

Excited state hyperfine structures in ^{133}Cs using quantum beat spectroscopy

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Quantum beats arising from the hyperfine interaction were observed in the fluorescence produced when the $8D_{3/2}$, $9D_{3/2}$, and $10D_{3/2}$ states of ^{133}Cs radiatively decayed to the $6P_{1/2}$ state. The period of the beats equals the reciprocal of the magnetic dipole coupling constant a since the ^{133}Cs nucleus has a negligibly small electric quadrupole moment. The results are $a = 3.95 \pm 0.01$, 2.38 ± 0.01 , and 1.54 ± 0.02 MHz for the $8D_{3/2}$, $9D_{3/2}$, and $10D_{3/2}$ states, respectively.

Des battements quantiques dus à l'interaction hyperfine ont été observés dans la fluorescence produite lors de la désexcitation des états $8D_{3/2}$, $9D_{3/2}$ et $10D_{3/2}$ de ^{133}Cs vers l'état $6P_{1/2}$. La période des battements est égale à l'inverse de la constante de couplage de dipole magnétique a , étant donné que le noyau ^{133}Cs a un moment de quadrupole électrique négligeable. Les résultats sont: $a = 3,95 \pm 0,01$, $2,38 \pm 0,01$ et $1,54 \pm 0,02$ MHz pour les états $8D_{3/2}$, $9D_{3/2}$ et $10D_{3/2}$, respectivement.

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1. Introduction

The measurement of energies with a sub-Doppler resolution is important for testing atomic theory. The alkali atoms are among the easiest atoms to model because they have only a single valence electron. Alkali elements such as cesium are used in a very wide range of applications including laser cooling (1), atomic clocks (2), and tests of parity violation (3). It is therefore essential that the atomic structure of cesium be well understood. Accurate measurements of hyperfine constants have revealed effects that cannot be explained by a simple hydrogenic picture of the alkali atoms (4–6). These effects are caused by the polarization of the inner shell electrons, relativistic corrections, and the magnetic and electric nuclear moments.

The hyperfine structure of the excited $nD_{3/2}$ states in ^{133}Cs has been studied using various methods including level crossing spectroscopy (6, 7), optical double resonance (8), and magnetic field decoupling (9). The most accurate measurements to date have been obtained by Deech *et al.* (10, 11) using quantum beat spectroscopy. We used the same method to determine the magnetic dipole constant of the $8D_{3/2}$, $9D_{3/2}$, and $10D_{3/2}$ states of ^{133}Cs shown in Fig. 1. The apparatus is much simpler than that used by Deech *et al.* and yields values of the magnetic dipole constant that are the most accurate yet reported. For the $8D_{3/2}$ state, the uncertainty in the magnetic dipole constant has been reduced by a factor of seven. Finally, the results are compared with calculated values.

2. Theory

The cesium atom can be modelled as a hydrogenic system using the following Hamiltonian.

$$[1] \quad H = H_0 + ah \mathbf{I} \cdot \mathbf{J}$$

H_0 represents the Coulomb and fine structure. The next term is the magnetic dipole hyperfine interaction whose magnitude depends on the coupling constant a . The electric quadrupole hyperfine interaction has been neglected since the electric quadrupole moment of ^{133}Cs is only -3 mb (12). \mathbf{I} is the nuclear spin and \mathbf{J} is the angular momentum of the outer electron. The spin of the ^{133}Cs nucleus is $7/2$.

To generate quantum beats, the atoms must be prepared in a coherent superposition of eigenstates. In our experiment, a pulsed laser excited the ground state. The excited state consists

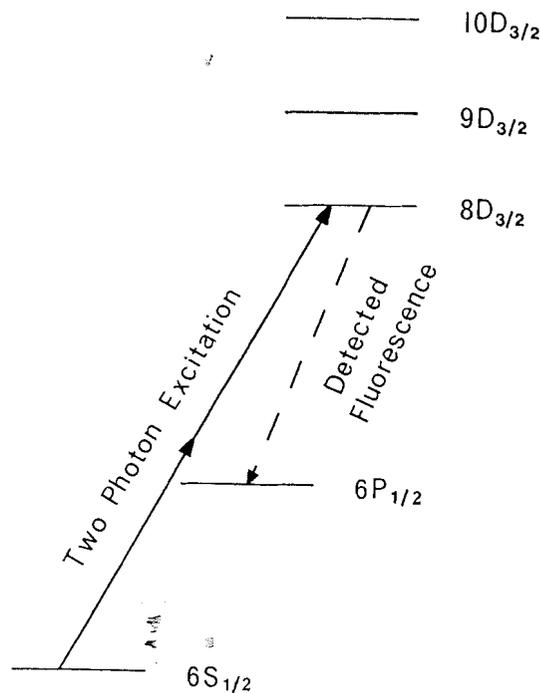


FIG. 1. Cesium energy levels accessed by the experiment. The various laser excitation and fluorescence wavelengths are listed in Table 1.

of a linear combination of hyperfine eigenstates if the laser line-width exceeds the hyperfine splitting. The excited state wavefunction then evolves in time according to Schrödinger's equation. Each hyperfine component of the wavefunction is multiplied by an exponential decaying factor and a complex phase factor whose exponent equals the product of the eigenfrequency and the time. Hence, temporal oscillations or quantum beats appear in the fluorescent intensity when the excited state radiatively decays. Optimum beat signals are obtained when the beat period is much greater than the duration of the exciting laser pulse and less than the fluorescent lifetime of the excited state.

Detailed reviews of quantum beats have been written by Dodd and Series (13) and by Haroche (14). They derive expressions

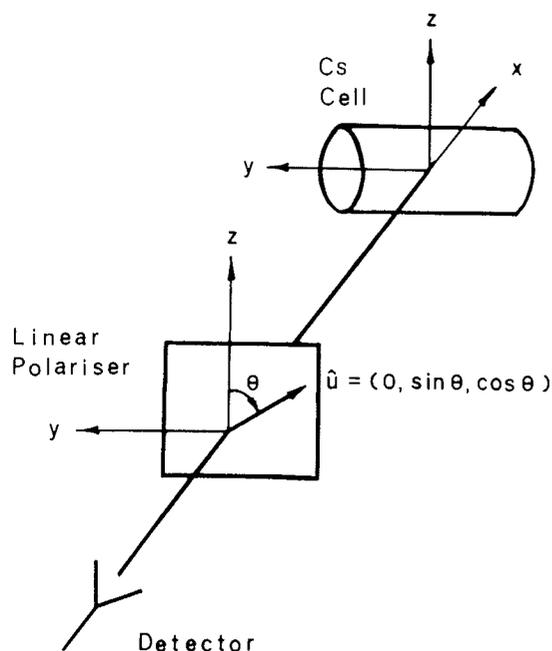


FIG. 2. Detection of linearly polarized fluorescence. Fluorescence is observed transverse to the laser propagation (\hat{y}) and polarization (\hat{z}) directions. The linear polarizer transmission axis is oriented along \hat{u} .

TABLE 1. Quantum beat frequencies

F	F'	$\omega_{FF'}/2\pi$
5	4	$5a$
5	3	$9a$
4	3	$4a$
4	2	$7a$
3	2	$3a$

for the fluorescent intensity at time t emitted by atoms excited at time $t = 0$. In our experiment, fluorescence produced by the decay of the $nD_{3/2}$ state to the $6P_{1/2}$ state was measured and is given by the following theoretical expression (13, 14).

$$[2] \quad I(t) = I_0 e^{-t/\tau} \left[1 + P_2(\cos \theta) \sum_{FF'} a_{FF'} \cos \omega_{FF'} t \right]$$

I_0 is a constant that includes factors such as the excited state number density, the finite solid angle of the detector, and the transmission efficiency of various optical filters and polarizers. τ is the radiative lifetime of the $nD_{3/2}$ state. $P_2(\cos \theta)$ is the second order Legendre polynomial, where θ is the angle between the z direction and the polarization axis of the detected fluorescence as shown in Fig. 2. $P_2(\cos \theta)$ equals 1, 0, and $-1/2$ when θ equals 0° , 54.7° , and 90° , respectively. The quantum beats occur at frequencies $\omega_{FF'}$, which are listed in Table 1, and with amplitude $a_{FF'}$. F is the magnitude of the total angular momentum, which equals the sum of the electronic and nuclear angular momenta. The modulation frequencies equal integral multiples of the magnetic dipole constant a , and are independent of factors such as the laser pulsewidth and linewidth that affect the $a_{FF'}$ coefficients. Hence, the magnetic dipole constant equals the reciprocal of the recurrence period of the beat pattern.

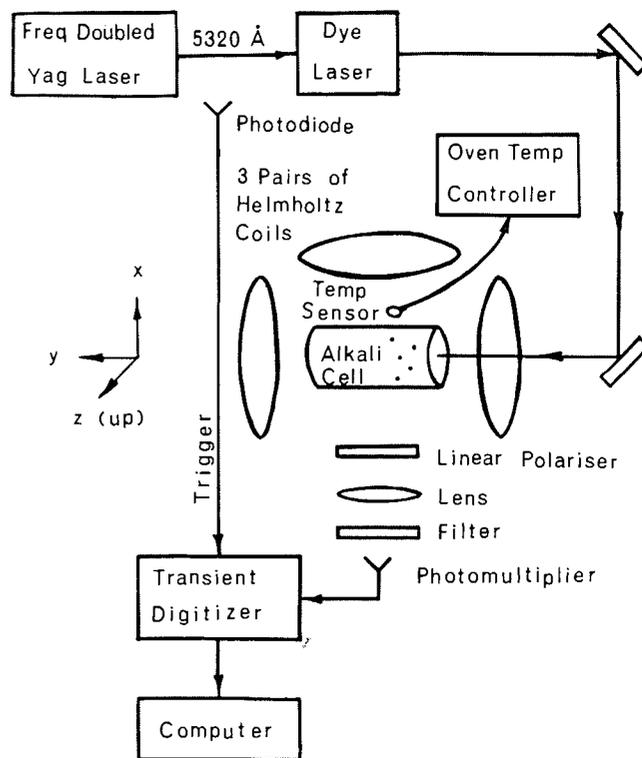


FIG. 3. Apparatus.

The hyperfine constant can also be determined by fitting [2] to the observed data. However, this requires knowing the beat amplitudes $a_{FF'}$. One can also find the beat frequencies by taking the Fourier transform of the experimental signal. These methods were not used since they are significantly more complex and yield less accurate results.

3. Experiment

3.1. Apparatus and procedure

The apparatus is shown in Fig. 3. A dye laser was pumped at a 10-Hz repetition rate by a frequency doubled Nd:YAG laser. The laser was linearly polarized along the vertical direction z . The laser wavelengths used to excite the cesium $6S_{1/2}$ ground state to the $nD_{3/2}$ via a two-photon excitation are listed in Table 2. The hyperfine splitting of the ^{133}Cs ground state is 0.3 cm^{-1} , while the laser linewidth is quoted to be 0.07 cm^{-1} by the manufacturer. The laser wavelength was tuned to maximize the amplitude of the observed quantum beats. The temporal profile of the laser pulse was measured using a fast photodiode and found to be closely approximated by a Gaussian function having a FWHM of only 7 ns.

The cesium atoms are contained in a cylindrical Pyrex cell having a diameter of 1 in. and a length of 10 in. (1 in. = 2.54 cm). Before the cell was filled with ^{133}Cs , it was simultaneously evacuated by a diffusion pump to a pressure of 1×10^{-7} Torr (1 Torr = 133.3 Pa) and baked overnight at several hundred degrees centigrade to remove impurities. The cell was located in an oven heated by jets of hot air. A feedback circuit stabilized the temperature to $\pm 0.1^\circ\text{C}$. The entire oven containing the cell was surrounded by three pairs of Helmholtz coils, which were used to cancel the Earth's field. The residual field was measured using a Hall Effect gaussmeter to be less than 10 mG.

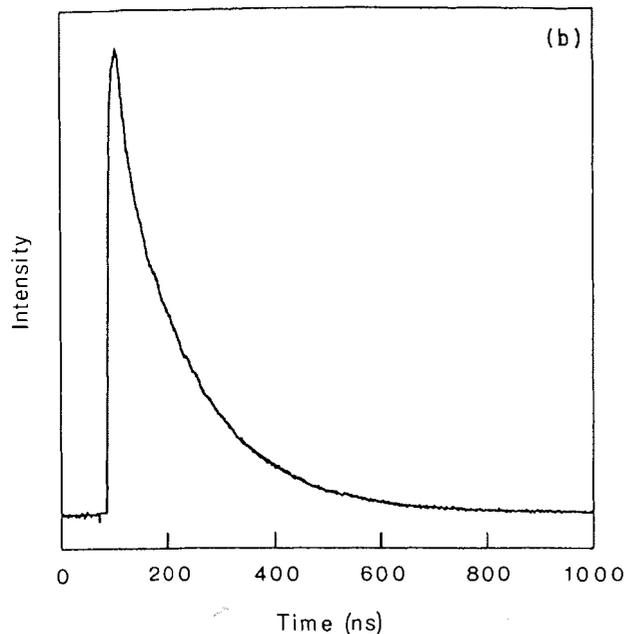
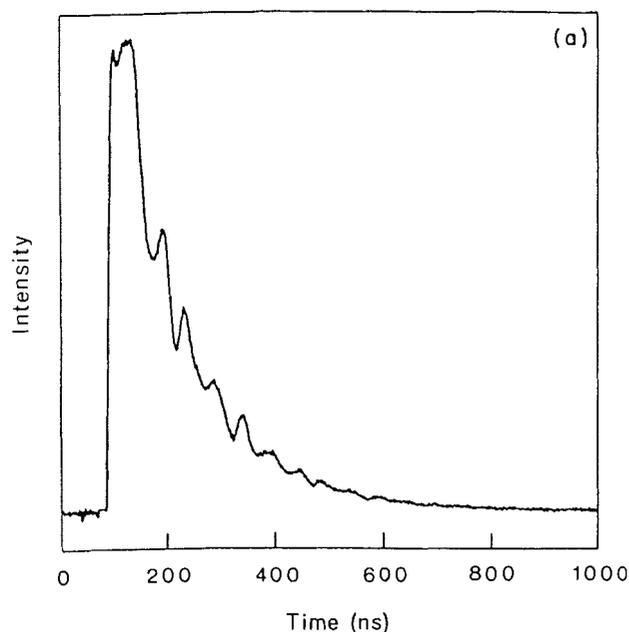


FIG. 4. Sample data. (a) These data were taken while a vertical linear polarizer was in front of the detector. (b) Signal when $\theta = 54.7^\circ$.

Fluorescent light was detected in the direction transverse to both the laser propagation and polarization directions, as shown in Fig. 3. The detected light first passed through a linear polarizer, oriented as shown in Fig. 2. The light was next collimated by a lens onto an interference filter. The filter transmits light emitted when the $nD_{3/2}$ state radiatively decays to the $6P_{1/2}$ state. The fluorescence wavelengths are listed in Table 2. Scattered laser light was blocked since the FWHM bandwidth of the filter was only 10 \AA ($1 \text{ \AA} = 10^{-10} \text{ m}$). Finally, the light was focussed onto a photomultiplier (Hamamatsu model R928), which has a manufacturer quoted risetime of 2.2 ns. The photomultiplier was contained in a mumetal housing to shield it

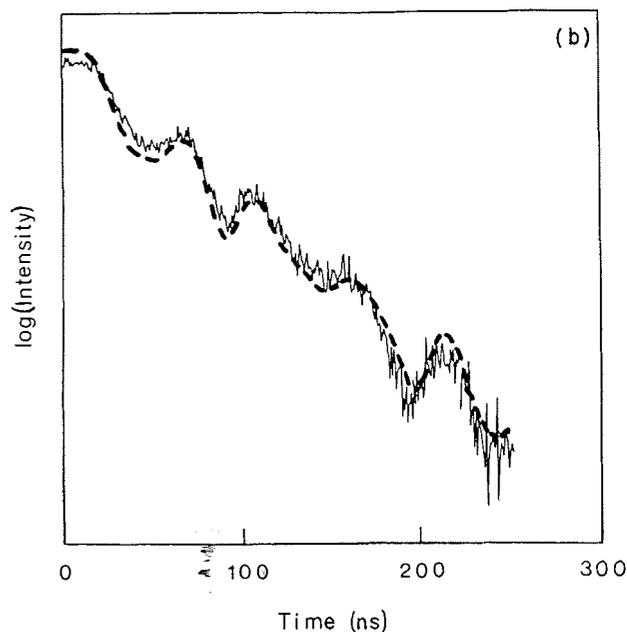
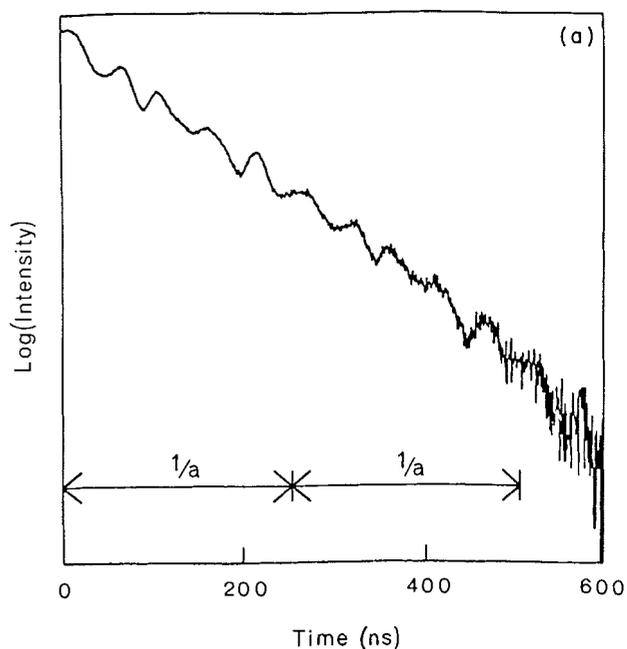


FIG. 5. (a) The logarithm of the data shown in Fig. 4a is plotted to more clearly show the quantum beats. (b) The first two periods of the beat pattern shown in part (a) have been superimposed.

from any external magnetic fields. It was operated at sufficiently low voltages to ensure that the output current was linearly proportional to the incoming light intensity. This was checked using a calibrated neutral density filter. Dark current and other sources of background noise were completely negligible. The photomultiplier signal was sent to a transient digitizer (LeCroy Waveform Digitizer 6880A), which was triggered by a fast photodiode that detects the output of the YAG laser. The digitizer has an analog bandwidth of 400 MHz and digitized the signal every 742 ps. The digitizer timing is accurate to 0.01% according to the manufacturer. The beat period found using the digitizer agreed with that observed on our oscil-

TABLE 2. Experimental results

Excited state	Excitation wavelength (Å)	Fluorescent wavelength (Å)	a (MHz)		
			This work	Other work	Theoretical
$8D_{3/2}$	7191	6012	3.95 ± 0.01	3.92 ± 0.10 (9) 3.92 ± 0.07 (10, 11) 3.98 ± 0.08 (7, 8)	2.085
$9D_{3/2}$	6937	5666	2.38 ± 0.01	2.32 ± 0.04 (10, 11) 2.37 ± 0.03 (7, 8)	1.266
$10D_{3/2}$	6787	5467	1.54 ± 0.02	1.51 ± 0.02 (10, 11) 1.52 ± 0.03 (7, 8)	0.825

loscope (Phillips 3296A). Typically, for a single run, data from 1000 laser pulses were additively accumulated in the digitizer and sent to the computer for analysis.

3.2. Data analysis

Samples of typical data are shown in Fig. 4. These data were taken when the $8D_{3/2}$ state decayed to the $6P_{1/2}$ state. The observed beats were strongly affected by the transmission axis of the linear polarizer in front of the detector as predicted in ref. 2. The beats had the greatest amplitude when vertical polarized light ($\theta = 0^\circ$) was detected, and disappeared when $\theta = 54.7^\circ$. Also, the two signals obtained using vertical and horizontal polarizers were observed to be out of phase.

Figure 5a shows a plot of the logarithm of the intensity versus time. The data show two repetitions of the beat pattern. The repetition time or beat period equals the reciprocal of the magnetic dipole constant a . It was determined as follows. Data collected during the second period were multiplied by a constant scaling factor and superimposed on the data collected during the first beat period as shown in Fig. 5b. The beat period was then equated to the time interval resulting in the optimum overlap of the first two beat patterns. Later data were not considered because they were too noisy.

The resulting values of the magnetic dipole constants are listed in Table 2. The quoted uncertainty is due to the finite time resolution of the transient digitizer. The uncertainty is larger for the $10D_{3/2}$ state, since data encompassing two complete beat periods could not be recorded by the digitizer. The beat periods measured were the same for dozens of different runs. No dependence on cell temperature or laser energy was observed. The cell temperature was varied from 70 to 100°C, corresponding to cesium densities of 10^{12} – 10^{13} atoms cm^{-3} (15). Data were taken with laser pulse energies between 1 and 10 mJ.

4. Discussion of results

The results of this experiment are in excellent agreement with previous measurements, obtained using a variety of different methods. They are more accurate than those of Deech *et al.* who also performed a quantum beats experiment using a photon counting apparatus. Data can be collected much faster and more simply in our experiment, since many fluorescent photons produced by a single exciting laser pulse are detected by a photomultiplier and recorded by a transient digitizer. The time resolution of the digitizer is 742 ps while that of Deech's apparatus is only 10 ns. This permits a more accurate measurement of the beat recurrence time and in turn the magnetic dipole constant.

The interaction of the valence electron of the alkali atom

with the nucleus shielded by the core electrons can be approximated by a hydrogenic potential. The following theoretical expression for the magnetic dipole constant has been derived by Kopfermann (16),

$$[3] \quad a_J = \frac{2\mu_B^2}{h} g_J \frac{L(L+1)}{J(J+1)} F_{Z,J} (1-\delta)(1-\epsilon) \langle r^{-3} \rangle$$

where r is the distance from the nucleus to the valence electron, which has orbital angular momentum L and total angular momentum J . $g_J = \mu_J/\mu_B J$ is the dimensionless ratio of the nuclear magnetic moment divided by the Bohr magneton and the nuclear spin. ^{133}Cs has a magnetic moment of 2.5777 nuclear magnetons. δ and ϵ are small corrections for the finite extent of the charge and magnetic dipole distribution of the nucleus. They were neglected in our calculation of a_J . $F_{Z,J}$ is a relativistic correction factor, which equals 1.066 for ^{133}Cs (16). The expectation value of $\langle r^{-3} \rangle$ is found using the Lande formula (16),

$$[4] \quad \langle r^{-3} \rangle = \frac{Z'}{n^*{}^3 L(L+1) \left(L + \frac{1}{2} \right) a_0^3}$$

where n^* is the effective quantum number and a_0 is the Bohr radius. Z' has been empirically found for D electrons to equal the atomic number Z minus 11 (16, 17). Large discrepancies exist between the calculated and experimental results listed in Table 2, illustrating the need for improved theory (18). A comparison between computed and measured hyperfine constants therefore provides a stringent test of any theoretical model.

Acknowledgements

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