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Hyperfine structure of some excited states of $^{133}\text{Cs}^+$ by collinear laser-ion beam spectroscopy

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(Received 13 March 1989)

Hyperfine structure of a few excited states of $^{133}\text{Cs}^+$ in the $5p^55d$ and $5p^56p$ configurations has been measured by laser-induced fluorescence with a collinear laser and slow-ion-beam (~ 1.5 keV) apparatus. The hyperfine spectrum for the optical transition at 612.86 nm has been recorded for the first time. The full width at half maximum of the laser-induced fluorescence spectra recorded with our apparatus is typically 45–75 MHz. The magnetic dipole (A) and electric quadrupole (B) hyperfine constants have been determined for the two lower levels $5p^55d\ ^3P_2$, 3F_4 and the two upper levels $5p^56p\ \frac{3}{2}[\frac{3}{2}]_1$, $\frac{3}{2}[\frac{5}{2}]_3$ at energies 112 803.99, 112 245.44, 129 998.63, and 129 116.56 cm^{-1} , respectively, above the ground state, $5p^6\ ^1S_0$.

INTRODUCTION

Before lasers were invented, the hyperfine structure of neutral atoms and ions was studied in the emission spectra with interferometers. In the 1930s Rabi *et al.*¹ developed the atomic beam magnetic resonance (ABMR) technique for neutral beams, and accurate hyperfine-structure (hfs) measurements were made. More recently, with the availability of high-power narrow-band lasers in the 1970s, it has been possible to attain very high spectral resolution to explore sub-Doppler regions.² Hyperfine transitions in an ion beam or neutral beam are excited by light of appropriate wavelength from a narrow-band tunable laser and its signature is recorded by observing the resulting fluorescence from the excited level. Very high resolution and precision in hfs measurements have become possible in laser-rf double-resonance spectroscopy.^{3,4}

In 1931, Kopfermann⁵ made accurate hyperfine-structure measurements of $^{133}\text{Cs}^+$, which indicated that the nuclear spin of ^{133}Cs was $\frac{7}{2}$. Sawyer and associates^{6,7} made extensive measurements of the spectra of Cs^+ , which resulted in a proper classification of the energy levels.

The collinear laser and fast-ion-beam geometry has been used for high-resolution hfs measurements for over a decade now. It takes advantage of kinematic compression resulting in a narrow velocity spread of the ion beam due to the acceleration. This velocity bunching was pro-

posed by Kaufman⁸ and almost immediately demonstrated by Wing *et al.*⁹ The velocity bunching resulted in a narrow Doppler profile so that hfs measurements were possible.

The fast ion beam and laser technique for the measurement of hfs of Cs^+ has been used by Alvarez and co-workers^{10,11} in a crossed ion beam and laser geometry with a 35–40-keV Cs^+ ion beam. Bengtson *et al.*¹² have used a collinear laser-ion beam at similar acceleration to study hfs of Cs^+ . In all these studies, the FWHM of the spectra are around 300 MHz and the hyperfine lines are poorly resolved. We have recently built a collinear laser and slow-ion-beam apparatus for laser-rf double-resonance spectroscopy.¹³ The laser-induced fluorescence spectra recorded with our apparatus have FWHM of about 45–75 MHz and the hyperfine components are very well resolved. We have recorded the hyperfine spectrum of $^{133}\text{Cs}^+$ at 612.86 nm for the first time with all its 15 transitions clearly resolved. In addition, we have measured the hyperfine structure for transitions at wavelengths 518.42 and 592.57 nm and made accurate determinations of the hyperfine constants A and B for four excited levels of $^{133}\text{Cs}^+$.

ENERGY LEVELS OF Cs^+

The Cs^+ ion is isoelectronic with neutral Xe and thus has a rare-gas-like structure. The ground-state configuration of Cs^+ is $5p^6\ ^1S_0$ and the singly excited states are of the configuration $5p^5nl$. In the 1920s, the

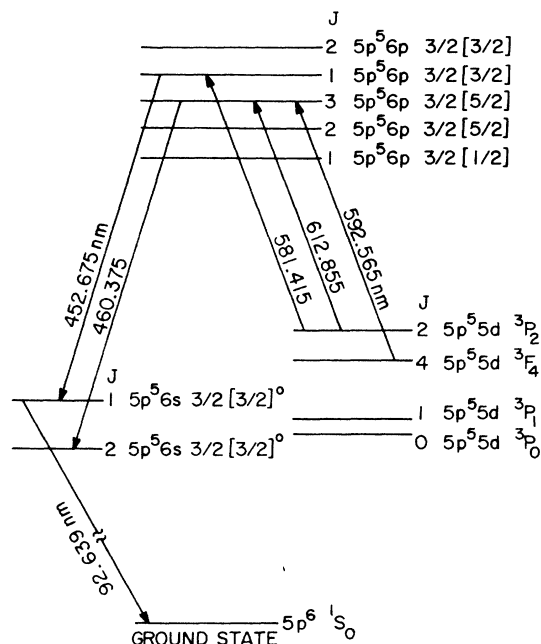


FIG. 1. Partial energy-level diagram of Cs^+ without the hyperfine splitting, showing the ground state and a few excited states of the ion. See Ref. 18 for the spectroscopic notations.

spectra of Cs^+ were observed by spark excitation and attempts were made by Sommer¹⁴ and by Laporte *et al.*¹⁵ to identify the energy levels of Cs^+ . Wheatley and Sawyer⁶ reported 79 excited levels of Cs^+ , of which spectral term designations for 43 levels were assigned in the jK coupling scheme by Moore.¹⁶ In the mid-1970s, Sansonetti¹⁷ made a thorough study of the energy levels,

which resulted in rejection and incorporation of new levels of Cs^+ . Reader¹⁸ reinterpreted the observed energy levels of Cs^+ reported by Wheatley and Sawyer through Hartree-Fock calculations and the published Zeeman-effect data of Zelikina and Semenov.¹⁹ Sansonetti and Andrew²⁰ have made a comprehensive analysis of the energy levels of Cs^+ and classified nearly 1700 lines, which are similar to Reader's. A partial energy-level diagram of Cs^+ based on the notation of Reader¹⁸ and relevant to our measurements is given in Fig. 1.

EXPERIMENT

Method and apparatus

When the Doppler-shifted frequency of the laser becomes equal to a transition frequency of the ion, the laser photon can be resonantly absorbed by the ion resulting in an excitation to a higher level, which subsequently decays to lower levels giving fluorescence. This fluorescence from the upper level contains the hyperfine splitting information for the transition.

The details of our apparatus have been given in Ref. 13. Briefly, the $^{133}\text{Cs}^+$ ions are produced in a colutron ion source, accelerated to 1.35 keV, and focused by an einzel lens and electrostatic quadrupole doublet lens, mass selected by a 90° magnet before entering the interaction region. Laser light from a tunable single-mode ring-dye laser (Coherent 699-21) pumped by an Ar^+ laser (Coherent CR-12) enters the chamber and superimposes collinearly with the ion beam in the interaction region. The interaction region is a cylindrical biased Faraday cage about 2 in. long through which the laser and the ion beam pass coaxially. The ions can be Doppler-tuned into resonance with a particular hyperfine transition inside the

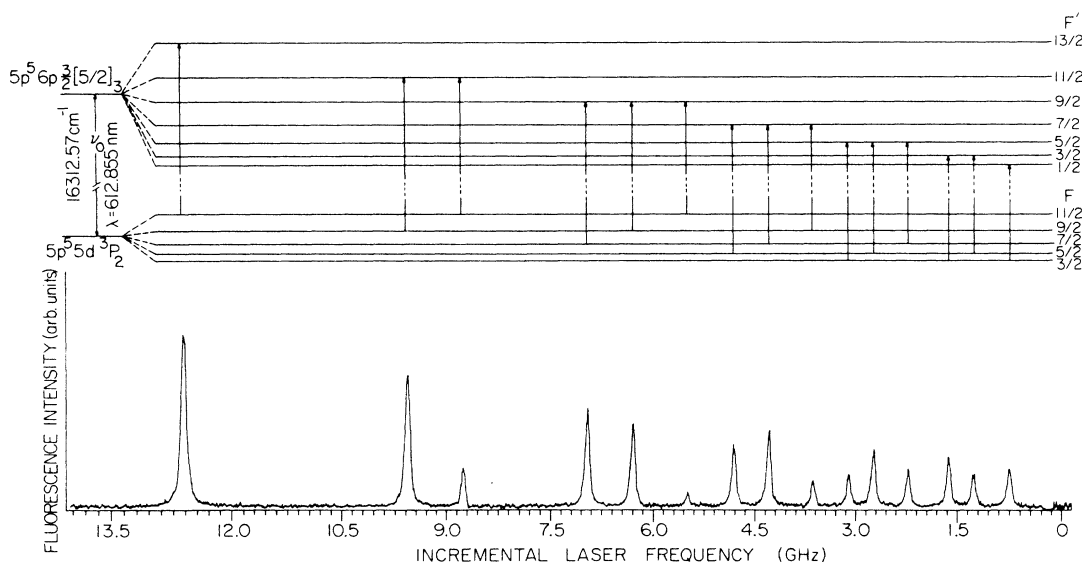


FIG. 2. Laser-induced fluorescence spectra of $^{133}\text{Cs}^+$ for the hyperfine transitions $5p^5 5d \ ^3P_2 \rightarrow 5p^5 6p \ \frac{3}{2} [\frac{5}{2}]_3$ at 612.855 nm. The upper part shows the hf splitting of the energy levels and the allowed hf transitions. The lower part is an actual recorded spectrum. The linewidths are typically 45–75 MHz and all the 15 allowed transitions are clearly resolved. The ion energy is 1.5 keV in the Faraday cage region.

TABLE I. Hyperfine constants for $^{133}\text{Cs}^+$.

Levels	Excitation energy (cm^{-1})	J	λ (nm)	A (MHz)	B (MHz)	Remarks	
$5p^56p \frac{3}{2}[\frac{3}{2}]$	129 998.63	1	581.42	786.6±0.7	7.3±5.0	This work	
				795			Kopfermann (Ref. 5)
				793±2	-20±20	Alvarez <i>et al.</i> (Ref. 11)	
				786.6±1.5		Sansonetti and Andrew (Ref. 20)	
$5p^56p \frac{3}{2}[\frac{5}{2}]$	129 116.56	3	592.57 612.86	588.8±0.5	12.3±8.7	This work	
				589.8±0.3	19.4±5.3	This work	
				585		Kopfermann (Ref. 5)	
				593±3	50±30	Alvarez <i>et al.</i> (Ref. 11)	
			589.2±3.3		Sansonetti and Andrew (Ref. 20)		
$5p^55d \ ^3P_2$	112 803.99	2	581.42 612.86	145.2±0.7	-8±7	This work	
				145.9±0.3	1.9±1.4	This work	
				147±5	-24±51	Alvarez <i>et al.</i> (Ref. 10)	
				147±1	5±6	Bengtson <i>et al.</i> (Ref. 12)	
			145.5±1.5		Sansonetti and Andrew (Ref. 20)		
$5p^55d \ ^3F_4$	112 245.44	4	592.57	370.4±0.4	-4±9	This work	
				376±3	-50±50	Alvarez <i>et al.</i> (Ref. 11)	
				375.0±18		Sansonetti and Andrew (Ref. 20)	

Faraday cage by either tuning the laser frequency or the bias voltage on the cage. In our experiment, the bias voltage on the Faraday cage was kept fixed around -100 to -300 V and the laser frequency was changed. The fluorescence from the cage following an optical excitation is collected by a spherical mirror and an aspherical lens doublet onto a cooled (-15°C) photomultiplier tube (Thorn EMI model 9658R) with appropriate filters. The signal from the photomultiplier tube is processed with standard electronics and sent to a strip chart recorder. Frequency markers on the same chart recorder were provided from a portion of the laser light going through a confocal Fabry-Perot interferometer of free spectral range 150 MHz. The ion beam current and the laser intensity were 0.4 μA and 100 mW, respectively.

Measurements

The hyperfine structure of two lower levels $5p^55d \ ^3P_2, \ ^3F_4$ and two upper levels $5p^56p \frac{3}{2}[\frac{3}{2}]_1, \frac{3}{2}[\frac{5}{2}]_3$ of $^{133}\text{Cs}^+$ at wave numbers 112 803.99, 112 245.44, 129 998.63, and 129 116.56 cm^{-1} , respectively, above the ground state $5p^61S_0$ (see Fig. 1) have been measured. These levels are about 14 eV above the ground state and are easily produced in sizable population in the ion source.

The hyperfine spectrum for the transition $5p^55d \ ^3P_2 \rightarrow 5p^56p \frac{3}{2}[\frac{3}{2}]_3$ at wavelength 612.855 nm is shown in Fig. 2. The nuclear spin for $^{133}\text{Cs}^+$ is $I = \frac{7}{2}$. With $J = 3$, there are $2J + 1 = 7$ hyperfine (hf) levels for the upper level. Also, the lower level with $J = 2$ splits into 5 hf levels. Following dipole selection rule $\Delta F = 0, \pm 1$, there are 15 allowed transitions as shown in the upper part of Fig. 2. The lower part of the figure shows the actual observed spectrum where all the 15 hyperfine

components have been clearly resolved and identified. The FWHM of these lines are about 50 MHz.

The hyperfine structures for the transitions $5p^55d \ ^3F_4 \rightarrow 5p^56p \frac{3}{2}[\frac{5}{2}]_3$ at wavelength 592.565 nm and $5p^55d \ ^3P_2 \rightarrow 5p^56p \frac{3}{2}[\frac{5}{2}]_1$ at wavelength 581.415 nm were also measured.

RESULTS

The magnetic dipole (A) and electric quadrupole (B) hfs interaction constants were determined from the spectra using the standard first-order theory.^{21,22} The A 's and B 's determined in this fashion are given in Table I where previous values reported by other groups are also compiled for comparison.

DISCUSSION

The hyperfine spectra of $^{133}\text{Cs}^+$ for three optical transitions at wavelengths 581.42, 592.57, and 612.86 nm have been measured by a collinear laser and slow-ion-beam apparatus. This is the first time where the 612.86 nm transition has been observed with all the hyperfine components clearly resolved. The FWHM of the laser-induced fluorescence spectra are typically 45–70 MHz, which is at least a factor of 5 better than some earlier measurements with laser by Alvarez *et al.*¹¹ and Bengtson *et al.*¹² We have demonstrated that rather good resolution is possible with a relatively slow ion beam if fluctuations in the acceleration potential are small enough.

The B constants for the two upper levels in the $5p^56p$ configurations (see Table I) are indicative of small but finite quadrupole interaction. The B 's for the lower two levels in the $5p^55d$ configuration are small, too. But because of the large uncertainties, they may suggest vanish-

ing quadrupolar interaction. The $5p^55d$ levels are strongly perturbed by the proximity of the $5p^56s$ configuration and it is difficult to estimate the configuration mixing. This may also suggest why the B 's have large uncertainties.

Also included in the table are the results of early work by Kopfermann.⁵ Sansonetti and Andrew²⁰ have measured 285 levels of $^{133}\text{Cs}^+$ by Fourier-transform spectroscopy,

but they have reported only the values of magnetic dipole hf constants (A). Their values are also given in Table I.

ACKNOWLEDGMENTS

This research was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, under Contract No. W-31-109-Eng-38.

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