Observation of the hyperfine structure of the $^2S_{1/2}$-$^2D_{5/2}$ transition in $^{87}Sr^+$

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The hyperfine structure of the $^2S_{1/2}$-$^2D_{5/2}$ quadrupole transition at 674 nm in $^{87}Sr^+$ has been observed. The ion was confined in a Paul trap and cooled using laser radiation at 422 and 1092 nm. The quadrupole transition was observed by monitoring quantum jumps in the 422-nm fluorescence. The odd isotope of strontium has ''clock'' transitions independent of the first-order Zeeman shift and the $^2D_{5/2}$ state hyperfine structure constants have been determined as $A_{D_{5/2}}=2.1743(14)$ MHz and $B_{D_{5/2}}=49.11(6)$ MHz. Standard uncertainties have been added in parentheses. These values allow the second-order Zeeman shifts to be calculated. The $^{88}Sr^+$-$^{87}Sr^+$ isotope shift for the 674-nm quadrupole transition has been measured to be 247.99(4) MHz.

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INTRODUCTION

In recent years, there has been significant progress towards the use of narrow optical transitions in cold trapped ions as optical frequency standards. A number of candidate ions are being investigated worldwide and some have been accurately measured with a femtosecond laser comb. At the National Physical Laboratory (NPL), an optical frequency standard based on $^{88}Sr^+$ is being developed. The frequency of the $^2S_{1/2}$-$^2D_{5/2}$ quadrupole ''clock'' transition at 674 nm in this ion is a recommended radiation for realization of the meter [1]. However, this isotope has zero spin and the ten Zeeman components of the clock transition all have a linear dependence on the magnetic field. The odd isotope $^{87}Sr^+$ has a spin of $I=\frac{9}{2}$ and so the $m_F=0$ levels of the $^2S_{1/2}$ and $^2D_{5/2}$ states are independent of the first-order Zeeman effect. A partial term scheme is shown in Fig. 1. The hyperfine splittings of the $^2S_{1/2}$ and $^2P_{1/2}$ levels are experimental values derived from measurements made in an ion trap [2] and a fast beam [3]. Following on from earlier NPL work, where a cooled $^{87}Sr^+$ ion was confined in a 1-mm-diam Paul trap [4], this paper describes the interrogation of the $^2S_{1/2}$-$^2D_{5/2}$ quadrupole transition and measurement of the $^2D_{5/2}$ level hyperfine constants. The trap is mounted inside a vacuum system within a μ-metal enclosure in order to reduce magnetic-field changes inside the trap. The oven contains a compound of isotopically enriched strontium-87. Three orthogonal coils are also mounted around the trap in order to reduce the dc magnetic field to ≈ 1 μT.

The cooling laser system is shown in Fig. 2 and comprises two frequency-doubled 844-nm diode laser systems [5,6]. These diode lasers are offset locked at $\approx$2.5 GHz, which is half the ground-state splitting. The two lasers are independently frequency doubled and the beams at 422 nm recombined before input into the trap. Additionally, the 422-nm laser driving the $^2S_{1/2}(F=5)$-$^2P_{1/2}(F'=5)$ transition needs to avoid pumping into a dark state, which will arise in the 1-μT magnetic field. This problem has been discussed in detail [7] and one solution is to split the 422-nm laser into two beams, one π polarized and one σ polarized and frequency separated by a few MHz. This is shown in Fig. 2 and the frequency separation of 7 MHz is generated by the frequency difference between the two acousto-optic modulators AOM_1 and AOM_2. It was verified experimentally that this frequency difference optimized the observed fluorescence, as predicted [7]. The second 422-nm beam, driving the $^2S_{1/2}(F=4)$-$^2P_{1/2}(F=5)$ transition can be switched using acousto-optic modulator AOM_3. In addition to the lasers at

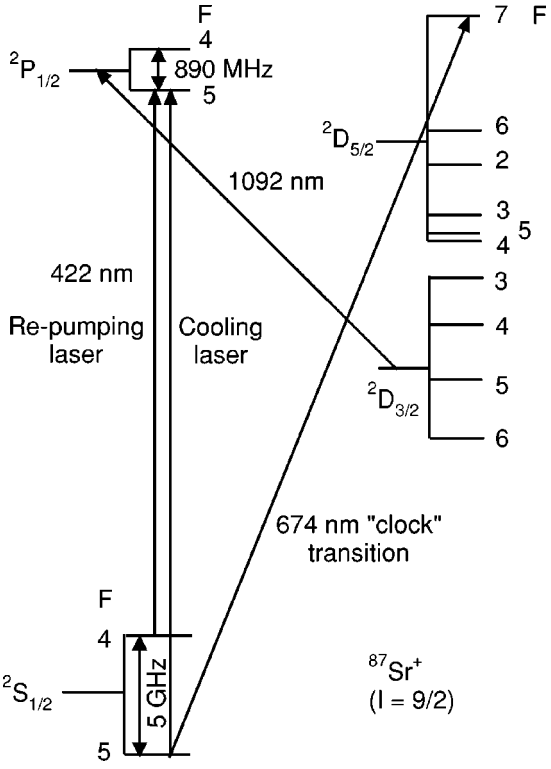

FIG. 1. Partial term scheme for $^{87}Sr^+$, showing the cooling laser transitions at 422 and 1092 nm, together with the quadrupole transitions at 674 nm.

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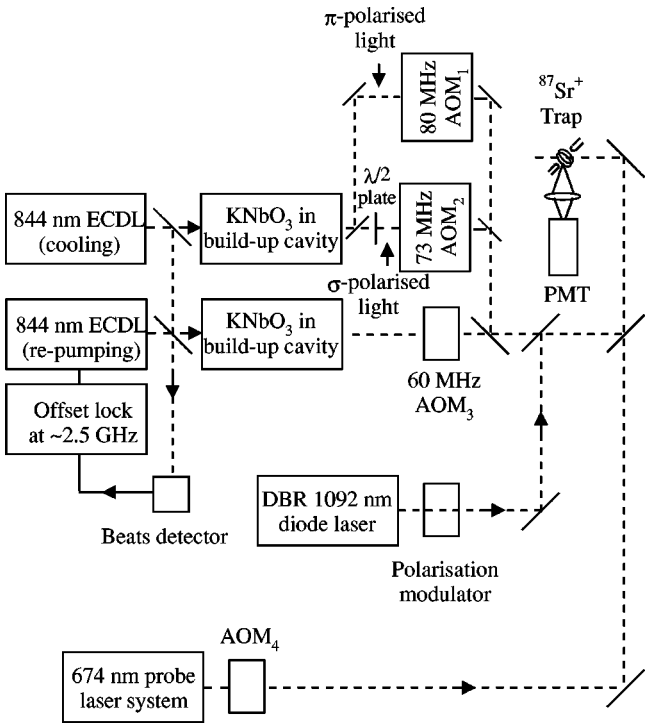


FIG. 2. Experimental arrangement for the $^{87}Sr^{+}$ cooling and probe laser systems. The system of probe lasers is designed to avoid optical pumping into a dark state of the $^{2}S_{1/2}(F=5)$-$^{2}P_{1/2}(F'=5)$ transition [7].

422 nm, a further laser is required at 1092 nm, tuned to the $^{2}P_{1/2}$-$^{2}D_{3/2}$ transition, to prevent the ion being driven into one of the $^{2}D_{3/2}(F'=3, 4, 5, \text{or } 6)$ manifold of lines. This is a distributed Bragg reflector laser, which is frequency modulated at a rate of 0.8 MHz over the few hundred MHz frequency range of the hyperfine structure. In addition, the laser is polarization modulated at 2 MHz in order to avoid the ion being pumped into dark states of the $^{2}P_{1/2}$-$^{2}D_{3/2}$ transition. This is the same technique as used previously with $^{88}Sr^{+}$ [8].

The quadrupole $^{2}S_{1/2}$-$^{2}D_{5/2}$ transition at 674 nm is driven by a Al-Ga-In-P diode laser. This laser is optically narrowed [9] and then locked to a highly stable, and nontunable, high-finesse cavity via a Pound-Drever lock, as shown in Fig. 3. This ultra-low-expansion (ULE) cavity has a measured free spectral range (FSR) of $\nu_{\mathrm{FSR}}=1504.360(7)$ MHz and finesse of $\approx 2\times 10^{5}$. For this lock, the laser drive current is modulated at 2.2 MHz and laser linewidths of 200 Hz have been reported for this system [4]. Laser tuning is provided by the use of acousto-optic modulators, in order to bridge the frequency interval between the nearest mode of the nontunable ULE reference cavity and the strontium transition. When interrogating $^{88}Sr^{+}$, a 370-MHz double-passed acousto-optic modulator is used, since the transition frequency is approximately 740 MHz higher in frequency than the nearest cavity mode frequency (ν_n). In order to drive the $^{2}S_{1/2}(F=5)$-$^{2}D_{5/2}$ manifold of lines in $^{87}Sr^{+}$, the probe laser is locked to a frequency $\nu_{n+2}=\nu_n+2\nu_{\mathrm{FSR}}$, two cavity modes higher than the frequency for $^{88}Sr^{+}$. The frequencies of the observed hyperfine structure for transitions from the $^{2}S_{1/2}(F=5)$ ground state are then between 227 and 284 MHz lower in frequency than ν_{n+2}. The 674-nm lines from the $F=4$ ground state are observed by locking the probe laser to the frequency $\nu_{n-1}=\nu_n-\nu_{\mathrm{FSR}}$. The frequencies of the hyperfine transitions from the $^{2}S_{1/2}(F=4)$ ground state are around 754 to 774 MHz lower in frequency than ν_{n-1}.

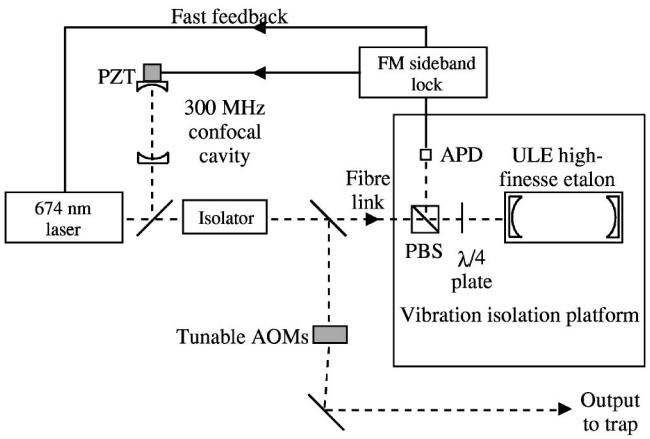


FIG. 3. Diode laser system at 674 nm, for probing the $^{87}Sr^{+}$ hyperfine structure.

HYPERFINE STRUCTURE OF THE $^{2}S_{1/2}$-$^{2}D_{5/2}$ TRANSITION IN $^{87}Sr^{+}$

Five transitions were observed at 674 nm from the $F=5$ ground state, namely $^{2}S_{1/2}(F=5)$-$^{2}D_{5/2}(F'=3, 4, 5, 6, \text{and } 7)$. These were observed and a sum of three scans over the $^{2}S_{1/2}(F=5)$-$^{2}D_{5/2}(F'=4)$ transition is shown in Fig. 4. The number of quantum jumps is plotted against the frequency offset from the $(n+2)$th mode of the ULE cavity. In a magnetic field of ≈ 1 μT, and with a probe laser linewidth of typically a few hundred Hz, the individual Zeeman components are not resolved. The observed linewidths are therefore determined by the frequency spread of the unresolved Zee-

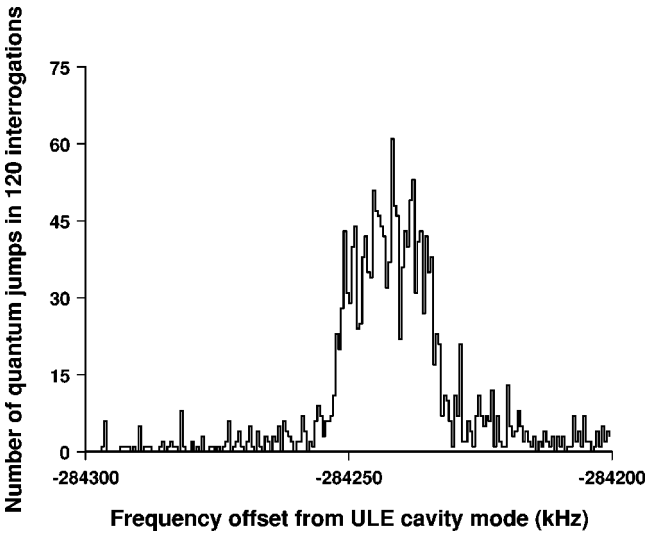


FIG. 4. Scan over the $^{2}S_{1/2}(F=5)$-$^{2}D_{5/2}(F'=4)$ transition in a 1-μT magnetic field. The data comprise a summation of three individual scans.

man components due to the ambient magnetic field. From the usual selection rules for quadrupole transitions ($\Delta m_F = 0$, ± 1, and ± 2) and the expected degeneracies of the various levels, it is possible to determine the maximum number of allowed Zeeman transitions. For transitions from the $^2S_{1/2}(F=5)$ ground state, this number ranges from 35 for transitions to the $^2D_{5/2}(F'=3)$ state to 55 for transitions to the $F'=7$ state.

In order to observe the hyperfine spectra, computer-controlled detection of quantum jumps and laser switching was used, similar to that for $^{88}Sr^+$ [5]. The detection cycle for observing the $^2S_{1/2}(F=5)$-$^2D_{5/2}$ transitions begins with the ion fluorescing in the trap, the 1092-nm laser and both 422-nm lasers on, but the 674-nm laser off. The two cooling beams driving the $^2S_{1/2}(F=5)$-$^2P_{1/2}(F'=5)$ transition are then turned off and the probe laser switched on for, typically, 3 ms. At the end of the probe pulse, the cooling laser is turned back on and the fluorescence level monitored to see whether the ion is in one of the $^2D_{5/2}$ hyperfine levels. The 422-nm repumping laser, driving the $^2S_{1/2}(F=4)$-$^2P_{1/2}(F'=5)$ transition and the 1092-nm laser, are kept on throughout. For detecting the $^2S_{1/2}(F=4)$-$^2D_{5/2}$ transitions, the same cycle is used, although the repumping laser is turned off during the probe pulse and the 422-nm cooling laser beams remain on.

The $S_{1/2}$ state splits into two levels, with $F=4$ and $F=5$, frequency shifted by $\nu_4 = -\frac{11}{4}A_{S_{1/2}}$ and $\nu_5 = \frac{9}{4}A_{S_{1/2}}$, respectively. Here, $A_{S_{1/2}}$ is the ground-state hyperfine constant, which has previously been measured to be -1000.4737 MHz [2]. The frequency of each of the five transitions from the $^2S_{1/2}(F=5)$ level can be expressed in the form [10]

$$\nu_{87} = \frac{1}{2}A_{D_{5/2}}K + B_{D_{5/2}}\frac{\frac{3}{2}K(K+1) - 2I(I+1)J(J+1)}{2I(2I-1)2J(2J-1)} - \frac{9}{4}A_{S1/2} - \Delta\nu_{87,88} + \nu_{88}. \quad (1)$$

In this equation, $K = F(F+1) - I(I+1) - J(J+1)$, $A_{D_{5/2}}$ and $B_{D_{5/2}}$ are the two $D_{5/2}$-state hyperfine constants. The $^{88}Sr^+$ $^2S_{1/2}$-$^2D_{5/2}$ transition frequency is denoted by ν_{88} and the isotope shift by $\Delta\nu_{87,88}$. As already noted, the isotope spin $I = 9/2$. A second equation, similar to Eq. 2 but with the term $-\frac{9}{4}A_{S_{1/2}}$ replaced by $+\frac{11}{4}A_{S_{1/2}}$, gives the transition frequencies from the $^2S_{1/2}(F=4)$ level.

In order to identify the components, sets of three of the five observed frequency offsets from the ULE cavity were used to calculate constants $A_{D_{5/2}}$ and $B_{D_{5/2}}$. These constants were then used to evaluate the frequencies of the other two transitions. Only one set of constants could be found which correctly predicted the frequencies of all five hyperfine components and the hyperfine quantum numbers could therefore be assigned unambiguously. The final values for the three constants were determined from the five frequency values by a least-squares fit. This gave values for the $A_{D_{5/2}}$ and $B_{D_{5/2}}$ coefficients of

$$A_{D_{5/2}} = 2.1743(14)\ \text{MHz}, \qquad B_{D_{5/2}} = 49.11(6)\ \text{MHz}. \quad (2)$$

The standard uncertainty of the determination is added in parentheses. These results are close to calculated values of $A_{D_{5/2}} = 1(1)$ MHz and $B_{D_{5/2}} = 52(4)$ MHz [11]. The results could also be used, together with the measured value for the ULE cavity FSR to obtain the isotope shift for the $^2S_{1/2}$-$^2D_{5/2}$ transition at 674 nm. In order to calculate the shift, allowance had to be made for the ULE cavity isothermal drift of 2.5 kHz/day [12]. The isotope shift was measured as 247.99(4) MHz, which compares with a calculated value of 206 MHz [13]. The standard uncertainty comprises contributions from the measurement statistics (25 kHz), the observed fluctuations in cavity length due to thermal fluctuations during the day (33 kHz) and twice the measurement uncertainty of the cavity FSR (14 kHz). These were then added in quadrature to give the total standard uncertainty of 40 kHz.

Results were also obtained on the $^2S_{1/2}(F=4)$-$^2D_{5/2}(F'=2, 3, 4, 5,$ and $6)$ transitions. This allowed determination of the ground-state splitting for comparison with previously published data, which served as a check on our data. The results gave a value of 5002.30(5) MHz, which compares well with the more accurate determination of 5002.368 365(55) MHz [2]. The uncertainty is similar to that for the isotope shift, except that the uncertainty calculation for the ground-state splitting involves three times rather than twice the ULE cavity FSR.

The values for the $A_{D_{5/2}}$ and $B_{D_{5/2}}$ coefficients, together with the isotope shift and previously published values for the ground-state splitting [2] and $^{88}Sr^+$ 674 nm frequency [14,15] may be used to calculate the absolute frequencies of

TABLE I. Frequencies of allowed quadrupole transitions from the $^2S_{1/2}(F=5)$ ground state in $^{87}Sr^+$.

$^2D_{5/2}$, $F'=$	Frequency difference from $F'=7$ (MHz)	Absolute frequency of the transition from the $^2S_{1/2}(F=5)$ ground state
2	−44.126	
3	−52.336	444 781 031.58(4) MHz
4	−57.553	444 781 026.36(4) MHz
5	−54.867	444 781 029.05(4) MHz
6	−38.138	444 781 045.78(4) MHz
7	0	444 781 083.91(4) MHz

TABLE II. Quadratic Zeeman shifts for the $^2D_{5/2}(m_F=0)$ levels, calculated from the experimentally determined constants of the hyperfine structure.

$^2D_{5/2}$, $F=$	Quadratic Zeeman shift (Hz/μT^2)
2	67
3	56
4	−341
5	189
6	20
7	6.4

the $^{87}Sr^+$ transitions. The frequencies for transitions from the $^2S_{1/2}(F=5)$ ground state are shown in Table I.

QUADRATIC ZEEMAN SHIFTS

With the experimental determination of the $^2D_{5/2}$ hyperfine constants in $^{87}Sr^+$, it is possible to calculate the quadratic Zeeman shift for the $m_F=0\rightarrow m_F'=0$ transitions. These are potential optical frequency references, which are free from the first-order Zeeman shift. For these levels, the quadratic Zeeman shift is dominated by the shift of the $^2D_{5/2}$ levels, since the hyperfine constants are much smaller for the $^2D_{5/2}$ level than the $^2S_{1/2}$ ground state. From second-order perturbation theory, the frequency shift $\Delta\nu$ in the presence of a small magnetic field B is given by [16]

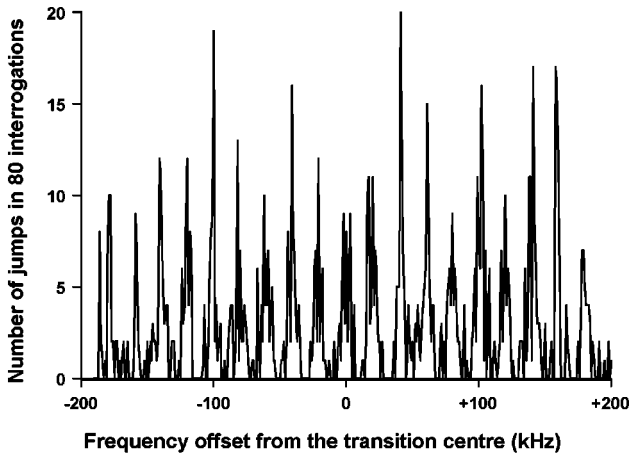
$$\Delta\nu(F')=\left(\frac{g_J eB}{4\pi m}\right)^2 \sum_{F''(F''\neq F')} \frac{|\langle IJF'm_F|J_z|IJF''m_F\rangle|^2}{(\nu_{F'}-\nu_{F''})}. \tag{3}$$

In this equation, g_J is the Landé factor for the $^2D_{5/2}$ state ($g_J=\frac{6}{5}$) and e and m are the electron charge and mass, respectively. For the six levels involved, the $^2D_{5/2}$ quadratic Zeeman shifts for the $m_F=0\rightarrow m_F'=0$ transitions are, therefore,

$$\Delta\nu(2)=-\left[\frac{1.964}{(3A_{D_{5/2}}-\frac{3}{10}B_{D_{5/2}})}\right]\left[\frac{g_J eB}{4\pi m}\right]^2, \quad F'=2 \tag{4}$$

$$\Delta\nu(3)=\left[\frac{1.964}{(3A_{D_{5/2}}-\frac{3}{10}B_{D_{5/2}})}-\frac{2.286}{(4A_{D_{5/2}}-\frac{17}{60}B_{D_{5/2}})}\right]\times\left[\frac{g_J eB}{4\pi m}\right]^2, \quad F'=3 \tag{5}$$

$$\Delta\nu(4)=\left[\frac{2.286}{(4A_{D_{5/2}}-\frac{7}{60}B_{D_{5/2}})}-\frac{2.068}{(5A_{D_{5/2}}-\frac{1}{6}B_{D_{5/2}})}\right]\times\left[\frac{g_J eB}{4\pi m}\right]^2, \quad F'=4 \tag{6}$$



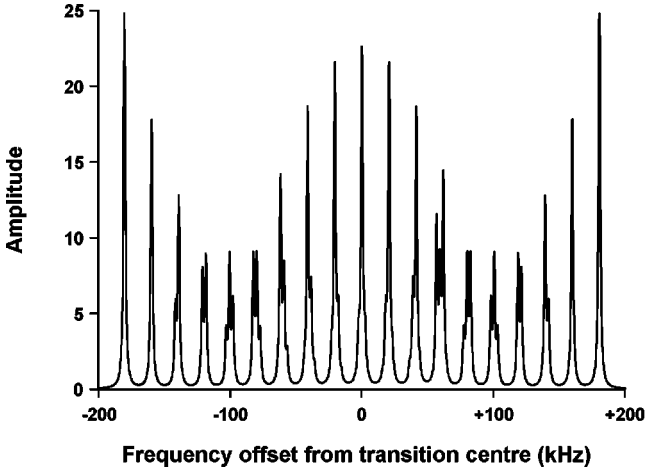


FIG. 5. Scan over the $^2S_{1/2}(F=5)$-$^2D_{5/2}(F'=7)$ transition. The data comprise a summation over two individual scans and have been compared with the calculated profile expected in a 6.5-μT field and 1-kHz laser resolution.

$$\Delta\nu(5)=\left[\frac{2.068}{(5A_{D_{5/2}}-\frac{1}{6}B_{D_{5/2}})}-\frac{1.566}{(6A_{D_{5/2}}+\frac{3}{40}B_{D_{5/2}})}\right]\times\left[\frac{g_J eB}{4\pi m}\right]^2, \quad F'=5 \tag{7}$$

$$\Delta\nu(6)=\left[\frac{1.566}{(6A_{D_{5/2}}+\frac{3}{40}B_{D_{5/2}})}-\frac{0.8654}{(7A_{D_{5/2}}+\frac{7}{15}B_{D_{5/2}})}\right]\times\left[\frac{g_J eB}{4\pi m}\right]^2, \quad F'=6 \tag{8}$$

$$\Delta\nu(7)=\left[\frac{0.8654}{(7A_{D_{5/2}}+\frac{7}{15}B_{D_{5/2}})}\right]\left[\frac{g_J eB}{4\pi m}\right]^2, \quad F'=7. \tag{9}$$

The numerical evaluations of the quadratic Zeeman shifts for the $^2S_{1/2}(m_F=0)$-$^2D_{5/2}(m'_F=0)$ transitions are shown in Table II. The uncertainty in the quadratic coefficients due to the uncertainty in the determination of the hyperfine constants is negligible.

For frequency standards applications, it is important to choose the level with the lowest quadratic shift and Table II shows that this is the $^2D_{5/2}F'=7$ level. Therefore, the preferred transition for a $^{87}Sr^+$-based optical frequency standard is $^2S_{1/2}(F=5, m_F=0)$-$^2D_{5/2}(F'=7, m_{F'}=0)$. This quadrupole transition is allowed when $F+F'$ is even, as in this case. The Zeeman structure of this line has been partially resolved in a magnetic field of 6.5 μT and the result is shown in Fig. 5. It is readily shown that the 55 Zeeman components are grouped into 19 sets, as shown in the calculated profile. However, in a 6.5-μT magnetic field and a 674-nm probe scan resolution of 1 kHz, the individual components within each group are not resolved. A higher magnetic field or narrower probe laser and smaller frequency step are required to achieve this and an improved probe laser system is being developed. The relative intensity of these groups depends on the orientation of the magnetic-field direction, linear 674-nm polarization and propagation direction. The calculation assumed that the magnetic field was 45° from the laser propagation direction and that the 674 nm polarization was 45° out of the plane defined by the laser beam and magnetic-field directions.

SUMMARY

The hyperfine structure of the $^2D_{5/2}$ level of the $^{87}Sr^+$ ion has been observed. The hyperfine constants and the isotope shift for the $^2S_{1/2}$-$^2D_{5/2}$ transition at 674 nm have been measured. This was achieved by measuring the frequency intervals of the $^2S_{1/2}(F=4)$-$^2D_{5/2}(F'=2, 3, 4, 5,$ and $6)$ and also the $^2S_{1/2}(F=5)$-$^2D_{5/2}(F'=3, 4, 5, 6,$ and $7)$ quadrupole transitions in a single cooled $^{87}Sr^+$ ion, confined in a Paul trap. The quadratic Zeeman shifts of the $m_F=0$ components have also been calculated, and the level with the lowest quadratic shift identified as $F'=7$. The preferred transition for an optical frequency standard is therefore $^2S_{1/2}(F=5, m_F=0)$-$^2D_{5/2}(F'=7, m_{F'}=0)$. The reproducibility of an optical frequency standard based on $^{87}Sr^+$ is likely to be limited by a number of factors, including the quadrupole shift. However, the contribution to the uncertainty from the Zeeman shift will be considerably smaller than for $^{88}Sr^+$, amounting to only 1 part in 10^{16} if the magnetic field is reduced to less than 0.1 μT.

ACKNOWLEDGMENTS

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