

Hyperfine structure of $^{151,153}\text{Eu}^+$ in the state $4f^7(8S^0)5d^9D_4^0$ by collinear laser-rf double resonance

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An apparatus has been constructed for laser-radio-frequency double-resonance studies of very-low-energy-ion beams with collinear laser-ion geometry. The ion energy of only 1–2 keV leads to extremely narrow linewidths (~ 60 kHz). This paper describes measurements of the hyperfine structure of $^{151}\text{Eu}^+$ and $^{153}\text{Eu}^+$ in the level $4f^7(8S^0)5d^9D_4^0$. The hyperfine constants A and B have been determined with a much higher accuracy than before. The present values of these constants (in MHz) are $A_{^{151}\text{Eu}^+} = 113.159(1)$, $B_{^{151}\text{Eu}^+} = -215.356(10)$, $A_{^{153}\text{Eu}^+} = -50.415(1)$, and $B_{^{153}\text{Eu}^+} = -548.357(10)$.

I. INTRODUCTION

The hyperfine structure of $^{151}\text{Eu}^+$ and $^{153}\text{Eu}^+$ in the ground state and a few excited states has been measured interferometrically by Krebs and Winkler,¹ Winkler,² and Guthöhrlein.³ More recently, Arnesen *et al.*⁴ and Dörschel *et al.*⁵ have determined the hyperfine constants A and B with some accuracy by laser-excited fluorescence of a fast (30–40 keV) ion beam. Although Arnesen *et al.* have taken advantage of the velocity bunching⁶ of a fast ion beam and a collinear laser beam, their linewidths are typically 100–150 MHz. Consequently, they were unable to resolve a number of the $^{151}\text{Eu}^+$ hyperfine components in typical optical lines. The errors for the hyperfine constants A and B reported by them are 1% and 8%, respectively.

In this paper we study the transitions from the hyperfine levels of the metastable state $4f^7(8S^0)5d^9D_4^0$ to those of the excited state $4f^7(8S^0_{7/2})6p_{3/2}$; $J=4$ in $^{151}\text{Eu}^+$ and $^{153}\text{Eu}^+$ by laser-excited fluorescence. We report the first high-precision measurement of the hyperfine intervals of the lower state ($5d^9D_4^0$) by the laser rf double resonance technique. The slow (1.35 keV) ion beam and the laser beam are collinear. The lower state and the upper state are designated according to the LS and $j-j$ coupling scheme, respectively.

We use a slow ion beam and a long rf region to achieve greater transit time and consequently obtain narrower line profiles. The rf region is about 50 cm long with an ion transit time of 12 μs at 1.35 keV. The Earth's magnetic field is shielded by a heat-treated permalloy enclosure. The typical linewidths obtained for these ions by laser rf double resonance are 58–70 kHz. For a monoenergetic beam, the linewidth⁷ for optimal rf power is $\sim 0.75/T$, where T is the transit time in the rf region. This suggests that the expected width is ~ 63 kHz which is consistent with the observation.

We have also recorded laser-excited fluorescence spectra of these slow ions and the lines are typically 50–75 MHz wide and very well resolved. An example of a fluorescence spectrum recorded with our apparatus is shown in Fig. 1.

II. EXPERIMENTAL PROCEDURE

The Eu^+ ions are produced in an arc-discharge source (Colutron), accelerated to 1.35 keV, focused by an einzel lens, and mass selected by a 90° magnet before they pass through the interaction regions. A tunable single mode dye laser (Coherent 599-21) pumped by an Ar^+ laser (Coherent CR-12) gives light at 604.951 nm which over-

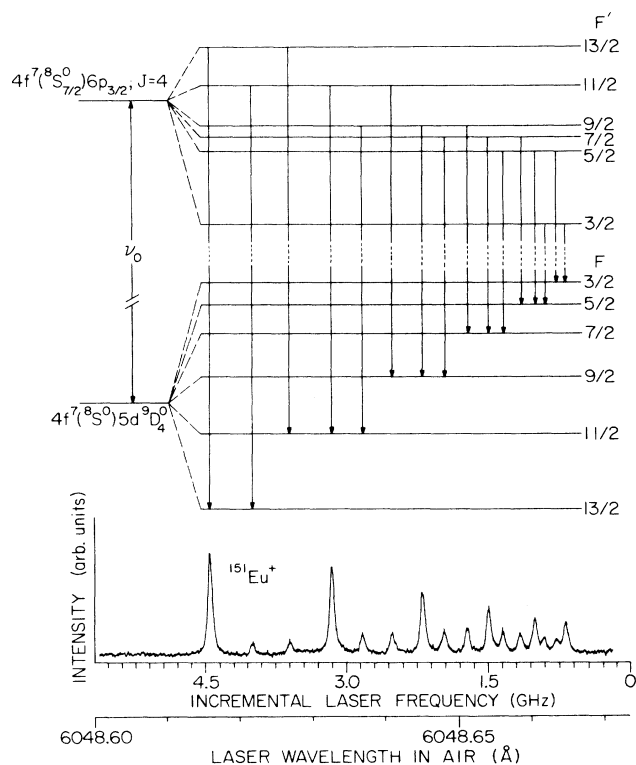


FIG. 1. Energy level diagram of the $6p_{3/2}$; $J=4$ and $5d^9D_4^0$ states of $^{151}\text{Eu}^+$ showing the hyperfine structure and the allowed transitions. A typical laser excited fluorescence spectrum taken with our apparatus is shown in the lower part.

laps the ion beam collinearly in the interaction regions over a length of approximately 1 m.

In the laser rf double-resonance method,⁸ the pump, the rf and the probe regions are conventionally designated as the *A*, *C*, and *B* regions respectively and are in that order along the ion beam (see Fig. 2). The typical ion beam and the laser intensity at the *A* region are 50–100 nA (for each Eu^+ isotope) and 30 mW, respectively. The *A* and *B* regions are identical in construction. Each consists of a Faraday cage about 10 cm long with gridded openings on the sides and collimating light baffles at the ends. Each cage can be biased to some convenient potential (typically –100 to –500 V) thus changing the ion beam velocity inside the cage. The rf power is applied at the region *C* through a triplate rf line⁹ where the ion beam interacts with the rf magnetic field. When the ions inside the cage are brought into resonance with the light by tuning the laser or the Faraday cage voltage, they are not resonant with the light elsewhere. In particular, they are far off resonance (~ 2.5 GHz) in the rf region.

For the laser-excited fluorescence measurements, only the region *A* is used. The Eu^+ ions in the metastable state $4f^7(8S^0)5d^9D_4^0$ are excited by 604.951 nm light to the $4f^7(8S^0_{7/2})6p_{3/2}$; $J=4$ level which decays primarily ($\lambda=397.2$ nm) to a lower state $4f^7(8S^0)6s^7S_3^0$. The fluorescence light is efficiently collected by a spherical reflector and an aspheric lens doublet and detected by a cooled photomultiplier with an appropriate interference

(pass-band) filter. The signal from the photomultiplier is amplified, discriminated and sent to a count-rate meter from which the analog signal is sent to a strip charge recorder. As the laser is scanned, the fluorescence spectrum is recorded. A typical hyperfine spectrum for the above transition ($^{151}\text{Eu}^+$, 604.951 nm) is shown in Fig. 1. Also shown in the figure is an energy-level diagram with the hyperfine components of the upper and lower states and the transitions between them in accordance with the selection rule $\Delta F=0, \pm 1$. The quantity ν_0 shown in the figure is the separation of the upper and the lower level with no hyperfine structure ($\nu_0 \cong 16\,525.68\text{ cm}^{-1}$). The linewidths are typically 50–70 MHz.

For the laser rf double-resonance measurements, first, the cage *A* is grounded and *B* is biased to some potential (say –300 V). The laser is tuned into resonance with one hyperfine transition which is observed as a peak in the photomultiplier signal at the *B* region. The cage at *A* is now biased to the same potential as *B* and the fluorescence at *B* decreases indicating the depletion of the particular lower hyperfine level due to the pumping at *A*. This optical pumping affects only the chosen hyperfine level and leaves others alone. Now, the rf is applied at the region *C*. If the rf is precisely equal to the splitting between the depleted hyperfine level and an adjacent level in the lower state, the depleted level will be repopulated to a certain extent according to the magnetic dipole hyperfine selection rule $\Delta F=0, \pm 1$. This increase in the population

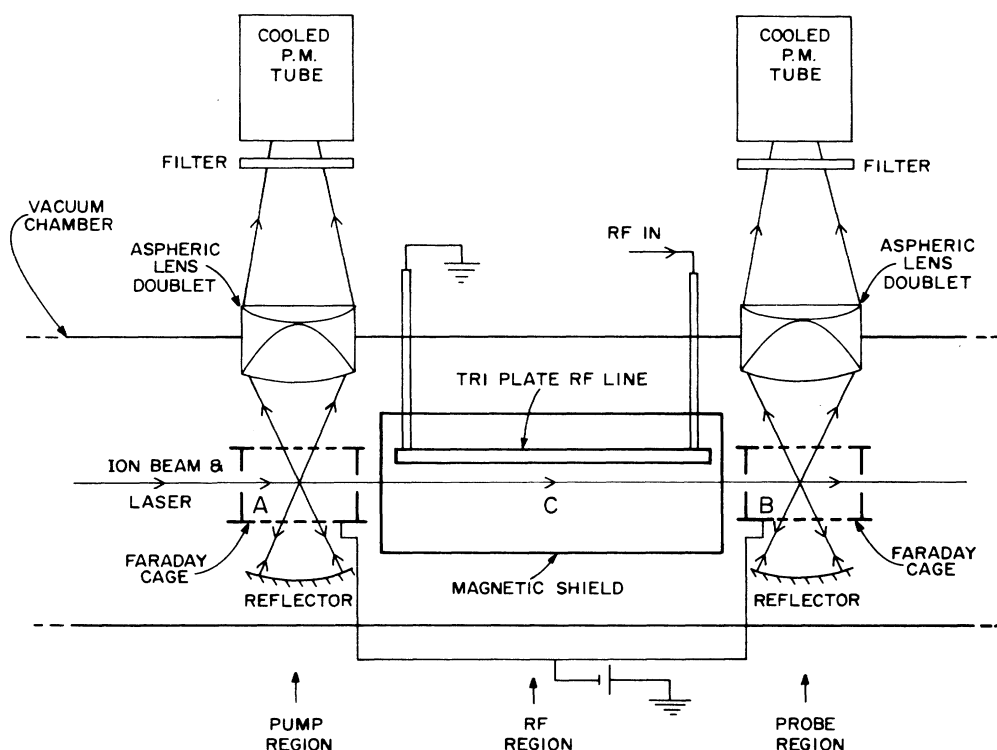


FIG. 2. Part of the collinear laser and slow ion beam apparatus showing the laser rf double-resonance region. The pump, the rf, and the probe regions are shown as *A*, *C*, and *B*, respectively.

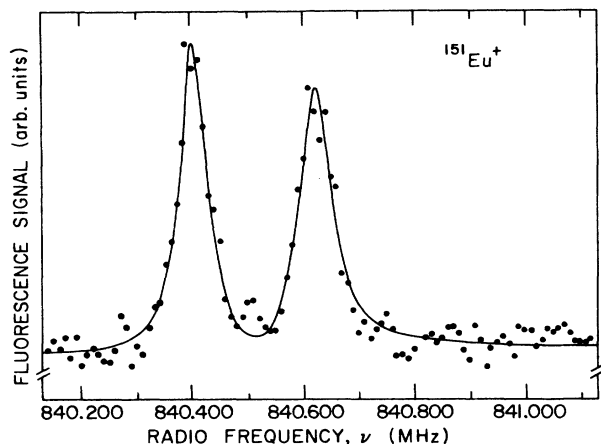


FIG. 3. A typical laser rf double-resonance spectrum for the transition $F = \frac{13}{2} \leftrightarrow F = \frac{11}{2}$ in the lower optical state $5d^9D_4^0$. The two peaks are the result of the interaction of the ion beam with the forward and reflected rf waves. The solid line is a Lorentz fit of the data points. The frequency of the hyperfine interval is taken as the average of the two peak positions.

in the C region will be reflected as an increase in the resonant fluorescence signal at B .

In an actual case, the rf is scanned over a short frequency interval around the anticipated resonance frequency. A typical laser-rf double resonance is shown in Fig. 3. The two peaks in the figure are due to the independent interaction of the forward and reflected rf waves of the shorted 50 ohm line with the ion beam and are Doppler shifted in opposite directions. The average of the two peak positions is taken as the frequency of the spacing between the two adjacent hyperfine levels. The separation of the peaks

(two times the Doppler shift) is consistent with the known ion velocity, and is proportional to the rf frequency.

III. RESULTS AND DISCUSSIONS

From the rf resonance measurements the hyperfine intervals for the lower optical state ($5d^9D_4^0$) are determined very accurately. Figure 3 shows the rf resonance for the transition $F = \frac{13}{2} \leftrightarrow F = \frac{11}{2}$. As mentioned earlier, the two peaks are due to the forward and reflected rf waves. A Lorentz fit of the data points is shown as the solid curve. The linewidth in this example is 62 kHz.

We have used only the magnetic dipole and electric quadrupole interaction terms¹⁰ involving the hyperfine constants A and B to analyze our data, and second-order hfs is ignored. With $I = \frac{5}{2}$ for Eu^+ and $J = 4$ for both the states ($5d^9D_4^0$ and $6p_{3/2}; J = 4$) each is split into $2I + 1$ or six sublevels (see Fig. 1). A theoretical spectrum, computed in terms of A and B values using the proper selection rules, is compared with the experimental laser-excited fluorescence spectrum. The hyperfine intervals of the upper and lower levels are expressed in terms of the corresponding A 's and B 's and their values are obtained from making a least-square fit to the measured intervals. The observed relative intensities are in reasonable agreement with those calculated for the transitions.

All the measured intervals from rf resonance are given in Table I. The values of A and B obtained from the best fit are given in Table II. The hyperfine intervals computed with these values of A 's and B 's are also given in Table I. The differences between the experimental and calculated hyperfine intervals are within experimental error. In the case of $^{151}\text{Eu}^+$ all the five intervals could be measured whereas for $^{153}\text{Eu}^+$ only two intervals could be determined by the rf resonance method due to the congestion of the optical hfs components and the consequent simultaneous pumping of adjacent hfs levels.

The errors quoted in Tables I and II are one standard deviation of the mean. A possible source of systematic er-

TABLE I. The hyperfine intervals of the $5d^9D_4^0$ state of $^{151}\text{Eu}^+$ and $^{153}\text{Eu}^+$ from rf resonance measurements.

| Mass No. | Transition $F + 1 \leftrightarrow F$ | Experimental hyperfine interval (MHz) ^a | Calculated hyperfine interval (MHz) | Difference (MHz) ^b |
|----------|---|--|-------------------------------------|-------------------------------|
| 151 | $\frac{13}{2} \leftrightarrow \frac{11}{2}$ | (-)840.522(3) | -840.522 | 0.000 |
| | $\frac{11}{2} \leftrightarrow \frac{9}{2}$ | (-)635.067(3) | -635.067 | 0.000 |
| | $\frac{9}{2} \leftrightarrow \frac{7}{2}$ | (-)467.684(4) | -467.684 | 0.000 |
| | $\frac{7}{2} \leftrightarrow \frac{5}{2}$ | (-)331.451(4) | -331.451 | 0.000 |
| | $\frac{5}{2} \leftrightarrow \frac{3}{2}$ | (-)219.445(5) | -219.445 | 0.000 |
| 153 | $\frac{13}{2} \leftrightarrow \frac{11}{2}$ | (-)595.021(3) | -595.022 | -0.001 |
| | $\frac{11}{2} \leftrightarrow \frac{9}{2}$ | (-)309.596(2) | -309.596 | 0.000 |
| | $\frac{9}{2} \leftrightarrow \frac{7}{2}$ | | -121.113 | |
| | $\frac{7}{2} \leftrightarrow \frac{5}{2}$ | | -11.945 | |
| | $\frac{5}{2} \leftrightarrow \frac{3}{2}$ | | +35.532 | |

^aThat the intervals have negative signs is determined from the laser fluorescence results; it implies that the level ordering is inverted from the "normal" order and arises from the fact that nuclear dipole moments of ^{151}Eu and ^{153}Eu are negative.

^bThat all residuals are zero may indicate that the experimental precision is better than that indicated in column 3.

TABLE II. Hyperfine constants A and B for the $5d\ ^9D_4^0$ and $6p_{3/2}; J=4$ levels of $^{151}\text{Eu}^+$ and $^{153}\text{Eu}^+$.

| Level | A_{151} (MHz) | B_{151} (MHz) | A_{153} (MHz) | B_{153} (MHz) | Reference |
|-----------------|--------------------|--------------------|--------------------|--------------------|-----------|
| $5d\ ^9D_4^0$ | -113.159(1) | -215.356(10) | -50.415(1) | -548.357(10) | a |
| | -113.6(3) | -214(3) | -52.7(3.0) | -521(29) | b |
| | -115.3(8) | -203(17) | -51.9(3.9) | -547(56) | c |
| $6p_{3/2}; J=4$ | 57.1(4) | 147(6) | 25.5(3.0) | 393(32) | b |
| | 57.3(7) | 156(15) | 23.2(3.3) | 439(52) | c |

^aPresent work: laser rf double resonance.

^bPresent work: laser-excited fluorescence.

^cArnesen *et al.*, 1981.

ror is the light shift¹¹ of the resonant frequency due to the presence of the detuned laser field in the rf region. We have varied the velocity of the ion beam (i.e., the extent of detuning) in the Faraday cages widely and measured the rf resonance. There is no change in the resonant frequency indicating a very small effect of light shift (less than our experimental uncertainty of about 3 kHz).

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