by Desesquelles, Gaillard, and Silver.[8]

In Fig. 2(c) we show the level structure for the transition observed and indicate the hyperfine frequencies which are expected to contribute to the quantum beats in the $3p\ ^3P$ decay. Using the notation of Ellis,[4] and assuming a spin-independent interaction at the foil, we can write the intensity of the decay curve, neglecting cascades, in the form

$$I(t) = I_0 e^{-\Gamma t}\{B + \sum G(JF, J'F') \cos(\omega_F - \omega_{F'})t\}, \qquad (1)$$

the sum being taken over all pairs of upper levels, where

$$B=\sum_{JF} G(JF, JF) \tag{2}$$

and

$$G(JF, J'F')=[L][F][F'][J][J']\sum_k (l^k\cdot\rho^k)\begin{Bmatrix} F F' k \\ J' J I\end{Bmatrix}^2\begin{Bmatrix} J J' k \\ L L S\end{Bmatrix}^2\begin{Bmatrix} L L k \\ 1 1 L_f\end{Bmatrix}. \tag{3}$$

The density matrix ρ and the light-polarization detection operator l have here been expanded in terms of irreducible tensor elements ρ_q^k and l_q^k, and $[a]\equiv 2a+1$. If we define the z axis along the beam, and the foil normal is always in the y-z plane, then reflection symmetry with respect to the y-z plane requires that only the following five independent density matrix parameters for $k\leq 2$ can be nonzero: ρ_0^0, proportional to the total intensity; ρ_1^1, the orientation parameter; and ρ_0^2, ρ_1^2, ρ_2^2, the alignment parameters.[9]

The relative Stokes parameter M/I can then be written in the form

$$M/I=I_1(3\rho_0^2+6^{1/2}\rho_2^2)(311+235\cos 2\omega t+190\cos 3\omega t+14\cos 5\omega t), \tag{4}$$

where small oscillating terms in the denominator have been neglected and where I_1 is a constant determined by Eq. (1). For our case of small alignment, the denominator $I=I^{\parallel}+I^{\perp}$ is dominated by a term proportional to ρ_0^0, which is independent of frequency. Note that the $F=0$ to 1 frequency ω does not appear in Eq. (4).

For the perpendicular foil, where axial symmetry obtains, the orientation vanishes and the alignment can be described by the single parameter ρ_0^2 which is proportional to the difference of the excitation cross sections to $m_L=\pm 1$ and $m_L=0$ in the upper P state. As the foil is tilted, it may happen that $\rho_2^2\neq 0$ corresponding to coherence between different m_L states, but the relative amplitudes of the quantum beats are unchanged. This is shown clearly in Figs. 1(a) and 1(b) where only the overall beat amplitude changes due to possible changes in ρ_0^2 and ρ_2^2. The fractional polarization is small, and the 5ω frequency (which may be only partially resolved within the resolution of this experiment) is too weak to be observed. From the value of ω, the value for the hyperfine coupling constant of the upper level $A=695\pm 15$ MHz is obtained. A simple calculation of A from the contact term of the $2s$ electron gives a value of 560 MHz, in qualitative agreement with this measurement.

Similarly, S/I may be written in the form

$$S/I=I_2\circ\rho_1^1(121+6\cos\omega t+21\cos 2\omega t+14\cos 3\omega t). \tag{5}$$

In the circular polarization, the 5ω frequency should not appear, and it can be seen from Fig. 2 that the $F=0$ to $F=1$ quantum beat appears to dominate the polarization curve. It is most probable that the higher frequencies 2ω and 3ω have reduced amplitudes resulting from loss of spatial resolution. This is due in part to the finite slit width in front of the 45° foil, and also to deterioration of the foils with changes in thickness during the measurement; up to six foils were used for each single measurement of approximately 20 h. The 5×10^5 counts recorded for each polarization direction at each point gives a statistical error of 0.14% per point compared to the beat amplitude of (0.25 ± 0.05)%. The resulting frequency corresponds to a hyperfine coupling constant $A=750\pm 50$ MHz, in good agreement with the previously quoted result.

Our observation of quantum beats in circular polarization shows that the titled-foil final surface interaction produced a very weak coherence between the different m_L states of the $3p\,^3P$ state of N IV. In contrast to previous measurements of collision-induced coherence,[6] we find that the coherence-induced orientation is much smaller than the alignment produced from the differences in m_L cross sections $\sigma(|m_L|)$, which were already small for this state. The prediction that $F=0$ to $F=1$ quantum beats—unobservable for straight foils—can be observed in circular polarization with tilted-foil excitations has been verified. Moreover, the fact that the amplitudes of the observed beats in circular polarization are proportional to the atomic orientation rather than the alignment may have important consequences. Very little alignment has been observed in the beam-foil excitation of heavy ions. If substantial orientation can be produced by the technique suggested here, significant fine- and hyperfine-structure measurements can be carried out even in the absence of alignment.

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[9]These parameters are proportional to the Fano-Macek parameters of Ref. 5. The relations given in Berry, Curtis, Ellis, and Schectman, Ref. 6, need to be corrected by a factor of $\frac{2}{5}$, i.e., $A_0^{\rm col} = \frac{2}{5} \times (\frac{3}{10})^{1/2} \rho_0^{\,2}$.