Hyperfine-structure measurements in the $4p\ ^2P_{3/2}$ state of ^{41}K using polarization quantum-beat spectroscopy

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A pump-and-probe approach was used to investigate polarization quantum beats resulting from hyperfine interactions in the resonant $4p\ ^2P_{3/2}$ level of ^{41}K. Coherent time evolution of the electronic polarization was observed over eight radiative lifetimes of the $4p\ ^2P_{3/2}$ level. Analytical expressions for the resulting polarization beat signals were fitted to the data to determine the magnetic-dipole constant, $A=3.325(15)$ MHz, and the electric-quadrupole constant, $B=3.230(23)$ MHz. These results represent an improvement of about one order of magnitude over previous measurements.

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I. INTRODUCTION

Interpretation of many spectroscopic experiments depends critically on the availability of reliable atomic excited-state wave-function calculations [1]. Many recent measurements of atomic lifetimes and hyperfine coupling constants have been carried out, in part to test the quality of calculated wave functions. Alkali atoms have had a special place in these studies due to their traditional role as model atoms in spectroscopy, and to their importance in recent parity violation experiments. Very precise determinations of hyperfine coupling constants in the rubidium and cesium resonant P levels using frequency-stabilized laser diodes and atomic beams serve as illustration of the exceptional quality of some recent measurements [2,3].

Because of the approximately Z^3 scaling of atomic parity violation effects, current interest in precise measurement of atomic observables has focused primarily on the heaviest alkali atoms. Beyond this, general theoretical approaches that can reliably predict physical properties in one atomic system should be able to be applied with confidence to other similar atomic systems as well. In this paper exceptionally precise experimental values of the hyperfine-structure constants for the $4P\ ^2P_{3/2}$ level of ^{41}K are reported. The precision of these measurements, and of others made recently by a similar technique, go well beyond the characteristic accuracy obtained in theoretical calculations of excited-level hyperfine structure.

A pump-probe technique applied to quantum beats in atomic electronic polarization was used to conduct the measurements. As reported previously [4], this method permits high precision determinations of excited-state hyperfine constants in systems which may be hard to study in other ways. The method is especially useful for investigations of low-lying P and D levels in alkali [4,5] and alkaline earth atoms.

II. EXPERIMENTAL APPROACH AND SETUP

A partial energy-level diagram for potassium which includes the levels relevant to this report is shown in Fig. 1. In the pump-probe quantum-beat approach illustrated there, a linearly polarized broadband pump laser is tuned to an atomic transition, producing an initial electronic alignment in an intermediate level, which is the state of interest. The electronic alignment, and its time evolution due to hyperfine interactions, is monitored by a probe laser, which has its frequency set to a second atomic transition originating in the level populated by the pump laser. For linearly polarized pump and probe beams, such monitoring is possible because the probability of the second transition depends on the time evolution of the spatial electronic charge distribution in the level under investigation.

The probe linear polarization is alternatively switched from parallel to perpendicular to that of the pump. The relative numbers of atoms promoted to the final state are compared and used for evaluation of the electronic polarization

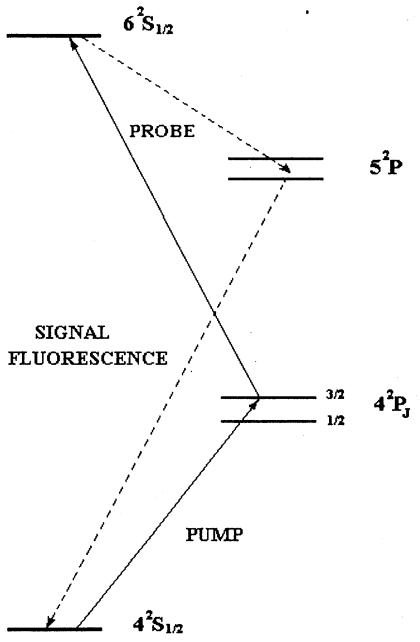

FIG. 1. Partial energy-level scheme for atomic potassium including levels involved in the experiment.

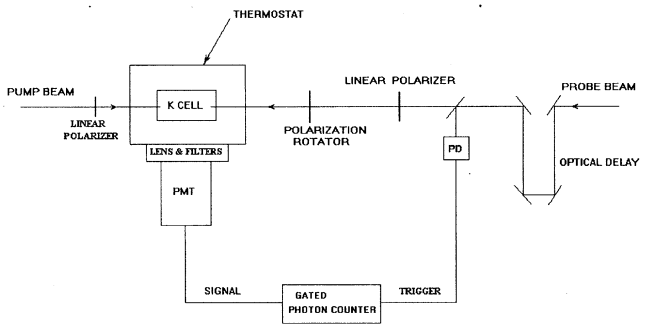

FIG. 2. Schematic diagram of the experimental setup.

of the intermediate level at any time. Atoms in the final state are detected by their direct or cascade fluorescence, generating signals I_{par} and I_{per}. Measurement of the fluorescence intensities as a function of the delay time between pump and probe allows for a reconstruction of the time dependence of the atomic alignment due to hyperfine interactions. This, in turn, permits determination of the hyperfine coupling constants A and B. The method has sufficient sensitivity that periodic coherent evolution of the alignment can be observed over about ten lifetimes of the investigated level.

The hyperfine separations in the $4p\ ^2P_{3/2}$ level of ^{41}K are among the smallest found in resonant levels and are consequently difficult to measure. Spectroscopic frequency-domain resolution of the hyperfine components is practically impossible. The magnetic-dipole and the electric-quadrupole hyperfine constants are both small and nearly the same magnitude. As a result, two of the four hyperfine sublevels fall within a 0.1-MHz frequency range, and the third is only 3 MHz away, while the natural width is about 6 MHz. Although the hyperfine structure (hfs) in the $4p\ ^2P_{3/2}$ level in other isotopes of potassium has been remeasured using the modern techniques of radio-frequency-modulated laser beams and atomic beams [6], for ^{41}K the initial level-crossing result [7] and the modified atomic beams measurement of Boroske and Zimmermann [8] have remained the only measurement in nearly 25 years. Utilizing the unique features of a pump-probe version of the quantum-beat method, we have been able to determine A and B with much-better-than-natural-width resolution and to reduce their uncertainties by an order of magnitude.

Our experimental setup is shown in Fig. 2. In the experiment, a saturated ^{41}K vapor was contained in an evacuated Pyrex cell that was kept at a temperature of 317 K. Pump-and-probe light pulses were produced by two tunable dye lasers pumped by a single neodymium: yttrium aluminum garnet (Nd:YAG) laser emitting approximately 6-ns pulses at a 10-Hz repetition rate. The green output from the YAG second harmonic generator had a pulse energy of 200 mJ. Only a small portion of the green output (roughly 5%) was used in excitation of each dye. The dye lasers were of the grazing incidence type with linewidths of about 0.01 nm and produced approximately 50 kW at the desired wavelengths. The unattenuated power of the pump and probe lasers was sufficient for a strong saturation of both atomic transitions of interest ($4s\ ^2S_{1/2} \rightarrow 4p\ ^2P_{3/2}$ at 766.9 nm, and $4p\ ^2P_{3/2} \rightarrow 6s\ ^2S_{1/2}$ at 694.0 nm). However, data used for analysis were collected with both beams attenuated by a factor of at least 100 to ensure weak excitation, as was assumed in the model used in interpretation of the beat signals. At the lower power levels used in the measurements, light intensity effects on the observed polarization were suppressed well below the statistical error limit. Decreasing or increasing the intensity by a factor of 3 produced no detectable change in the data. As a rule, the saturation levels were kept well below 25% at all times. Removal of all attenuating neutral density filters from either of the beams produced at least a fourfold increase of the fluorescence signals.

The two beams were counterpropagating within the resonance cell, were about 5 mm in diameter, and were made to overlap within the interaction region of the cell. The interaction region was about 2.5 cm long and maintained free of electric and magnetic fields. Two pairs of Helmholtz coils were used to compensate the horizontal and vertical components of the earth field to better than a 5-mG level. Changes of the probe polarization were made every 1000 shots using a Fresnel rhomb-type polarization rotator. Crystal polarizers with an extinction ratio of 10^5 were used to assure clean linear polarization of the beams and to verify the $\lambda/2$ retardation of the rhomb. The cascade fluorescence at about 404 nm from the $5p\ ^2P_j$ levels to the ground state was monitored by a RCA 31034 photomultiplier tube (PMT) working in a single photon counting mode. The photon counter was gated. The gate was opened by signals from a photodiode monitoring the probe laser beam. This ensured that the timing of the signal collection was constant as the delay was varied. The gate remained open for 300 ns, slightly longer than the combined lifetimes of the potassium $6s\ ^2S_{1/2}$ and $5p\ ^2P_{3/2}$ levels. The PMT was thermoelectrically cooled and had a dark count rate of about 30 counts per second or 10^{-5} dark counts per observation window. Blocking either of the beams eliminated the fluorescence signals. The signals were kept below one photon per pulse by means of attenuating filters in the laser beams. Since the overall time resolution of the PMT–electronic-counter (Stanford Research model 465) combination was about 10 ns the probability of photopulse overlap was negligibly small. Photoelectron counts for two different probe polarizations were used to calculate the linear polarization degree $P_L=(I_{\text{par}}-I_{\text{per}})/(I_{\text{par}}+I_{\text{per}})$ for a given pump-probe delay. Probe delays were changed by varying the optical path length of the probe beam relative to that of the pump beam. Delay values were estimated from the direct distance measurements by using a metrological tape and by adopting a refraction index for air of 1.000 29. Placing a fast photodiode in the position of the resonance cell and observing signals on a 1-GHz oscilloscope allowed for an independent time calibration for selected delays. Direct and electronic estimations agreed within the ± 0.5-ns time resolution of the electronics.

III. RESULTS

As the delay of the probe with respect to the pump was increased, periodic variations in the polarization were observed. The frequencies involved are directly related to the

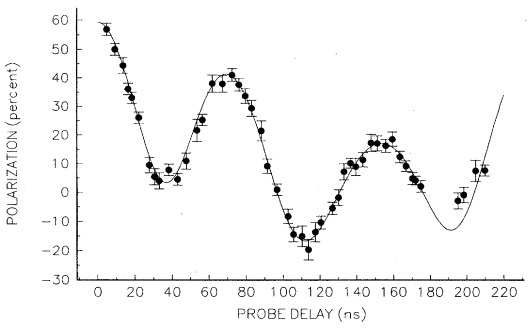

FIG. 3. Experimental data and best-fit curve showing the time evolution of the electronic polarization caused by the hyperfine interactions in the ^{41}K 4p $^2P_{3/2}$ state.

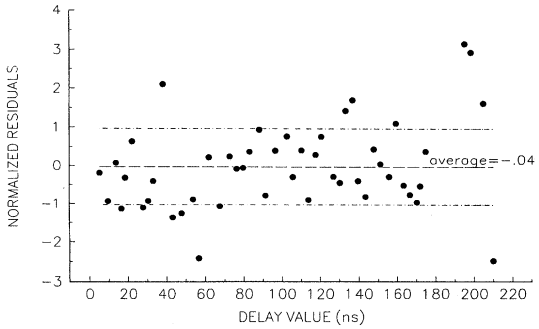

FIG. 4. Residuals of the fit of the analytical expressions to the polarization data. Values are normalized to the statistical uncertainty of each data point.

hyperfine energy separations in the intermediate level. The polarization data points are shown in Fig. 3. In the case of an instantaneous excitation and probing, and in the absence of collisional or other depolarization processes, the polarization is given by

$$P_L = 3g(t)/[4+g(t)] \ .$$

The depolarization coefficient $g(t)$ is given by

$$g(t) = \tfrac{27}{100} + \tfrac{7}{20}\cos[2\pi(3A+B)t] + \tfrac{21}{200}\cos[2\pi(5At)]$$

$$+ \tfrac{3}{20}\cos[2\pi(2A-B)t] + \tfrac{1}{8}\cos[2\pi(3A-2B)t] \ .$$

The coefficients in $g(t)$ are given in terms of angular momentum degeneracy factors and Racah coefficients $W(j,f,j,f';i,2)$, where the electronic angular momentum $j=\frac{3}{2}$, the nuclear spin $i=\frac{3}{2}$, and f and f' are the possible total angular momentum values in the 4p $^2P_{3/2}$ level.

The above formulas have been derived from general expressions given elsewhere [9,10]. A and B are the hyperfine coupling constants and are considered as the only freely adjustable parameters. A standard least-squares fit using the P_L formula gave an acceptable reduced-χ^2 value. However, the 5–7-ns duration time of the dye laser pulses used in the experiment cannot be totally neglected in comparison with the 4p $^2P_{3/2}$ lifetime or the hyperfine precession periods. Two kinds of effects are expected. First, the finite lifetime makes later-excited, earlier-probed atoms contribute relatively more to the signal. Second, the hyperfine frequency oscillations in the observed atomic polarization will be somewhat washed out when integration of the polarization over the pump and probe duration takes place. Both effects are rather small in our case but undoubtedly present. Assuming a square temporal pulse shape for the laser pulses, the effect of finite pulse duration can be treated analytically and is equivalent to a slight modification of the relative amplitudes and phases of the cosine-type contributions to the signal. When the modified formulas were used, a slight improvement in the quality of the fits combined with a shift in the derived A and B was observed. The reduced-χ^2 value goes through a minimum at 1.3 when the dye laser pulse durations are about 7 ns, as expected. By rejecting the four "noisiest" data points, which correspond to the longest delays and the lowest signals, the reduced-χ^2 value may be made smaller yet. However, this procedure had a negligible effect on the final parameters of the fit, which were obtained using all the data presented in Fig. 3.

Because the finite pulse corrections to A and B were smaller than their statistical errors, we elected to use the analytical square-pulse corrections as described, rather than a more tedious full scale numerical deconvolution procedure involving more realistic Gaussian-shaped pulses. A comparison of computer-generated signals corresponding to both square and Gaussian pulses of similar width convinced us that the square-pulse corrections were indeed adequate. The fit of the pulse-length-corrected formula to the experimental data is shown in Fig. 3.

Figure 4 shows the distribution of the residuals of the fit, normalized to the statistical error in each point. The overall agreement is seen to be very satisfactory. Note that several major rearrangements of the mirrors in the optical delay line accompanied by new measurements of all distances had to be done during the course of experiment. This may be responsible for the suggestion of clustering of low or high readings in the data. In any case, such systematic effects should be no larger than about 1 ns. Finally, fluorescence signals were observed at probe delays even larger than 220 ns, but the low intensity of the signal and signs of deterioration of mechanical stability in the >70-m-long delay path did not permit reliable polarization measurements.

Each experimental point in the polarization graph represents fluorescence intensities averaged over typically 40 000 laser shots. The least-squares fit to the formula describing the time evolution of the polarization gives values for A and B of 3.325(15) MHz and 3.230(23) MHz, respectively. The quoted errors represent one standard deviation.

TABLE I. Summary of experimental determinations of the hyperfine coupling constants A and B.

A (MHz)	B (MHz)	Reference
3.325(15)	3.230(23)	This work
3.40(8)	3.34(24)	[7]
3.6(3)	2.8(8)	[8]

IV. DISCUSSION

Our values for A and B are consistent with previous determinations by Ney [7] and by Boroske and Zimmermann [8] (see Table I), but are about an order of magnitude more accurate. As discussed previously, conscientious efforts have been made to ensure that any systematic errors are small with respect to the cited random error. To our knowledge there are no hyperfine-structure calculations carried out specifically for this level. It is believed, however, that techniques such as the multiconfiguration Dirac-Fock model when applied to the lowest atomic state of a given symmetry should be able to produce theoretical results of comparable precision [11].

We are presently extending this work to measurements of the hyperfine structures of the lowest D states in alkalis. Many of the higher-lying D states have been investigated recently with considerable success [12–14], but it appears that some of the low ones, which may be of most interest to theorists, have yet to be studied at all.

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