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Hyperfine Quantum Beats Observed in Cs Vapor under Pulsed Dye Laser Excitation*

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We report observation of hyperfine quantum beats in the light emitted by the $7^2P_{3/2}$ level of Cs^{133} after excitation by a short resonant dye laser pulse. Frequency analysis of the beats provides a measurement of the hyperfine splittings in agreement with previous work. The usefulness of quantum-beat detection as a new method of laser spectroscopy without complications from the Doppler effect is discussed.

The fluorescent light emitted by an atom prepared in a coherent superposition of states by a pulse excitation may exhibit modulations at frequencies corresponding to the energy splittings between these states.[1] These modulations, called quantum beats, have been detected in many experiments over the last ten years, the pulse excitation being provided either by an optical pulse,[2] a pulsed electron beam,[3] or the collision of an atomic beam with a thin carbon target (beam-foil spectroscopy).[4,5] In this latter type of experiment, high time resolution can be achieved by observing the spatial modulation of the fluorescence along the path of the atoms. As a result, beats at several hundred megahertz arising from the coherent excitation of fine[4] and hyperfine[5] structure levels in various atomic species have been recently studied.

The optical-pulse quantum-beat experiments[2] have so far been unable to detect such high-frequency modulations. In these experiments, the optical pulses, produced by conventional spectral sources shuttered by a Kerr cell, were rather long (a few hundred nanoseconds). Therefore, all observations have been restricted to the study of very low-frequency (a few megahertz) beat signals, arising from the coherent excitation of Zeeman substates whose degeneracy was removed by an external magnetic field. Furthermore, due to the weakness of the signal, long averaging techniques had to be used to extract the modulation from the noise.

The advent of widely tunable lasers[6] renews the interest of such experiments. It has recently been shown[7] that these lasers may be successfully used to detect quantum beats corresponding to weak-field Zeeman splittings, with a signal-to-noise ratio considerably improved over previous experiments. It then seemed interesting to extend the same technique to the study of modulated fluorescence at the much higher frequencies corresponding to hyperfine structure splittings. In this paper, we report the first observation, to our knowledge, of such optically excited hyperfine quantum beats.[8] We have studied the light emitted by the $7^2P_{3/2}$ level of Cs^{133} after excitation by a short laser pulse.

We show in Fig. 1 the level diagram of the Cs $6^2S_{1/2}$ and $7^2P_{3/2}$ levels with their hyperfine structure (optical transition at 4555 Å). The ground-state splitting Δg is resolvable by classical spectroscopic methods, while the excited-state splittings $\nu_{54} = 82.9$ MHz, $\nu_{43} = 66.5$ MHz, and $\nu_{32} = 49.9$ MHz, are much smaller than the Doppler width (about 750 MHz), and have been measured using double-resonance or level-crossing techniques.[9] Let us suppose that the atom is excited to the $7^2P_{3/2}$ state by a short light pulse whose duration τ and spectral bandwidth Δ obey the following compatible conditions:

$$1/\tau > \nu_{52} \quad (\nu_{52} = \nu_{54} + \nu_{43} + \nu_{32}), \qquad (1)$$

$$\Delta < \Delta g; \qquad (2)$$

and let us call (a) and (b) the sets of transitions starting respectively from the hyperfine levels
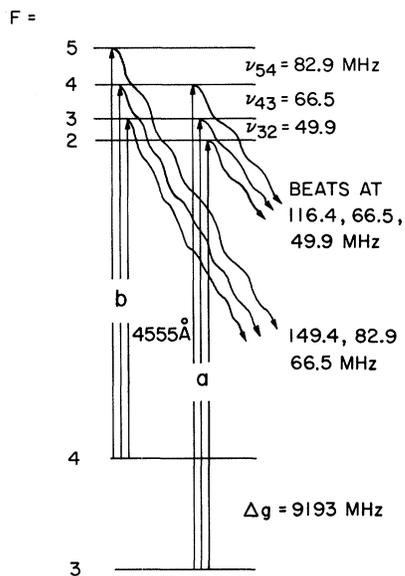
FIG. 1. Hyperfine structure of the $6^2S_{1/2}$ and $7^2P_{3/2}$ levels of cesium. Transitions starting, respectively, from levels $F = 3$ and $F = 4$ in the ground state are labeled by letters (a) and (b). The sets of quantum-beat frequencies expected in each case (a) and (b) are indicated.

$F = 3$ and $F = 4$ in ground state (see Fig. 1). Condition (2) ensures that transitions (a) and (b) are well separated, the light pulse irradiation providing a selective excitation from one of the ground-state levels. However, condition (1) implies that the excited-state structure is not resolved, so that the atoms can be prepared in a coherent superposition of excited levels with different energies. More precisely, in each case (a) and (b), the atoms are excited to a superposition of the three hyperfine states which, according to the selection rule $\Delta F = 0, \pm 1$, are coupled to the relevant hyperfine ground state. After this excitation process, which, according to (1), may be considered as instantaneous, the hyperfine coherence starts evolving at the three eigenfrequencies corresponding to the mutual energy splitting between the excited levels, and is damped at a rate Γ corresponding to the spontaneous emission lifetime $(1/\Gamma \approx 135$ nsec$)$.[10] Consequently, the intensity of the fluorescent light emitted by the atom must exhibit the same temporal behavior. We indicate in Fig. 1 the two different sets of light modulation frequencies which are expected in cases (a) and (b).

These modulations may be observed in every re-emission channel starting from the $7^2P_{3/2}$
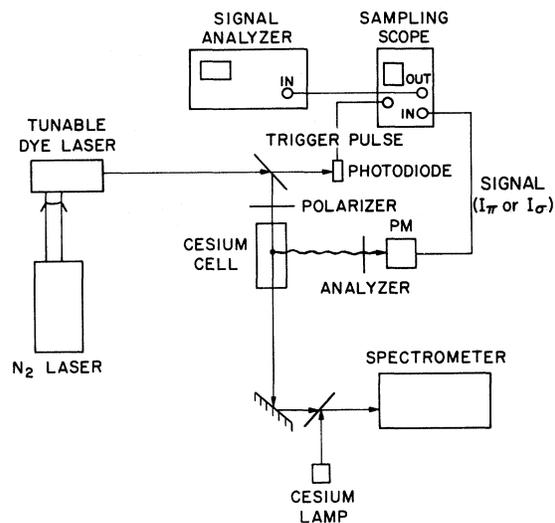
FIG. 2. Experimental setup for quantum-beat observation and analysis.

state (transitions back to the levels $6^2S_{1/2}$, $7^2S_{1/2}$, $5^2D_{3/2}$, and $5^2D_{5/2}$). The intensity $I(t)$ of the light emitted in any of these channels is readily calculable using the formalisms of Ref. 1. The signal obtained depends not only on the hyperfine state F from which the excitation started [case (a) or (b)], but also on the respective polarizations $\vec{\epsilon}$ and $\vec{\epsilon}'$ of the incident and fluorescent light. Let us call, respectively, $I_\pi(t)$ and $I_\sigma(t)$ the signals corresponding to $\vec{\epsilon}$ and $\vec{\epsilon}'$ parallel and perpendicular. It may be shown quite generally that the modulated parts of I_π and I_σ are in phase opposition, the modulation in I_π being twice as big as the one in I_σ.

The above discussion dictates the experimental conditions which have to be fulfilled for the study of these hyperfine beats. The setup is shown in Fig. 2. The optical source is a N_2 laser-pumped tunable dye laser[6] which delivers repetitive light pulses whose duration (~ 2 nsec) and bandwidth (~ 1 GHz) satisfy conditions (1) and (2). The laser is tuned to one of the two resonances (a) or (b) by comparing, with a spectrometer, its spectrum with that of a Cs lamp. The laser radiation is linearly polarized and sent through a Cs cell, temperature regulated at about 70°C. The fluorescent light emitted at right angles is detected by a photomultiplier (RCA 1P28) which is sensitive only to the transition back to the ground state (4555 Å). An analyzer allows the detection of either the I_π or I_σ signals. The repetitive output of the photomultiplier is sent to a sampling scope (Tektronix 661) which allows the reconstruction
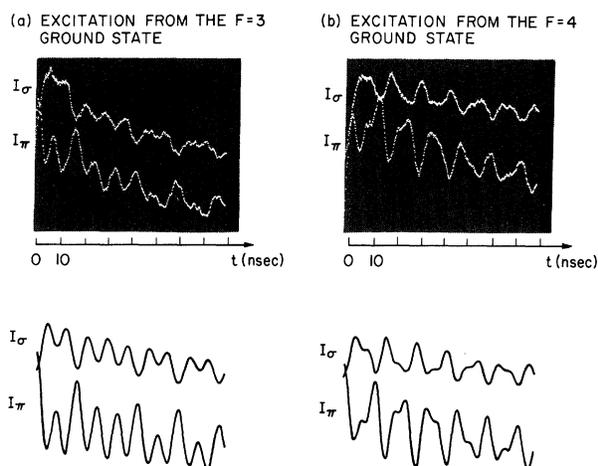
FIG. 3. Pictures of the quantum-beat signals in cases (a) and (b). The lower and upper trace in each picture correspond, respectively, to I_π and I_σ. Each trace contains 500 sampling points and is averaged over 256 runs. The base line has been offset between the recordings of I_π and I_σ to avoid overlapping of the traces. The corresponding theoretical plots of the beats are shown under each picture, with the same time scale.

FIG. 4. Time recording and frequency analysis of the $I_\pi - I_\sigma$ signal in cases (a) and (b). Signals I_π and I_σ have been recorded separately (500 sampling points; 256 runs) and then subtracted. The frequency interval between points in the spectra $S_a(\nu)$ and $S_b(\nu)$ is 2.34(2) MHz.

of the signal on a long-term basis, approximately 10 sec instead of a few hundred nanoseconds real time, enabling us to store and process the data. The sampling technique introduces, however, noise due to the pulse-to-pulse amplitude fluctuation of the dye laser. A Hewlett-Packard 5480A signal analyzer is used to enhance the signal-to-noise ratio by signal averaging methods. Figure 3 shows the I_π and I_σ beat signals corresponding to resonances (a) and (b). One can compare them to the corresponding theoretical plots shown under each picture. The patterns of the beats in the different signals correspond very well to the theoretical predictions. In particular, the I_π and I_σ signals are always in phase opposition, as predicted by the theory. In order to enhance the modulation depth, we have thus been led to subtract the I_π and I_σ curves. This is done using a Hewlett-Packard 2115A computer interfaced to the signal analyzer. Figure 4 shows the $I_\pi - I_\sigma$ beat signals in cases (a) and (b), recorded on a time scale longer than that used in the previous pictures. Using the computer, we have performed a fast Fourier-transform analysis of $I_\pi - I_\sigma$, the typical power spectra $S_a(\nu)$ and $S_b(\nu)$ being shown opposite to the time-evolution curves in Fig. 4. The zero frequency peaks correspond to the unmodulated part of the fluorescent decay. The three successive

peaks, indicated by arrows, correspond to the three eigenfrequencies in each beat signal. From $S_a(\nu)$, we find the hyperfine intervals to be $\nu_{43} = 65.5 \pm 1.5$ MHz, $\nu_{32} = 49.1 \pm 1.5$ MHz, and $\nu_{42} = 114.6 \pm 2$ MHz; from $S_b(\nu)$, $\nu_{54} = 81.9 \pm 2$ MHz, $\nu_{43} = 66.7 \pm 1.5$ MHz, and $\nu_{53} = 148.6 \pm 2$ MHz. These values are in agreement with those obtained in previous works[9] (compared with the expected frequencies indicated in Fig. 1). The quantum-beat technique is so far less accurate, but its precision could be improved by sampling more points and recording the beats on a longer time scale.

This experiment shows that the quantum-beat technique using pulsed lasers can be used practically as a new method in optical spectroscopy without complications from the Doppler effect. As such it can be compared with double-resonance,[11] level-crossing,[12] and saturation spectroscopy.[13] The attractive feature of this technique is that the atoms, selectively excited in the levels of interest, provide, without being perturbed, their own spectrum; no search or scan of the line is necessary. Clearly, this method should be a useful tool for a first determination of still unknown hyperfine structures (for example, in high-lying states of alkalis).

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Resonant Raman Scattering from Iodine: Spectral Changes with Excitation Frequency

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We have recorded high-resolution resonant Raman spectra of molecular iodine which show very pronounced systematic spectral band-shape variations with incident laser frequency. This frequency dependence and the presence of the multiple overtones in resonant Raman scattering from iodine may be explained by a theory in which the scattering intensity depends on Franck-Condon overlap integrals. We have obtained semi-quantitative agreement between the observed spectra and spectra calculated from this theory.

Reports of previous Raman studies on molecular iodine have discussed light scattering below[1] and above[2,3] the $B(^3\Pi_{0^+u})$-state dissociation limit (20162 cm^{-1}). Resonant Raman scattering above the dissociation limit[2,3] is characterized by several overtones, each exhibiting a complex structure. The details of this structure were attributed to O, Q, and S ground-electronic-state $X(^1\Sigma_{0^+g})$ vibrational-rotational branches of several thermally populated hot bands.[3] Although it has been shown[3] that frequencies of the discrete features of the spectra result from bandheads of S and Q branches (the O branch results only in broad structureless scattering[3]), no explanation has been given for the relative intensities of these features. Furthermore, prior investigations of scattering above the B-state dissociation limit

have reported only on spectra obtained with 4880-Å excitation and no mention was made of structural changes occurring with different laser frequencies. In this Letter we wish to report very marked changes in the spectrum as a function of the incident laser frequency above the B-state dissociation limit. We attribute these changes to a variation in the magnitude of the Franck-Condon overlap integral and discuss a theory from which we have calculated spectra. This calculation both explains the occurrence of the multiple overtones in resonant Raman scattering from molecular iodine and accounts for the structural changes with incident laser frequency.

The Raman data reported here were obtained in a standard manner. The gaseous iodine in a sample cell was held at a pressure of about 25