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Observation of Fine-Structure Quantum Beats Following Stepwise Excitation in Sodium *D* States

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We report the observation of fine-structure and Zeeman quantum beats in highly excited sodium *D* states prepared, for the first time in a quantum-beat experiment, by stepwise excitation using two synchronously pulsed dye lasers. We have used this new technique to measure the previously unknown fine-structure intervals of levels $9D$ and $10D$ of sodium, the values being, respectively, 124.5 ± 1.5 and 91.5 ± 1 MHz.

Quantum-beat spectroscopy (QBS) using pulsed dye lasers[1] has proven to be a useful technique in various experiments. Measurements of Zeeman splittings in atoms[2] or molecules[3] and of hyperfine intervals[4] have been recently performed by using QBS. The method consists of preparing by a short light pulse a coherent superposition of the states of interest and observing the subsequent spontaneous emission which exhibits modulations at the Bohr frequencies corresponding to level separations. The exciting light pulse and the detected fluorescence have to be properly polarized and the laser-pulse duration must be smaller than the period of the expected beats.

If a single laser pulse is used to prepare the excited states, the method is obviously restricted to the investigation of those levels which can be optically reached from the ground state (namely *P* states in alkali atoms). In order to extend QBS to the study of optically inaccessible states (e.g., *S* and *D* states), we have used two successive laser pulses tuned at the frequencies of two transitions sharing an intermediate common level (stepwise excitation). By this method, which has already been developed in other types of experiments with spectral lamps[5] and continuous[6] or pulsed[7] lasers, one efficiently prepares a large fraction of atoms in highly excited states of the same parity as the ground state. By monitoring the light emitted by the sodium *D* state produced in this way, we have been able to detect quantum beats either in the presence of a magnetic field (Zeeman beats), or in zero magnetic field where the beats arise from the fine-structure splitting of the *D* state into $D_{5/2}$ and $D_{3/2}$ components.[8] In this way, we have measured the previously unknown fine-structure intervals of the $9D$ and $10D$ levels. This, to our knowledge, is the first time that fine-structure quantum beats are observed following laser-pulse excitation.

Figure 1(a) shows the excitation scheme of Na *D* states. A first pulse, tuned at the wavelength $\lambda_1 = 5890$ Å of the $3S_{1/2}$-$3P_{3/2}$ transition (solid arrow), is followed by a second pulse tuned at the wavelength λ_2 of the $3P_{3/2}$-nD transition (dashed arrow). This second pulse excites in a linear superposition of states the fine-structure and the Zeeman components of the nD level. The light emitted subsequently at the frequency of the nD-$3P$ transition (wavy line) exhibits the modulations of interest. The experimental setup is sketched

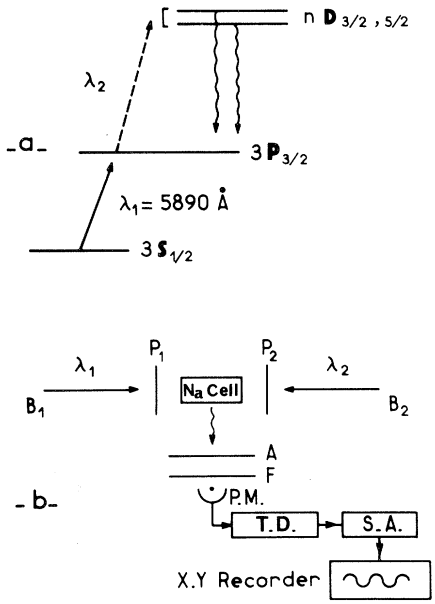

FIG. 1. (a) Level diagram of Na showing the stepwise excitation of nD states. Full line, first excitation; dashed line, second excitation; wavy line, subsequently detected fluorescence. (b) Diagram of QBS apparatus. $P_{1,2}$, polarizers of beams $B_{1,2}$; A (detection analyzer) and F (filter) in front of P.M. (photomultiplier); T.D. and S.A., transient digitizer and signal averager, respectively.

in Fig. 1(b). The two beams B_1 and B_2 are produced by two pulsed dye lasers,[9] pumped by the same nitrogen laser (Molectron UV 1000), which insures a good synchronization of the two successive excitations. The two pulses (3 to 4 nsec long) propagate in opposite directions through the Pyrex resonant cell, whose cold point is regulated at about 130°C. The second pulse is delayed by about 2 nsec with respect to the first one, and thus excites the atoms from the level $3P_{3/2}$ before they decay back to the ground states (the lifetime of the $3P_{3/2}$ level is 16 nsec.) The two beams are tuned to the resonant frequencies with the help of a high-resolution grating monochromator (Jobin-Yvon THR2). For fine tuning of the first beam, one monitors the yellow fluorescence from the $3P_{3/2}$ state at 5890 Å. This wavelength is then filtered out with the help of a Wratten filter F which eliminates also all the uv light components emitted by higher nP levels in the cascades originating from the nD states. The photomultiplier (RCA 1P28) which collects the photons emitted at right angles to the exciting beams is thus only sensitive to the light emitted at the nD-$3P$ transition frequency which exhibits the quantum-beat modulations.

The amplitude and the phase of the beats depend strongly on the polarizations of the exciting pulses ($\vec{e}_1$, $\vec{e}_2$) and detected fluorescence ($\vec{e}_d$). Thus, two polarizers P_1 and P_2 and an analyzer A are placed in the beam paths and before the photomultiplier to select these respective polarizations. The beats in the current of the photomultiplier have been detected by two different techniques. In a first stage of the experiment, we have fed the photomultiplier signal into a Tektronix-661 sampling scope and averaged the output of this scope with a signal averager, according to the technique already described in Ref. 4. Although we have observed Zeeman beats in this way, the technique has proven to be difficult. The pulse-to-pulse instabilities of the dye lasers gave rise to fluctuations in the signal which resulted in a large amount of noise on the sampled trace. Since 200 pulses had to be used to reconstruct a single trace, the recording and averaging of one beat pattern took a long time and the signal-to-noise ratio remained moderate. In a second stage of the experiment, we utilized the newly marketed Tektronix transient digitizer R 7912 which enabled us to store and to analyze the beat patterns in real time. All the noise inherent in the sampling technique was then suppressed and a transient was recorded in one pulse instead of 200, thereby leading to a considerable gain in time. Some noise, coming partly from the detection inself and partly from the nitrogen laser electrical radiation, was still present in the trace, so that one had to average the output of the transient digitizer, which was interfaced to our signal averager. At a rate of 5 pulses/sec, 1000 runs were obtained in a little more than 3 min.

Using this detection procedure, we have looked for zero-magnetic-field fine-structure quantum beats in levels $7D$ to $10D$ of Na (whose lifetimes are in the range of several hundred nanoseconds[10]). Two different polarization schemes have been used. Calculation shows[11] that the beat signal is optimum when the two exciting pulses are polarized at right angles. We have thus set P_1 and P_2 so that $\vec{e}_1$ and $\vec{e}_2$ are perpendicular. We then detected the light with $\vec{e}_d$ either parallel to $\vec{e}_1$ (case a) or to $\vec{e}_2$ (case b). Figure 2 shows the signal observed under these conditions in the $9D$ level. One observes in traces a and b two beat patterns with opposite phases, as expected from the theory.[11] The signal should be purely sinusoidal

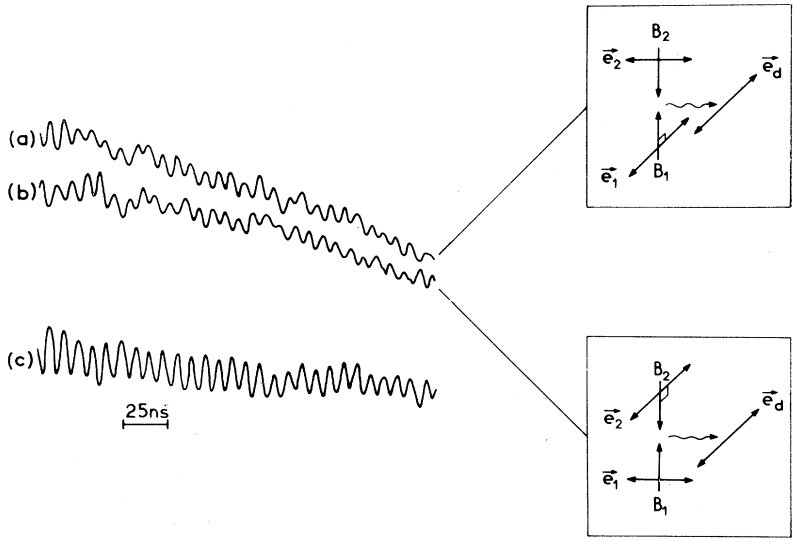

FIG. 2. Recording of fine-structure beats in $9D$ level (averaging of 1000 runs). Trace a: signal obtained with configuration of polarizers $\vec{e}_1 \perp \vec{e}_2$, $\vec{e}_d \parallel \vec{e}_1$ as shown in inset; trace b: signal obtained with $\vec{e}_1 \perp \vec{e}_2$, $\vec{e}_d \parallel \vec{e}_2$ as shown in inset; trace c: result of subtracting trace b from trace a.

since there is a single Bohr frequency in the studied structure (namely, the $9D_{5/2}$-$9D_{3/2}$ interval). However, some systematic noise beats against the signal, so that the trace is slightly distorted. By subtracting trace b from trace a, one gets trace c, in which the noise has been almost entirely eliminated and which exhibits a rather pure sinusoid. The measurement of the frequency of this sinusoid yields the fine-structure interval which we found to be 124.5 ± 1.5 MHz, corresponding to a fine-structure constant $A_{9D} = 49.8 \pm 0.6$ MHz.

We have also observed quantum beats under the same conditions in the $10D$ level. A similar analysis for this level gives an interval of 91.5 ± 1 MHz and $A_{10D} = 36.6 \pm 0.4$ MHz. However, we have not been able to observe the corresponding beats in levels $7D$ and $8D$. The laser-pulse duration (~4 nsec) and the rise time of the photomultiplier (~2 nsec) set up an upper limit of about 150 MHz on the modulation frequencies we were able to detect. It thus seems that the fine-structure intervals of these states are larger than this value. In order to check this hypothesis and to rule out the possibility of a very small fine structure in these states (smaller than the natural linewidth for example), we have applied a magnetic field H_0 to the atoms and observed the Zeeman beats corresponding to the coherent excitation of $\Delta m = 2$ magnetic substates. The calculation shows that in the low-field range ($\mu_B H_0 \ll A$ where μ_B is the Bohr magneton and A the fine structure constant), the dominant frequency of the beats must be equal to $2g_J \mu_B H_0/\hbar$, where $g_J = 1.2$ is the Landé factor of the $D_{5/2}$ state. In the high-field domain ($\mu_B H_0 \gg A$), the beat pattern is made of a "carrier signal" at the frequency $2\mu_B H_0/\hbar$ modulated at a low frequency corresponding to the fine-structure splitting. We have checked that in fields up to 20 G the frequency of the Zeeman beats is equal to $2g_J \mu_B H_0/\hbar$ with $g_J \approx 1.2$. It thus seems clear that, for H_0 in this range, we remain in the low-field domain and that the fine-structure interval should be rather large in levels $7D$ and $8D$.

With the measurements reported in this Letter, the fine-structure intervals of nD levels of Na are known from $n = 3$ to $n = 10$ (with the exception of $n = 7$ and 8). The $n = 3$ to $n = 6$ determinations have been made long ago by classical interferometry on atomic beams[12] and very recently for $n = 4$[13-15] and $n = 5$[16] by two-photon spectroscopy.

Table I summarizes the values obtained for the fine-structure intervals by various authors. It is well known that the structure is inverted for $n = 3$ to 6 (e.g., $D_{5/2}$ lies above $D_{5/2}$). Our quantum-beat experiment does not give the sign of fine structure since linear polarizations are used to excite the atoms. If one uses circularly polarized light pulses in a magnetic field corresponding to the intermediate decoupling range ($A \approx \mu_B H_0$), calculation shows[11] that the beat pattern must be sensitive to the sign of A. Experiments to detect the beats under these conditions are in progress

TABLE I. Summary of measurements of fine-structure interval in D states of sodium.

n	Fine structure interval (MHz)	Sign	Reference
3	1482(50)	−	12
4	1038(50)	−	12
	1025(6)		13
	1035(10)		14
	1028.5(3)		15
5	690(50)	−	12
	618(12)		16
6	372(50)	−	12
7	>150		This work
8	>150		This work
9	124.5(1.5)		This work
10	91.5(1)		This work

and should unambiguously yield the sign of A in levels $9D$ and $10D$. At the moment however, it is reasonable to suppose that this structure is inverted as in the lower levels. An inspection of the figures in Table I shows that a reversal of the sign of A between levels $6D$ and $9D$ would imply a very small absolute value for the structure in levels $7D$ or $8D$, which is not in accordance with our present observations.

Whether the fine structure in the D series of Na is always negative or changes sign for highly excited states remains an open question which we hope to resolve in the near future with QBS experiments performed on higher D levels.

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[10]In the course of our QBS measurements, we have made preliminary determinations of these lifetimes which, to our knowledge, have not been previously published.

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