HYPERFINE INTERACTION CONSTANTS IN THE $9\,^2D_{3/2}$ STATE OF ^{85}Rb MEASURED BY THE QUANTUM BEAT METHOD*

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The magnetic-dipole interaction constant A and the electric-quadrupole interaction constant B in the $9\,^2D_{3/2}$ state of ^{85}Rb were measured using the quantum beat method. The results are $|A| = 0.561(11)$ MHz, $|B| = 0.20(3)$ MHz and $B/A > 0$.

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1. Introduction

Recently the results of hyperfine structure (hfs) measurements in several $n\,^2D_{3/2}$ states of ^{85}Rb have been reported. The most impressive ones were obtained for $n = 5$ [1]. The optical frequencies of hyperfine components in the $5\,^2S_{1/2}$–$5\,^2D_{3/2,5/2}$ two-photon transitions of rubidium atoms were determined with an uncertainty of 10^{-11}. From various combinations of these frequencies the magnetic-dipole interaction constant A and electric-quadrupole interaction constant B for both isotopes, ^{85}Rb and ^{87}Rb, were calculated. The sign of the constants was also established. The hyperfine structures for rubidium $5\,^2D_J$ states are now the most accurately determined from among those on the excited states of alkali metal atoms.

The ^{85}Rb states with higher n such as $8\,^2D_{3/2}$ [2] and 10 and $11\,^2D_{3/2}$ [3] were investigated by quantum beat spectroscopy [4, 5]. The rubidium atoms were excited from the ground $5\,^2S_{1/2}$ to $n\,^2D_{3/2}$ state by two-photon absorption of laser light. The time-resolved fluorescence was registered and the observed modulations of the decay were analysed. The absolute values of the constants A and B and their relative sign were obtained. In the present work in an IIIIIexperiment on the $9\,^2D_{3/2}$ state of ^{85}Rb the quantum beat method was also employed. The obtained results fill a gap in hitherto available data.

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2. Experimental

The experimental set-up was the same as used in Ref. [3], as was the excitation and detection scheme. In the inset of Fig. 1 the transitions involved and corresponding wavelengths are shown. The nitrogen-pumped dye laser produced pulses of about 4 ns duration and the intracavity Fabry–Perot etalon narrowed the laser linewidth to about 3 GHz. The fine structure in the $9\,^2D$ state of rubidium is equal to 21 GHz, therefore a selective excitation of the $9\,^2D_{3/2}$ state was easily achieved. The half-wave plate converted the linear polarization of the laser beam from horizontal to vertical. The cylindrical Pyrex cell containing isotopic rubidium was kept at the temperature of 59°C which corresponds to rubidium vapour density of 4×10^{-11} atoms cm^{-3} [6]. Three pairs of orthogonal Helmholtz coils reduced the Earth magnetic field to less than 20 mG.

The fluorescent light was observed perpendicularly to the exciting laser beam and to its polarization vector. Edge and interference filters were used to select the light corresponding to the transitions $9\,^2D_{3/2}$–$5\,^2P_{1/2}$ and $9\,^2D_{3/2}$–$5\,^2P_{3/2}$. The interference filter had maximum transmittance at 518 nm, so the first component of the doublet was mainly transmitted, but a series of measurements with an edge filter alone was also made. The fluorescence components with polarization vector either parallel ($I_\parallel$) or perpendicular ($I_\perp$) to that of the laser light were detected by a cooled photomultiplier (Phillips XP2020).

In time-resolved measurements the photon-counting technique was employed. The electronic equipment consisted of ORTEC modules (including pile-up inspector), analog-to-digital converter (POLON 712) and microcomputer. In the time-to-amplitude converter (TAC) the signal from the fast photodiode detecting the small amount of the laser light served as a "Start", and the signal from the photomultiplier as a "Stop" pulse. The TAC output signal was digitized and stored in the microcomputer working as multichannel analyser. The signal was discarded if following a laser pulse more than one pulse from the photomultiplier appeared. The time scale of multichannel analyser was calibrated using a variable delay generator. The delays were measured with a quartz controlled frequency/interval counter.

3. Results and discussion

The $9\,^2D_{3/2}$ state of ^{85}Rb (nuclear spin $I = 5/2$) is split into four hyperfine substates described by quantum number $F = 4,3,2,1$. Due to the coherent excitation of this hfs manifold with a short laser pulse modulation of the fluorescence appears. The modulation frequencies $\omega_{FF'}$ depend on the hyperfine structure: expressed in terms of the hfs constants A and B they are shown in Table. The corresponding amplitudes $A_{FF'}$ are also given. They were calculated in Ref. [2] for the uniform population of the ground state and for the exciting linewidth which is broad relative to the ground state hyperfine splitting. The relative amplitudes do not depend on which component of the doublet $n\,^2D_{3/2}$–$5\,^2P_{1/2,3/2}$ is considered.

The experimental data for the $9\,^2D_{3/2}$ state were fitted to the unmodulated term $X_0 \exp(-t/\tau_0)$, sum of the five components $\sum_{F>F'} X A_{FF'} \cos\omega_{FF'} \times \exp(-t/\tau_0)$ and the background. The τ_0 is a natural lifetime and $A_{FF'}$ and

TABLE

Quantum beat frequencies expressed in terms of hfs constants and the relative amplitudes $A_{FF'}$, for broad line excitation and for excitation from a single ($f = 2$ or $f = 3$) hyperfine sublevel of the ground state.

FF'	$\omega_{FF'}/2\pi$	$A_{FF'}$	$A^{(2)}_{FF'}$	$A^{(3)}_{FF'}$
43	$4A + 4B/5$	15/8	15/14	45/56
32	$3A - 9B/20$	2/5	8/35	6/35
21	$2A - 4B/5$	7/20	$-1/20$	2/5
42	$7A + 7B/20$	6/7	12/49	30/49
31	$5A - 5B/4$	28/25	4/25	24/25

$\omega_{FF'}$ are given in Table. The possibility of existence of two different time constants for unmodulated and quantum beat terms was also taken into account but the obtained values of A and B were the same as those from the fits with τ_0 only. At rubidium vapour pressure used in the experiment the collisional effects which cause that the population and alignment decay are different were negligible [7]. The multiple time constants were found for data acquired with lower laser power and with the interference filter removed. In this case the unmodulated decay was no longer exponential due to contribution of fluorescence from S and D states populated in cascades. However, the fitted hfs constants remained very consistent with those obtained from the other series of data.

In the described experiment the laser linewidth was not broad compared to the hyperfine splitting of the ground state thus the preferential excitation from one of the two hyperfine levels of the ground state was possible. To take such effect into account the relative amplitudes $A^{(f)}_{FF'}$ of the quantum beats following absorption from a single hfs sublevel f of the ground state were calculated. They are also listed in Table. In the fitting procedure the term $\sum_f \sum_{F>F'} X_f A^{(f)}_{FF'} \cos\omega_{FF'}$ replaced the former sum of modulated components, but again no systematic change in the hyperfine constant values was noticed.

All of the above mentioned alternative fitting procedures were applied to sets of data obtained for two polarized components of fluorescence, $I_\parallel$ and $I_\perp$. The example of experimental results and one of the possible fits are shown in Fig. 1.

The experimental values of hfs constants were found to be: $|A| = 0.561(11)$, $|B| = 0.20(3)$, $B/A > 0$. The errors account for several factors: the spread of the results for various sets of data and the detection of two polarizations, the different fitting procedures, an uncertainty in the origin of the time scale and in the time scale calibration.

The experimental value of the constant A is in good agreement with the values 0.561(3) and 0.553(3). The former was obtained by scaling the result for ^{87}Rb with the relation $A_{85} = A_{87}(\mu_{85}/\mu_{87})(I_{87}/I_{85})$. The value for A_{87} was obtained by Belin et al. [8] by means of level crossing spectroscopy and the magnetic moments μ of rubidium isotopes were taken from Ref. [9]. In the calculation of the second comparison value for A it was assumed that the ratios A_{85}/A_{87} in the $n\,^2D_{3/2}$ states

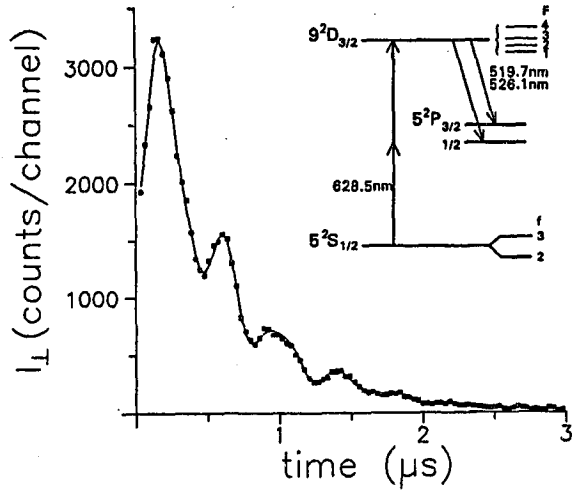

Fig. 1. Example of experimental quantum beat signal (squares) and one of the possible fits. The inset shows the excitation and detection scheme: two-photon absorption and fluorescence to $5\,^2P_{1/2,3/2}$ states. The corresponding wavelengths are also given.

are the same for $n = 9$ and $n = 5$, which assumes the same hyperfine anomaly for both n. The value for B also agrees with the scaled value 0.22(6) calculated from the relation $B_{85} = B_{87}Q_{85}/Q_{87}$, where Q denotes the electric-quadrupole moment [9] and B_{87} is taken from Ref. [8].

The presently determined value for A is less accurate than the scaled ones. The same occurs for the 10 and 11 $^2D_{3/2}$ states when the hfs measurements for both rubidium isotopes were performed by the same method [3, 10]. The quantum beat pattern in ^{85}Rb carries only a few undulations which is contrary to the situation for ^{87}Rb. Each of these undulations consists of five overlapping signals which limits the final accuracy. An improvement in hfs determination in ^{85}Rb could be expected if an additional quantum beat experiment with simultaneous hyperfine pumping was performed and the excitation took place only from the sublevel $f = 2$. In this case one modulation component (ω_{43}) would dominate. Another way of improving the experimental precision would be to use Zeeman optical pumping, as has been proposed in Ref. [11]. For fully oriented ground state (only sublevel (3,+3) or (3,−3) populated) and linear π two-photon excitation, the beat pattern simplifies to only single ω_{43} frequency.

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