Lifetime measurement of the $T=23\,537\ \mathrm{cm}^{-1}$, $J=9/2$ odd-parity level in Nd II using time-resolved collinear fast-beam laser spectroscopy

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Time-resolved collinear fast-beam laser spectroscopy has been used to measure the lifetime of the $T=23\,537\ \mathrm{cm}^{-1}$, $J=9/2$ odd-parity [abbreviated as $(23\,537)^{\circ}_{9/2}$] level in Nd II. Nanosecond pulsed excitation was achieved with the Doppler switching technique. The fast ions were selectively excited from the metastable $4f^4 5d\ ^6K_{9/2}$ level to the $(23\,537)^{\circ}_{9/2}$ level by the light of a cw dye laser. The decay of the resonance fluorescence photons emitted in the transition to the ground $4f^4 6s\ ^6I_{7/2}$ level, as a function of the flight distance, was observed. The decay length was scaled into the decay time by the ion velocity, which was determined using the optical resonance itself. The lifetime is 27.7(9) ns.

I. INTRODUCTION

Atomic transition probability data are needed in several applied-physics fields, such as astrophysics,[1] laser technology,[2–4] and controlled thermonuclear fusion research,[5] etc. Owing to the difficulties in the theoretical calculations, especially for heavier elements, experimental measurements play a major role in obtaining them. Corliss and Bozman[6] have measured and compiled transition probabilities of a wide variety of elements based on arc spectra. Although the absolute values of the transition probabilities in this compilation may be wrong by an order of magnitude, especially for the rare-earth elements, the relative values for transitions from a common upper level are often more accurate. Atomic-level lifetime data are needed to put the relative transition probabilities on an improved absolute scale.

Atomic-level lifetimes can be measured using various experimental methods, e.g., Hanle effect,[7] multichannel method of delayed coincidence with electron-impact excitation,[8] beam foil,[9] laser excitation of fast beams.[10] Among these methods, the laser excitation of fast beams is the most accurate (with accuracy up to 0.1%) and reliable method since the selective excitation avoids cascading. In particular it is a powerful tool for lifetime measurements of elements whose spectra are very complex, e.g., the rare-earth elements.

In the present paper, we report our work on the lifetime measurement of the $T=23\,537\ \mathrm{cm}^{-1}$, $J=9/2$ odd-parity [abbreviated $(23\,537)^{\circ}_{9/2}$] level in Nd II using the collinear fast-beam laser spectroscopy method.

II. EXPERIMENTAL PRINCIPLE

The principle of the time-resolved collinear fast-beam laser spectroscopy method using Doppler switching and time-of-flight techniques has been described by Winter and Gaillard[11] and later by Ceyzeriat *et al.*[12] In the Doppler switching concept, the ions are brought suddenly in and out of resonance with the applied laser light by in-flight changes of the ion velocity. This is easily achieved by applying a localized electric field along the common beam path. With a large electric field gradient, pulsed excitation in the nanosecond range can be achieved. The decay of the fluorescence is then monitored as a function of the flight distance after the resonant excitation region.

III. EXPERIMENTAL ARRANGEMENT

The experimental arrangement is shown in Fig. 1. A beam of isotopically pure singly-charged ^{142}Nd ions with energy of 30 keV was supplied by a isotope separator (Danfysik 1080-30) equipped with a hollow cathode discharge ion source (Danfysik 911A). The beam entered a target chamber. A Farady cup at the end of the ion beam line was used to collect the momentum-analyzed ions deflected by an externally applied magnetic field. The collected charge produced a signal for normalization purposes. The beam current at the exit slit of the isotope separator can be as high as tens of microamperes when the ion source is charged with neodymium chloride. In order to avoid collisional quenching, collision-induced ion beam excitation, and radiation trapping, the background pressure in the target chamber was maintained below 2×10^{-6} Torr by a 450 l/s turbomolecular pump.

A laser beam was extracted from a cw ring dye laser (Coherent 699-21) pumped by an argon-ion laser (Coherent Innova-20) and split into four beams. The first beam was guided through the target chamber, where it collinearly interacted with the ion beam. The second beam entered a prism monochromator for the purpose of

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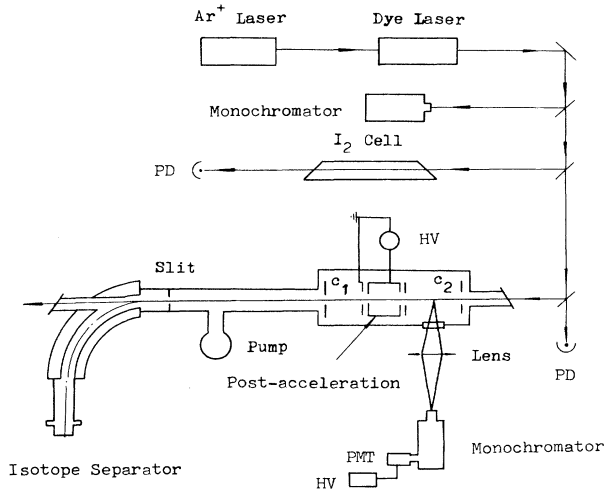

FIG. 1. Schematic diagram of the experimental arrangement.

the rough readout of the laser wavelength. The third beam was guided through a room-temperature iodine vapor cell and detected by a photodiode for the purpose of absolute laser wave-number calibration. The fourth beam entered another photodiode and was detected for normalization purposes. With the Rhodamine 6G dye, the dye laser can cover an optical range of 5600–6250 Å. When the dye laser is operated in the single-frequency mode, the bandwidth of the laser is below 1 MHz, and it can scan automatically over a maximum range of 30 GHz under a control unit.

In Fig. 1 the collimators C_1 and C_2 have apertures of 3 mm in diameter and are spaced 60 cm apart. These collimators, together with the exit slit of the isotope separator, were used to guarantee that the two beams remained collinear to within 2 mrad. The two outer disks of a set of postacceleration and postdeceleration electrodes were electrically grounded, while the 10-cm-long (10 decay lengths for $\tau=50$ ns) high-potential tube was held at a positive potential of 600 V. The whole set of electrodes was mounted on the movable platform which was driven by a stepping motor with a step size of 0.250 mm.

Fluorescence photons were analyzed with a grating monochromator viewing perpendicular to the beam axis through an 11-cm focal length lens which imaged the 0.25-mm entrance slit of the monochromator in a 1:1 ratio on the ion beam path. The detection system included a photomultiplier tube (EMI 9789QB) and typical single-photon-counting electronics.

To minimize the background light, mainly the scattered laser light, the inner and outer surfaces of the set of postdeceleration electrodes and the inner surfaces of the target chamber were blackened, and the fluorescence photon observation and detection systems were light-shielded. As a result, the scattered laser light count rate was reduced by an order of magnitude, and was comparable to the photomultiplier tube (PMT) dark-current background count rate.

With the arrangement described above, lifetimes ranging from a few to several hundred nanoseconds can be measured. The lower limit is set by the time width of the instrumental window function, while the upper limit is set by the maximum movable length of the set of postdeceleration electrodes (10 cm).

IV. ION VELOCITY

One of the basic problems of the lifetime measurements using the fast-beam laser spectroscopy method is the accurate determination of the ion velocity. Andrä *et al.*[10] used an electrostatic energy analyzer to determine the ion velocity, while several authors[11–13] used the optical resonance itself. It was found that final accuracy in the $\pm 0.1\%$ range can be achieved by the optical resonance method, which is competitive with method based on an electrostatic energy analyzer.

We have measured the ion velocity using the optical resonance itself. The excitation and detection channels are shown in Fig. 2. The ions are selectively excited from the metastable $4f^45d\ ^6K_{9/2}$ level to the $(23\,537)^\circ_{9/2}$ level using laser radiation of wavelength 5706 Å (the Doppler shifted wavelength), while the fluorescence photons emitted in transition to the ground $4f^46s\ ^6I_{7/2}$ level are observed at a wavelength of 4247 Å. It was found that there was sufficient content of the metastable ions in the neodymium ion beam extracted from the ion source for the experiment to be successful.

During the measurement, fluorescence photons were observed through one of the four holes in the high-potential tube of the set of postdeceleration electrodes. Signals from the dye laser light transmitted through the iodine vapor cell and from the PMT were used as gate inputs to two multichannel analyzers (MCA). This allowed the ramp voltage from the dye laser control unit to be digitized and stored in the two MCA's, as well as allowing the dye laser to be scanned quickly, tens of times, over a range of 30 GHz centered at 5706 Å. The resonant excitation spectrum of the transition $4f^45d\ ^6K_{9/2}-(23\,537)^\circ_{9/2}$ is shown in Fig. 3.

By interpolation using the iodine reference spectrum, the resonant excitation wave number in the laboratory

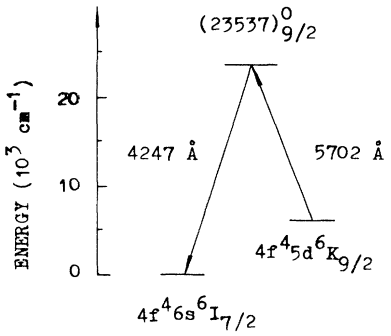

FIG. 2. Schematic energy level diagram of Nd II showing the transitions investigated. The transition $4f^45d\ ^6K_{9/2}-(23\,537)^\circ_{9/2}$ (5702 Å) is used as the excitation channel, while the transition $(23\,537)^\circ_{9/2}-4f^46s\ ^6I_{7/2}$ (4247 Å) is used as the detection channel.

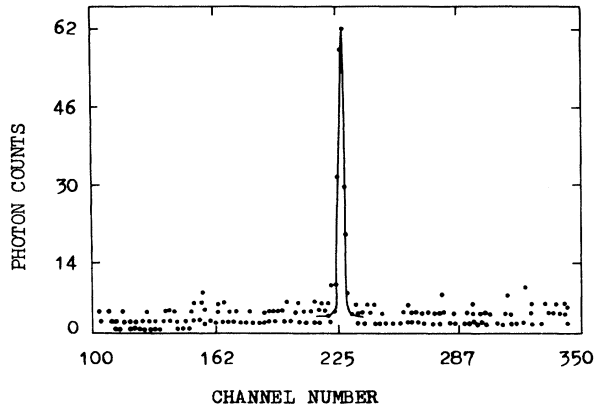

FIG. 3. The resonant excitation spectrum of the transition $4f^45d\ ^6K_{9/2}-(23\,537)^{o}_{9/2}$ in Nd II.

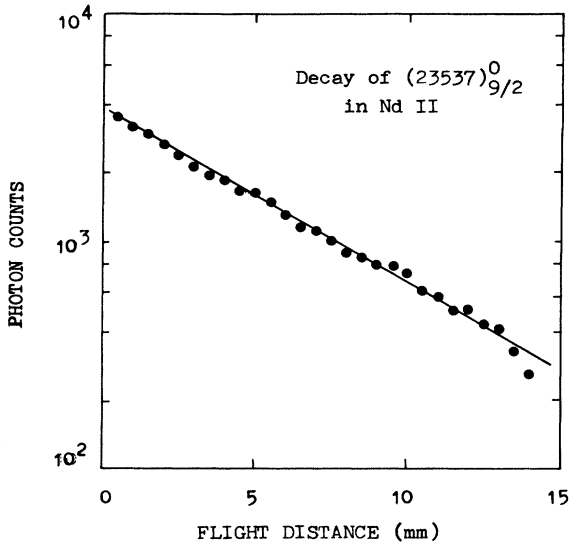

FIG. 4. A semilogarithmic plot of the fluorescence photon counts as a function of the flight distance after subtraction of the background. The solid line is a one-exponential least-squares fit.

frame was obtained,

$$\tilde{\nu}_l = 17\,520.152(3)\ \mathrm{cm}^{-1}\ . \tag{1}$$

The quoted error is mainly due to the uncertainty in the iodine wave number[14] (0.002 cm^{-1}), and the uncertainties in determining the centers of the iodine peaks (0.001 cm^{-1}) and the optical resonance (0.0005 cm^{-1}). Blaise *et al.*[15] and Ahmad and Saksena[16] have measured the corresponding transition wave number in the ion frame:

$$\tilde{\nu}_a = 17\,532.067(8)\ \mathrm{cm}^{-1}\ . \tag{2}$$

The Doppler formula can be used to determine the ion beam velocity inside the electrode assembly

$$\beta = \frac{(\tilde{\nu}_a)^2-(\tilde{\nu}_l)^2}{(\tilde{\nu}_a)^2+(\tilde{\nu}_l)^2}\ , \tag{3}$$

thus the ion energy corresponding to the exact resonance has been found,

$$E = 30.55(4)\ \mathrm{keV}\ . \tag{4}$$

The ion energy outside the electrode assembly is then

$$E_0 = E + eV$$
$$= 31.15(4)\ \mathrm{keV}\ , \tag{5}$$

where V is the voltage applied to the postdeceleration electrodes. This corresponds to an ion velocity in the field-free region of

$$v = 2.0581(13)\times 10^5\ \mathrm{m/s}\ . \tag{6}$$

V. LIFETIME AND DISCUSSION

A lifetime measurement was carried out by moving the set of postdeceleration electrodes upstream of the observation point of the transverse fluoresence photon optics. During the motion of the set of electrodes, we have measured two kinds of normalized background counts, i.e., the normalized residual-gas-induced fluorescence photon counts and the normalized scattered laser light counts. The former background was found to remain essentially constant with distance from the electrode assembly. The mean count rate is about 8 counts/s for an ion beam current of 500 nA. The latter background was found to decrease quickly from about 11 to 4 counts/s and then remain constant for a dye laser power of 100 mW measured at the entrance of the target chamber. This can be easily understood: near the outer grounded disk of the electrode assembly, the scattered laser light from the aperture of the disk can go directly into the fluorescence observation system, so a relatively high background is expected. When the scattering aperture goes beyond the limited observation region, the scattered laser light can only get into the observation system through a multiscattering process, hence the background is expected to be relatively weak, and to remain constant with the increasing distance. Consequently, the total normalized background downstream from the set of electrodes remains essentially constant except for the first 2–3-mm region behind the electrode assembly.

Figure 4 shows the decay of the fluorescence photon intensity. The maximum count rate is about 170 counts/s for an ion beam current of 500 nA and a dye laser power of 100 mW. The decay data were least-

TABLE I. Measured lifetime of the $(23\,537)^{o}_{9/2}$ level in Nd II.

Measurement	Lifetime (ns)
Present	27.7(9)
Ward *et al.*[a]	25.5(8)
Corliss and Bozman[b]	115

[a]Reference 13.
[b]Reference 6.

squares fitted with an analytic function of the form

$$y = a_1 \exp(-x/a_2) + a_3 , \quad (7)$$

where x and y corresponds to the flight distance and the normalized resonance fluorescence photon counts, respectively. Since the nearest observation point is 2 mm away from the resonant excitation region (corresponding to a flight time of 10 ns), and the time width of the instrumental window function is about 3 ns, the window function effect has been eliminated.

Table I contains the result of the present measurement and those of earlier measurements. The measurements of Ward *et al.*[13] have been carried out using the crossed fast-beam laser spectroscopy method, while those of Corliss and Bozman[6] were based on arc spectra. It can be seen that our result is in fair agreement with that of Ward *et al.*[13]

The main error of the present result comes from the uncertainty ($\pm 3\%$) in the fit parameter a_2. This uncertainty originates from the limited counting statistics and from the change in relative overlap of the laser and ion beams due to deviations from a perfect collinear geometry. In the present experiment, the data acquisition time was limited by the isotope separator operation time, e.g., a few hours with good beam stability when the ion source was charged with neodymium chloride; and the two beams were guaranteed collinear only to within 2 mrad. It can be expected that the accuracy of the measured lifetime could be enhanced if the data acquisition time were increased and the beam parallelism improved.

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