Sub-Doppler Nuclear Detection of Laser-Induced Orientation of $^{85}Rb^m$

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Using laser-induced nuclear-orientation and optical-saturation techniques, we have produced sub-Doppler changes in the anisotropy of the angular distribution of the 514-keV γ rays from the 1-μs isomer $^{85}Rb^m$. This permits high-resolution measurements of $D1$ and $D2$ hyperfine transitions of atoms of the isomer, which give the isomer's nuclear electric quadrupole moment, -0.73 ± 0.17 b, and its nuclear magnetic dipole moment, $(6.043 \pm 0.005)\mu_N$. The isomer shift, -113 ± 8 MHz relative to the ground state of ^{85}Rb, corresponds to a change in the mean-square charge radius of $+0.174 \pm 0.008$ fm^2.

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We report the sub-Doppler modulation of the spatial distribution of γ radiation emitted from atoms with unstable nuclei. This achievement significantly advances our ability to use laser-induced nuclear orientation to measure optical hyperfine spectra of radioactive atoms and so to determine their nuclear parameters. We have optically pumped radioactive rubidium atoms at both the $D1$ (795 nm) and $D2$ (780 nm) transitions and produced resonant changes in the angular distribution of the 514-keV γ rays, revealing hyperfine structure features of 90 MHz, approximately 10% of the Doppler width. This tenfold improvement in resolution from our previous work[1] has allowed for the first time the measurement of the hyperfine splittings of closely spaced optical $D2$ transitions in atoms of $^{85}Rb^m$, yielding a value of the nuclear electric quadrupole moment. Also, higher-resolution measurements of the $D1$ splittings give more precise and accurate values for the isomer shift and nuclear magnetic dipole moment of this isomer. The demonstration of this sub-Doppler technique is especially timely given the recent improvements in on-line cells for studying the optical spectroscopy of radioactive atoms.[2] Further, the increased resolution and sensitivity obtained in these experiments may also be useful for proposed studies of β-decay velocity distributions.[3]

The production of a sub-Doppler change signal in nuclear radiation and its use to detect optically induced nuclear orientation is the latest step in a series of developments begun by Kastler.[4] His work made it clear that absorption of polarized light by electronic transitions could lead to nuclear orientation, a possibility later verified by Bitter *et al.*[5] In the 1970s Otten and his colleagues showed that such optically induced orientation could be detected by the changes it produced in the angular distribution of nuclear emissions from radioactive atoms.[6] In 1979 Murnick and Feld[7] proposed that changes in the spatial distribution of nuclear radiations could be produced with sub-Doppler resolution. Our present results constitute the first experimental demonstration of this idea.

For these experiments we have used a system particularly convenient for studying laser-induced nuclear orientation.[1] Radioactive atoms with nuclei that are 1-μs, 514-keV isomers of ^{85}Rb are produced by the β decay of 10.7-yr ^{85}Kr contained at a pressure of few Torr or less in a cell of a few cubic centimeters volume. The gas serves both as a source of the isomer and as a buffer, lengthening the time the isomers remain in the laser beam. Optically pumping atoms of this rubidium isomer on a hyperfine transition induces spatial anisotropy in the nuclear γ rays. The detection of such anisotropy occurs when the laser is tuned to an optical resonance of the atom. Experimental investigation over a wide range of parameters[8] shows that the system is well modeled using the approach developed by Quivers[9] and is therefore especially suitable for the quantitative characterization of the physical processes affecting nuclear orientation.

The need for sub-Doppler resolution was evident from earlier work.[1] Those experiments yielded curves of anisotropy versus laser frequency which had resolution limited by the Doppler full width at half maximum of 890 MHz. This resolution is insufficient for studying more closely spaced hyperfine levels such as those of the $^2P_{3/2}$ state, which have spacings of $\sim$300 MHz. With sub-Doppler resolution we have now been able to resolve the $^2P_{3/2}$ hyperfine splittings, find the B hyperfine coefficient, and extract a value for the nuclear electric quadrupole moment of $^{85}Rb^m$.

Sub-Doppler modulation of the γ-ray angular distribution is achieved by using standing-wave saturation spectroscopy. This technique can be used because the population transfer among the sublevels of the isomer and the resulting anisotropy are altered by an intense resonant laser field. In our experiments the laser beam is split into two overlapping beams of equal power, which are passed through the cell collinearly and in opposite directions. Each beam interacts only with those atoms in the Maxwellian distribution that have a velocity which Doppler shifts them into resonance with the laser light. When the laser frequency is detuned from an atomic-

transition center frequency the beams interact with two distinct velocity groups symmetrically located on opposite sides of the Doppler velocity profile, and the resulting signal is proportional to the sum of the number of atoms in those groups. On resonance, however, both beams interact with the same zero-velocity group and, as a result, the system saturates more strongly. Therefore, each time the applied field is tuned through the center of an atomic transition, a dip is created in the anisotropy signal. This saturation effect is similar to the Lamb dip observed in optical-fluorescence experiments, but here it is observed in the spatial distribution of the γ rays emitted from the short-lived isomers.

Figure 1 shows an example of a measured sub-Doppler change signal and a prediction of the change signal obtained from a version of the model developed by Quivers.[9] In this model the Zeeman sublevels are grouped into equivalent levels and the population distribution is then found by integrating the rate equations. The velocity-dependent aspects are taken into account by dividing the Doppler distribution into a set of subgroups, and solving the steady-state rate equations for each group. The equations are coupled through stimulated and spontaneous emissions and by velocity-changing collisions, which shift atoms from one velocity group to another. The collisions are treated in the strong-collision limit. There are no free parameters in the model. Agreement between the predictions of the model and the experimental observations is quite good, especially in view of the simplifying assumptions made.

The difference between the Doppler-broadened anisotropy signal produced by a traveling wave and the anisotropy produced by the standing wave is defined as the change signal. This quantity is plotted in Figs. 1 and 2. The line shape has two distinct features, a sharp narrow feature (conventionally called a "dip") and a broad pedestal. The dip represents optically pumped atoms which do not undergo velocity-changing collisions, and the pedestal represents optically pumped atoms which have undergone velocity-changing collisions.[9] In our model, and in our experimental results, the dip is well represented by a Lorentzian line shape and the pedestal by a Gaussian line shape. As buffer-gas pressure is increased the pedestal grows at the expense of the dip; at high enough pressures the dip disappears.

Details of the experimental arrangement are as follows. The isomers are formed in a Pyrex cell 40 mm long and 12 mm in diameter filled with 300 mTorr of Kr containing $\sim$0.25 mCi of 10.7-yr ^{85}Kr.

Only 0.4% of the ^{85}Kr β decays form the isomeric state at 514 keV in ^{85}Rb. The laser beams were produced by a Coherent autoscan ring dye laser using LD700 dye pumped with a 6-W krypton-ion laser. The output power of the dye laser was approximately 300 mW at 795 nm and 500 mW at 780 nm. A set of three-axis coils canceled the Earth's magnetic field and supplied a 1-G keeper field along the quantization axis perpendicular to the axis of the laser beam. The γ rays were detected with two 3-in. NaI(Tl) detectors placed at 0° and 180° relative to the quantization axis, the positions of largest anisotropy.

A cell pressure of 300 mTorr was chosen for this experiment as a compromise between a lower value at which the γ-ray count rate would become unacceptably

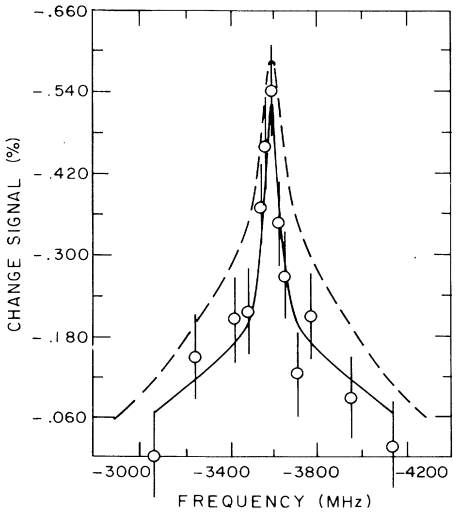

FIG. 1. Sub-Doppler change signal for the $^2S_{1/2}$ $F=5$ to $^2P_{1/2}$ $F=4$ transition. The solid line is least-squares fit by a composite Lorentzian and Doppler signal. The dashed line is model prediction with no free parameters. The frequency refers to ^{87}Rb $D1$ $F=2$ to $F'=1$ transition.

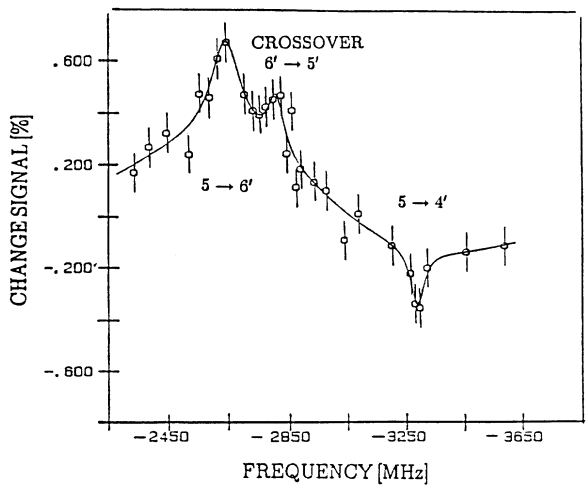

FIG. 2. Sub-Doppler change signals for $D2$ transitions. Sharp features visible from higher to lower frequency are the $^2S_{1/2}$ $F=5$ to $^2P_{3/2}$ $F'=6$ transition, the $^2P_{3/2}$ $F=6$ to $^2P_{3/2}$ $F=5$ crossover, and the $^2S_{1/2}$ $F=5$ to $^2P_{3/2}$ $F'=4$ transition. The frequency refers to ^{87}Rb $D2$ $F=2$ to $F'=1$ transition.

weak due to low density, and a higher value where the number of velocity-changing collisions would produce only a pedestal, erasing the sub-Doppler dip entirely. It is important that the $^{85}Rb^m$ rapidly form neutral atoms, because the mean life of the isomers is only 1.4 μs. Although Kr is an effective buffer gas, it is an inefficient charge exchanger, so a small amount of natural Rb having a cross section for resonant charge exchange with the isomers on the order of 500 Å^2 was put into the cell. The vapor density of the natural rubidium was controlled by an oven heated to 150 °C. The sensitivity of the experiment can be seen from the fact that the density of the isomeric atoms at any time is only 0.1/cm^3 and the count rate is about 1200/s.

Data were taken at selected frequencies for 10-min intervals to minimize effects of thermal drift in the laser. Each data point required a total of 5 h of counting time. Anisotropy signals were measured as a difference in the γ-ray count rate with laser on and off for 30-s intervals. The anisotropy-change signal was then calculated as the difference between the anisotropies measured with one laser excitation and excitation from two oppositely traveling laser beams (standing wave).

In this work all optical pumping was done with linearly polarized light. Strong γ anisotropy signals were obtained for two $D1$ transitions, two $D2$ transitions, and a $D2$ crossover resonance. Measured change signals of one $D1$ and the three $D2$ transitions are shown in Figs. 1 and 2. Other transitions strong enough to study could have been obtained by pumping the atoms from both ground-state hyperfine levels with two lasers or by using circularly polarized light. However, the measured transitions were sufficient to yield all the nuclear parameter information available from hyperfine spectra.

Table I shows the observed $^{85}Rb^m$ transitions and their relative frequencies. Because the lines of $^{85}Rb^m$ lie closest to those of ^{87}Rb, these were used as reference lines and were located with high precision by laser saturation spectroscopy in a reference cell operated in parallel with our apparatus. The frequency calibration was carried out with the Coherent autoscan 699-29 dye laser to an accuracy of ± 3 MHz. An overall fit of the data points enabled us to locate the line centers within ± 6 MHz. Using the data[10] on hyperfine splittings of ^{87}Rb and the isotopic shift of 80.1 MHz between ^{87}Rb and ^{85}Rb,[11,12] the relative frequencies $\Delta\nu$ were calculated with respect to the ^{85}Rb center of mass of the $D1$ ($D2$) multiplets.

The analysis to determine nuclear parameters is straightforward. From the $D1$-transition data, we determine the sum of the $^{85}Rb^m$ hyperfine constants $A_{S_{1/2}} + A_{P_{1/2}}$. Using simple scaling arguments we combine this sum with the corresponding sum of hyperfine constants and magnetic moment of ^{85}Rb and obtain an accurate value of the magnetic moment of $^{85}Rb^m$. From this result we then determine $A_{S_{1/2}}$, $A_{P_{1/2}}$, and $A_{P_{3/2}}$ for $^{85}Rb^m$ and use the value of $A_{P_{3/2}}$ to extract B from our $D2$-transition data. In obtaining these constants we have assumed that electronic effects are identical in corresponding atomic states of $^{85}Rb^m$ and ^{85}Rb atoms and that there is no appreciable magnetic hyperfine anomaly. An average value of the isomer shift I_s, which is the frequency difference between $^{85}Rb^m$ and ^{85}Rb electronic transitions, is then calculated from the above constants and the known data[12] on ^{85}Rb. Following the procedure of Thibault *et al.*,[11] the quadrupole moment is obtained from the B value and the change in the mean-square charge radius $\langle \delta r^2 \rangle (^{85}Rb^m - ^{85}Rb)$ is obtained from the isomer-shift value.

Table II lists atomic and nuclear parameters derived in this study for $^{85}Rb^m$.

The value for the magnetic dipole moment agrees with earlier measurements made using a Doppler-broadened signal.[1] The isomer shift is substantially different from the previously reported value, for which an insufficient correction was made for absorption of pump light by the natural Rb. Such a correction is unnecessary in the present work because the resonance is narrow and the measurements are made at lower Rb density, where absorption is greatly diminished.

The magnetic moment of the $\frac{9}{2}^+$ isomer $^{85}Rb^m$ is close to the value expected for a single-particle nucleus with a $g_{9/2}$ proton. The value is also close to the magnetic moment $(5.598 \pm 0.002)\mu_N$ of the $\frac{9}{2}^+$ isomer $^{81}Rb^m$. There is similar agreement[11] with the quadrupole moment of $^{81}Rb^m$, which is -0.743 ± 0.057 b. Deformation parameters for $^{85}Rb^m$ and $^{81}Rb^m$, which can be extracted from the isomer shifts, are also similar, 0.26 and

TABLE I. Observed $^{85}Rb^m$ transitions and their relative frequencies. $\Delta\nu$ values in the last column are the frequencies of $^{85}Rb^m$ transitions with respect to ^{85}Rb center of mass of $D1$ ($D2$) multiplets.

	Observed $^{85}Rb^m$ transitions	^{87}Rb reference transition	$\Delta\nu$ measured (± 6 MHz)	$\Delta\nu$ reference, ^{85}Rb (± 6 MHz)
$D1$	$F=5 \rightarrow F'=4$	$F=2 \rightarrow F'=1$	−3602	−6593
$D1$	$F=4 \rightarrow F'=5$	$F=1 \rightarrow F''=2$	2800	7457
$D2$	$F=5 \rightarrow F'=4$	$F=2 \rightarrow F'=1$	−3291	−6004
$D2$	$F=5 \rightarrow F'=6$	$F=2 \rightarrow F'=1$	−2641	−5354
$D2$	$(6 \rightarrow 5)$ crossover	$F=2 \rightarrow F'=1$	−2806	−5519

TABLE II. Hyperfine coefficients, isomer shift, and corresponding nuclear parameters.

$A_{S_{1/2}} = 2510 \pm 2$ MHz	$A_{P_{1/2}} = 299.5 \pm 0.7$ MHz
$A_{P_{3/2}} = 62.04 \pm 0.07$ MHz	$B = -68.2 \pm 15$ MHz
$\mu = (6.043 \pm 0.005)\mu_N$	$I_s = \Delta\nu(^{85}\mathrm{Rb}^m - ^{85}\mathrm{Rb}) = -113 \pm 8$ MHz
$Q = -0.73 \pm 0.17$ b	$\langle \delta r^2 \rangle (^{85}\mathrm{Rb}^m - ^{85}\mathrm{Rb}) = +0.1740 \pm 0.008$ fm^2

0.27, respectively. These new high-resolution nuclear data should be useful for testing nuclear shape and surface models.[13]

As discussed above, under the conditions of this experiment, the sub-Doppler γ anisotropy-change signal originates from an alteration in the ground-state velocity distributions of the M_F components of the isomeric atoms. As pointed out in Ref. 7, under the appropriate optical pumping conditions this change signal should be accompanied by a sub-Doppler feature in the *spectral distribution* of γ rays, as observed along the axis of the laser beam. Observation of this effect requires a $\Delta F = -1$ transition optically pumped with linearly polarized laser light,[7] just as in the experiments reported here. In the present experiment, this narrowed spectral component, which would occur even with a traveling-wave field, is about 10% of the $\sim$0.5-eV Doppler width of the emitted γ rays, and may be tuned across the γ-ray Doppler profile by tuning the laser field through the 890-MHz Doppler profile of the isomeric $D1$ ($D2$) resonances. Experiments to utilize this tunable γ-ray signal would be of interest.

In conclusion, optical-saturation spectroscopy has been used to produce changes in the spatial distribution of γ rays from a short-lived isomer exhibiting resonant features with a width 10% of the Doppler broadening. Using this technique, we have determined the nuclear electric quadrupole moment of a 1-μs isomer. This is the shortest-lived atom for which the hyperfine structure has been measured using optical techniques. The laser-induced nuclear-orientation technique may be useful in on-line studies where it could yield higher-resolution spectra from short-lived atoms than previously possible with resonance cells.

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