PHYSICAL REVIEW
LETTERS

VOLUME 30 | 29 JANUARY 1973 | NUMBER 5

Hyperfine-Structure Studies of ^{3}He by Zero-Field Quantum Beats

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(Received 26 June 1972)

A new beam-velocity–independent data analysis is used in a zero-field quantum-beat measurement for the determination of the magnetic-dipole hyperfine-interaction constants of the 3^3P and 4^3P states of ^{3}He.

In recent years the method of zero-field quantum beats[1-3] subsequent to beam-foil excitation has been developed as a new tool for high-resolution measurements in atomic spectroscopy. This method is based on an interference effect arising from the decay of coherently excited, closely spaced and aligned levels.[4] This effect can be observed as superimposed oscillations in the time-differential detection of intensity decay curves.[5] Since the frequencies of these oscillations correspond to the level separations involved, the direct measurement of fine-[2,3,5-8] or hyperfine-structure (hfs) splittings becomes possible. However, no hfs measurement has been reported yet, and the achievement of accurate measurements has been inhibited so far by the difficulty in measuring beam velocities accurately enough to enable this method to compete with other methods.

Recently, the authors have noted that the ratio of simultaneously observed frequencies can be measured to high precision.[3] Thus any physical quantity, which is uniquely determined by beat-frequency ratios, can be measured quite accurately. This is the case for hfs coupling constants as soon as the $\Delta J=0$ and $\Delta J\geq 1$ beat frequencies with arbitrary ΔF can be resolved simultaneously. In order to demonstrate this possibility we have determined the magnetic-dipole hf coupling constant A of the 3^3P and 4^3P states of He^3 using this new velocity-independent data analysis in the first hfs measurement with zero-field quantum beats.

A $^3He^+$ beam was accelerated to 210 keV, corresponding to a velocity of 3.66 mm/nsec, and was sent through carbon foils with 8 μg/cm^2 and 3 mm diameter, which were placed and could be translated along the beam axis within a high-precision target chamber with side-on viewing[9] geometry (described earlier[3] in some detail). This observation geometry together with an optical solid angle of 0.183 sr results in an effective Gaussian slit function $g(x-x')$ with a full width at half-maximum of 0.18 mm, thus guaranteeing a time resolution of $\Delta t=5\times10^{-11}$ sec. The only external field present was Earth's magnetic field, which affects the final result less than one part in 10^3. The data were taken by a multiscaling technique with each channel correlated to a certain foil position by a stepping motor drive. This drive scanned the foil over the whole delay length more than forty times during each measurement. Thus all channels were counting at the same averaged beam velocity whose stability was proved by the resolved beat pattern itself, since an unstable velocity would wash out this pattern. With this experimental arrangement the 3^3P and 4^3P zero-field quantum beats of ^{3}He were measured in the 3^3P-2^3S 3889-Å and 4^3P-2^3S 3118-Å emission with adequate time resolution, as shown in Fig. 1(c) for the 3^3P state which is used as the example for the discussion throughout this paper.

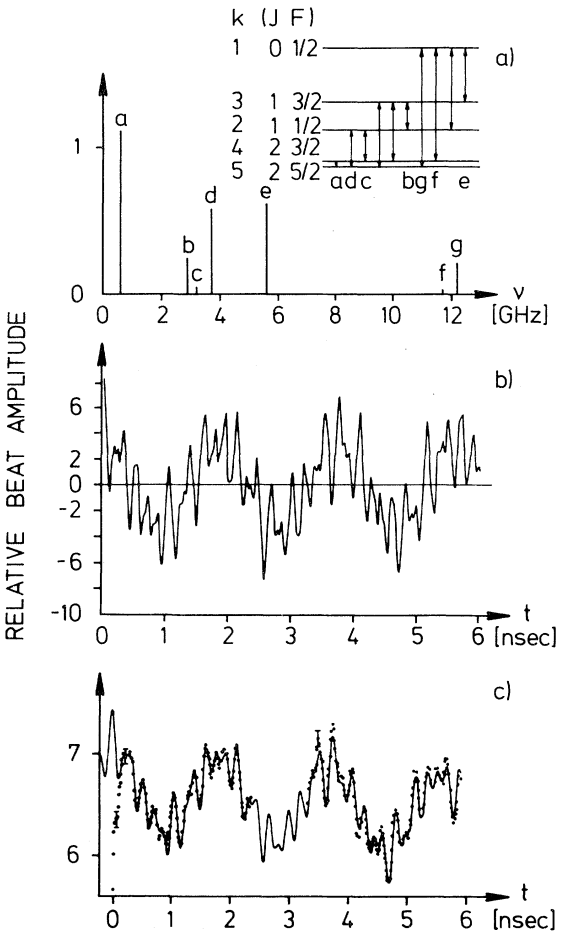

FIG. 1. (a) Expected quantum-beat frequency spectrum of 3^3P, calculated with $A_0 = -0.1440$ cm^{-1}. (b) Fourier transform of (a). (c) Measured intensity beats of the 3^3P-2^3S 3889-Å emission line as a function of time, computer fitted by Eq. (2). The decrease of intensity close to $t = 0$ is caused by optical shadowing due to the foil holder.

The gap in the data between 2.3 and 3.3 nsec was introduced on purpose, since such a measuring procedure is equivalent to the "separated oscillating field" technique in rf spectroscopy, as invented by Ramsey,[10] and allows for better frequency resolution, as will be discussed in a future publication.

The theoretical description of the zero-field quantum-beat phenomenon was basically given in earlier papers[1-3,5] and, as has been recently shown, can be given in a closed form for hfs measurements[11] provided that $LS(J)I, F$ coupling is valid for the excitation and emission process. This coupling scheme, with J being a good quantum number, holds to a good approximation only for such cases where the hfs splitting is small compared to the fs splitting, which is not true for the ^{3}He n^3P states in mind.[12,13] Therefore one has to use a more elaborate theoretical technique which is mainly concerned with the mixing of different J states with the same FM_F quantum numbers due to the hf interaction operator $\vec{I}\cdot\vec{S}$, as described in a fundamental paper by Güttinger and Pauli[14] for the hfs of Li^+. Thus one obtains new eigenstates $|k\rangle = \sum_J b_{Jk} |JFM_F\rangle$ of the atom in terms of the $|JFM_F\rangle$ states for which the energy and expansion coefficients b_{Jk} are calculated by diagonalization of the complete fs and hfs perturbation matrix in the JFM_F representation with the matrix elements given by Ref. 14. With these expansion coefficients b_{Jk}, the eigenfunctions are known and all coefficients can be calculated in the standard formula for zero-field quantum beats of P states,[1-3,6,8] which has to be convoluted with the slit function $g(x - x')$:

$$I(x/v) = \int_{-\infty}^{\infty} [a\sigma_0 + b\sigma_1 + (\sigma_0 - \sigma_1) \sum_{k,k'} c(k,k') \times \cos(\omega_{kk'} x/v)] g(x - x') e^{-\gamma x} dx'. \quad (1)$$

From this equation it is evident that, for the observation of quantum beats, the cross sections for the initial population of the M_L sublevel, given by $\sigma_{|M_L|}$, must fulfill the alignment condition $\sigma_0 \neq \sigma_1$. In the He^3 case this alignment can be assumed to be the same as that measured in the electronically equivalent ^{4}He states.[2,3,8] The beat frequencies $\omega_{kk'} = (E_k - E_{k'})/\hbar$ as well as the beat amplitudes $c(k,k')$ depend on A and the two fs separations Δ_{01} ($J = 0 \rightarrow J = 1$) and Δ_{02}. Although it would be possible to fit all three parameters in cases where at least three independent frequency ratios were measured, we would rather take the fs separations known with high accuracy from He^4 measurements.[15-17] Thus for our further considerations the beat frequencies $\omega_{kk'}$ and the relative beat amplitudes $c(k,k')$ depend only on A.

As a result, the frequencies $\omega_{kk'}$, although nonlinear, can be approximated by linear functions of A, $\omega_{kk'} = \omega_{kk'0} + m_{kk'}(A - A_0)$, in a narrow region around a theoretical value A_0. These functions are shown in Fig. 2 normalized to 1 at $A = A_0$. From this figure it is obvious that as a function of A only well-defined sets of relative frequencies (that is, well-defined beat patterns) are allowed in the experiment. Or, vice versa, a measured beat pattern uniquely determines A_{exp}. Moreover, since the beam velocity enters only as a common scaling factor for *all* frequen-

cies in a measured beat pattern, this pattern itself is independent of the velocity and therefore determines A_{exp} completely independent of the velocity. This is equivalent to the determination of A_{exp} from all measured frequency ratios. Thus by inserting the frequency interdependence of Fig. 2 into a fitting function

$$F=\left\{C_1+\sum_{k,k'}C_2(k,k')\cos[\omega_{kk'0}+m_{kk'}(A-A_0)]\frac{x-C_3}{v}\right\}\exp(-C_4x)+C_5, \tag{2}$$

which is used in least-squares fits to the data, one can leave A and v as free parameters and obtains A_{exp} essentially from the ratios of all measured frequencies. The constants C_1 to C_5 are also left as free parameters. C_1 represents the nonoscillatory part, and the $C_2(k,k')$ are the beat amplitudes of the frequency $\omega_{kk'}$, which have to be fitted independently because of the convolution with $g(x-x')$, which affects the calculated amplitudes as a function of frequency. C_3 represents the point of excitation, C_4 the inverse of the natural lifetime and the effect of the beam divergence, and C_5 the background and cascade contributions, which are considered to be mainly incoherent and constant in the time interval observed. (The problem of coherent cascading has been discussed by Macek.[18] Following his arguments we expect the beat amplitudes from feeding levels to be strongly reduced in the frequency region observed.)

For the application of this fitting procedure, A_0 has to be evaluated. Since the main contribution to A in an excited two-electron atom is resulting from the 1s-electron spin density at the nucleus, a first approximation is found with a hydrogen wave function for this electron, yielding $A=-0.1438$ cm^{-1}. Here the definition of A from Ref. 14 is used, which makes $A=\frac{1}{2}a_{s_1}$ independent of J with a_s defined by Kopfermann.[19] The contribution of the p electrons in ^3P states[20] and higher-order corrections[21] sum to a factor of 1.001. Thus we adopt $A_0=-0.144$ cm^{-1} for both 3^3P and 4^3P states.

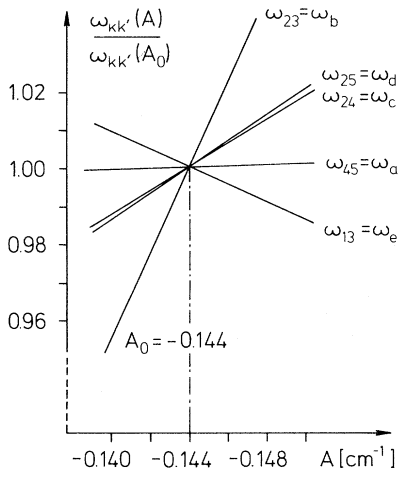

FIG. 2. Normalized beat frequencies $\omega_{kk'}$ of the 3^3P term of He^3 as a function of the hf coupling constant A.

With this A_0 value and the assumed alignment, we calculated the beat-frequency spectrum in Fig. 1(a) and the complete quantum-beat pattern in Fig. 1(b). Except for some washing out of high-frequency components due to the finite time resolution, one finds visually excellent agreement between Fig. 1(b) and the experimental result in Fig. 1(c). Thus A_0 is perfectly well suited as a starting value for the data-fitting procedure.

The fits obtained as in Fig. 1(c) yield rather accurate results, $A_{exp}(3^3P)=-0.1145\pm0.0004$ cm^{-1} and $A_{exp}(4^3P)=-0.1450\pm0.0020$ cm^{-1}, with the goodness of these fits determined by a χ^2 test,[22] yielding a confidence level of 0.04; that is, the probability of χ^2 to be above the critical χ_c^2 was only 4%. The reasons for the larger uncertainty limits of $A_{exp}(4^3P)$ are easily found: first in the lower intensity, and second in the fact that only two beat frequencies have markedly different slopes as a function of A in the case of 4^3P in comparison to five such frequencies in the case of 3^3P (Fig. 2).

The agreement with earlier experimental results[15] as well as with the theoretical evaluation is very good. Thus these measurements demonstrate that the main drawback of zero-field quantum-beat measurements—the uncertainty in the beam velocity—can be eliminated in hfs measurements by applying our new data-analysis technique, which is not limited only to cases where $\Delta J=0$ and $\Delta J\geq1$ beat frequencies with arbitrary ΔF can be resolved simultaneously. Applying external magnetic fields to the beam, the beats between Zeeman-split level separations with ΔJ, $\Delta F=0$ become observable at the same time, together with $\Delta J=0$, $\Delta F\neq0$ hf-splitting beats. The measured frequency ratios determine then again the hf separations at zero field, completely independent of the beam velocity, if the g values and the magnetic field strengths are known. That is, the use of external magnetic fields in conjunction

with our data analysis shifts the problem of measuring accurate beam velocities to the problem of determining accurate field strengths, which is a common difficulty to all other methods also.

In conclusion, we are convinced that by employing our data-analysis technique, the quantum-beat method extended to long delay times will become competitive with other methods and even superior as soon as ionic states can be studied.

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Optically Detected Electron Spin Echoes and Free Precession in Molecular Excited States

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Electron free precession and electron spin echoes have been detected for molecules in their excited triplet states in zero field by monitoring the microwave-induced modulation of the phosphorescence. The technique is capable of measuring as few as 10^4 excited-state spins and should be applicable to a wide variety of problems.

While it is generally recognized[1] that an ensemble of two-level systems coupled to a resonant oscillating electric or magnetic field is, in principle, capable of exhibiting free precession and echoes, it is not always apparent precisely how the coherence of the phenomena is manifested in the experimental observables. In magnetic resonance and more recently in optical spectroscopy, spin echoes[2] and their optical analogs, photon echoes,[3] have been observed and explained in terms of dephasing and rephasing of the magnetization of the spins or of the phase of the oscillating electric dipole associated with the electronic transition moment connecting the ground and excited states. On the other hand, when optical properties such as emission or absorption of light are used to monitor magnetic properties such as in optically detected magnetic resonance,[4] the coherent phenomena in many cases are not directly observable via a modulation of the emission or absorption of light at the Larmor frequency of the spins. In phosphorescent triplet states, or, for that matter, in any state where the emission from the individual spin states is unresolved optically, it has been shown[5] that modulation of the light at the Larmor frequency is unobservable unless the electric dipole transition from each of the individual spin levels is of the same polarization. In many excited triplet states, however, the spin sublevels of molecules have different spin-orbit symmetries[6] and, thus, direct detection of the Larmor term is usually forbidden. The result is that only rotary precession has been observed[7] optically, while coherent phenom-