CONF-8708164--3

DE88 002985

High-precision Hyperfine Structure Measurement in Slow Atomic Ion Beams by Collinear Laser-rf Double Resonance*

Amarjit Sen, William J. Childs and Leonard S. Goodman

Physics Division, Argonne National Laboratory, Argonne, IL, 60439-4843.

A new collinear laser-ion beam apparatus for slow ions (1 - 1.5 keV) has been built for measuring the hyperfine structure of metastable levels of ions with laser-rf double resonance technique. Narrow linewidths of ~ 60 kHz (FWHM) have been observed for the first time in such systems. As a first application the hyperfine structure of the $4f^7(^8S^0)5d\ ^9D_J^0$ metastable levels of $^{151,153}Eu^+$ has been measured with high precision.

* This research was supported by the U.S. Department of Energy , Office of Basic Energy Sciences, under Contract W-31-109-Eng-38.

1. INTRODUCTION:

Hyperfine structure of neutral atoms and ions have traditionally been studied in the emission spectra with the help of interferometers as early as the later part of the last century. More recently, with the advent of high power narrow band tunable lasers it has been possible to attain very high spectral resolution and hence explore the sub-Doppler regime. By combining a laser beam with an ion beam the hyperfine structure of these ions can be measured by laser-induced fluorescence.

The arrangement of collinear laser and fast ion beams has been used over a decade to achieve high resolution measurements of hyperfine structure. The method takes advantage of velocity bunching or kinematic compression which was originally proposed by Kaufman [1] and almost simultaneously realized experimentally by Wing et al. [2]. It has been shown [1] that by accelerating a beam of ions to a few tens of keV, the velocity spread can be reduced by about 3 orders of magnitude over the thermal spread. Consequently, the Doppler profile of the spectral lines can be made quite narrow so as to observe the hyperfine structure.

The first order Doppler width (ν_D) at a frequency ν_0 associated with the velocity spread (δv) of the ions is given by

$$\frac{\delta\nu_D}{\nu_0} = \frac{\delta v}{c} = \left\{ \frac{e}{2mc^2} \right\}^{1/2} \frac{\delta U}{U^{1/2}}$$

where e is the electronic charge, m is the mass of an electron, c is the speed of light and U is acceleration potential. Therefore, it is imperative that not only the acceleration voltage be made high but also the fluctuations of the high

voltage be made small in order to reduce the Doppler width. Typical linewidths observed in the laser-induced fluorescence spectra are 30 - 150 MHz (FWHM) at 10 - 100 keV ion energy. With this the hyperfine structure of the ions can be measured to an precision of a few MHz.

Further improvement in the precision by about 3 orders of magnitude has been attained by laser-rf double resonance technique [3] which is similar to the atomic beam magnetic resonance (ABMR) technique developed by Rabi et al. [4].

We have recently built a collinear laser-ion beam apparatus [5] for slow ions (~ 1 - 1.5 keV) with good stability of the acceleration potential. With this we have achieved linewidths of 45 - 75 MHz (FWHM) in the laser-induced fluorescence spectra, which are better than that obtained in some of the fast beam [6] machines. With the laser-rf double resonance technique we have observed [7] the narrowest linewidth (FWHM ~ 60 kHz) yet seen in similar apparatuses. The rf resonance linewidths are transit-time limited. Therefore, by choosing slower ion beam (low acceleration potential) the transit time in the rf region is made longer which translates into extremely narrow linewidths. Furthermore, by using a tri-plate rf line [8] the rf power required is significantly low (typical rf power used is ~ 0.06 W).

2. EXPERIMENTAL METHOD:

2.1 Laser-induced Fluorescence:

An ion moving with a velocity v observes the laser frequency ν_L in its rest frame as (Doppler effect)

$$\nu_{ob} = \nu_L(1-\beta\mathrm{Cos}\theta)(1-\beta^2)^{-1/2}$$

where $\beta = v/c$ and θ is the angle between the direction of propagation of the laser and the ion velocity. For small β,

$$\nu_{ob}\pm = \nu_L(1\mp\beta)$$

where ν_{ob}^{+} and ν_{ob}^{-} are the frequencies seen by the ions when the laser and the ion beam are collinear and propagating in the same and opposite directions, respectively. When ν_{ob}^{*} equals a transition frequency of the ion, the laser photon is resonantly absorbed by the ion causing excitation to the higher level which subsequently decays to lower levels giving fluorescence. (See Fig. 1).

2.2 Laser-rf Double Resonance:

In the laser-rf double resonance method the pump, the rf and the probe regions are designated as A, C and B respectively following the nomenclature of ABMR and are shown in Fig. 2 and Fig. 3. The A and B regions are two identical fluorescence regions where the ions can "see" the laser. By tuning either the ion velocity or the laser frequency the ions can be made to absorb the laser photon at A causing (say) a hyperfine transition $F \rightarrow F'$, whereby the population of the level F is depleted. The level may be repopulated by transition from a neighboring hyperfine level, $F \pm 1 \rightarrow F$ with an appropriate rf field at region C. The amount of repopulation is probed at the B region from the increase in the fluorescence . The rf resonance is monitored

by scanning the applied rf field and observing the fluorescence at B. The rf resonance linewidth is limited only by the transit time of the ions through the rf field region, C.

2.3 The Apparatus:

The ions (Eu^+, Cs^+ etc.) are produced in a Colutron ion source, accelerated to 1.35 keV, focused by an einzel lens and a quadrupole lens, mass-separated by a 90° magnet and are rendered parallel by another quadrupole lens. Laser light from a tunable single mode dye laser (Coherent 599-21) pumped by an Ar^+ laser (Coherent CR-12) enters the chamber and overlaps collinearly with the ion beam in the interaction region (see Fig. 3) which is about 1m long. The rf region (C) consists of a tri-plate line [8] and is located between the two identical fluorescence regions (A and B). Each fluorescence region has an isolated Faraday cage which can be biased to some convenient potential (typically 100 - 400 V) and thereby bring the ions into optical resonance inside the cage. The resulting fluorescence following an optical transition is collected by an aspherical lens doublet and a spherical mirror onto a cooled (-15° C) photomultiplier tube with appropriate interference filters. The signal from the photomultiplier output is processed with standard electronics and sent to a strip chart recorder. The rf signal is obtained from a frequency synthesizer controlled by a PDP-11 computer and sent to the center plate of the tri-plate line after proper amplification. Typical rf power used in the apparatus is ~ 0.05 W. The whole system is pumped by two oil diffusion pumps and the operating pressure in the interaction region is ~ 1×10^{-6} torr.

3. MEASUREMENTS:

We have used this apparatus to measure the hyperfine structure of Eu^+ ions. Europium has two stable isotopes with mass numbers 151 and 153 which are separated by the magnet. The $4f^7(^8S^0)5d\ ^9D_J{}^0$ metastable levels which are about 10000 cm^{-1} above the ground state of the ion (see Fig. 4) are easily populated in the arc discharge type of source (Colutron) used.

For laser-induced fluorescence, only the A region is used. The Faraday cage is biased to -300 V and the laser frequency is scanned to bring the ions into optical resonance which is observed from the fluorescence. The laser-induced fluorescence spectra for $^{151}Eu^+$ and $^{153}Eu^+$ at the optical wavelength 617.305 nm are shown in Fig. 5 and Fig. 6 respectively. The FWHM of the spectral lines are ~ 45 MHz and most of the lines are well resolved. The hyperfine transitions corresponding to the spectral lines have been identified and are shown in the figures.

For the laser-rf double resonance measurements, first the Faraday cage at A is connected to ground and the one at B is given a potential (say -300 V). By scanning the laser frequency, the fluorescence spectrum can be observed at B. Identification of the fluorescence lines and a rough determination of the hyperfine splittings of the levels are prerequisite to rf-resonance measurements. The laser is tuned to resonance peak corresponding to some known hyperfine transition while observing the fluorescence at B. Then the Faraday cage at A is biased to the same potential as at B whereby the fluorescence at B decreases indicating optical pumping at A. This results in the decrease of the population of a particular hyperfine level chosen. The rf power is applied and if the frequency matches the splitting between the depleted level and an adjacent level of the lower hyperfine manifold, there will be some repopulation of the depleted level

according to the magnetic dipole hyperfine selection rule $\Delta F=0,\pm 1$. The recovery of the population at C due to the rf resonance at C region is observed as the increase of the fluorescence at B.

A typical resonance spectrum for $^{151}Eu^{+}$ at an ion energy 1.35 keV is shown in Fig. 7. The occurance of two rf resonance peaks are due to effect of the rf waves propagating along and against the ion beam and are Doppler shifted in opposite directions from the true value. The true resonance position is taken as the average of the two peak positions. Half of the peak separation gives the Doppler shift of the radio frequency. The first order Doppler shift is directly proportional to the frequency and therefore when plotted they bear a linear relationship. A plot of the Doppler shift of the rf resonance versus the resonance frequency is shown in Fig. 8. The slope of the straight line measures β from which the ion velocity and the acceleration potential can be determined. The value of the acceleration potential determined from the linear graph is 1349 V which compares very well with the applied potential (1350 V).

The linewidth of the rf resonance peaks is determined by the transit time of the ions in the rf region. For a uniform velocity and optimal perturbation in a two level system, the FWHM of the transition is given by [9] $\sim 0.8/t$ where t is the transit time. For the case of $^{151}Eu^{+}$ at 1.35 keV the transit time in the 50 cm long rf region is 12 μs and the calculated linewidth is 65 kHz. The linewidth in Fig. 7 obtained by a Lorentzian fit of the data is ~ 59 kHz. The

linewidths observed with this apparatus are in the range 60 - 75 kHz which is an improvement of 3 orders of magnitude over the laser-induced fluorescence spectra.

4. RESULTS:

From the rf resonance measurements the hyperfine splittings of the $4f^7(^8S^0)5d\ ^9D_J{}^0$ metastable levels of $^{151}Eu^+$ and $^{153}Eu^+$ have been measured with very high precision (a few kHz) and accurate values of the magnetic dipole hyperfine constant (A) and the quadrupole constant (B) have been determined. A J-dependent hyperfine anomaly in the $^9D_J{}^0$ has been observed. The analysis also shows a strong admixture of the *s*-electrons which is presumed to be due to core polarization effects. Also accurate values of the nuclear quadrupole moments of these nuclei have been determined. The details of the measurements and analysis are given in ref. [10]. Measurements on Cs^+ ions are in progress.

5. SUMMARY:

We have built a collinear laser and slow ion beam apparatus for laser-rf double resonance measurements. The principal virtue of this apparatus is that in spite of its small size it is capable of measuring the narrowest linewidths ever achieved in similar systems. Also by a different design of the rf line the rf power requirement has been made significantly low. Both, laser-excited fluorescence and laser-rf double resonance measurements can be performed with

this apparatus. Atomic and molecular ions of positive and negative charge states can be studied at high precision. As a first application the measurements and analysis of the hyperfine structure of $^{151,153}Eu^{+}$ have been done.

REFERENCES:

[1] S. L. Kaufman, Opt. Commun. 17, 309 (1976).

[2] W. H. Wing, G. A. Ruff, W. E. Lamb and J. J. Spezeski, Phys. Rev. Lett. 36, 1488 (1976).

[3] S. D. Rosner, R. A. Holt and T. D. Gaily, Phys. Rev. Lett. 35, 785 (1975).

[4] I. I. Rabi, J. R. Zacharias, S. Millman and P. Kusch, Phys. Rev. 53, 318 (1938).

[5] A. Sen, L. S. Goodman, W. J. Childs (1987) Submitted to Rev. of Sci. Instrum.

[6] A. Arnesen, A. Bengtson, R. Hallin, C. Nordling, Ö Staaf and L. Ward, Phys. Scr. 24, 7474 (1981).

[7] A. Sen, L. S. Goodman, W. J. Childs and C. Kurtz, Phys. Rev. A 35, 3145 (1987).

[8] Handbook, Tri-Plate Microwave Components, ed. R. W. Peters (Sanders Assoc. Ind. Nashua, NH, 1956) p. 8.

[9] N. F. Ramsey, Molecular Beams (Oxford University Press, London, 1956), p. 119.

[10] A. Sen and W. J. Childs, Phys. Rev. A (1987) in press.

FIGURE CAPTIONS:

Fig. 1 Laser-induced fluorescence of an ion beam in a collinear geometry. The ions are tuned into optical resonance in the Faraday cage causing excitation from a level F to F'. The level decays by giving out fluorescence which is monitored to record the excitation.

Fig. 2 Laser-rf double resonance in a collinear laser and ion beam geometry. The ions are pumped from lower level F to upper level F' at A whereby the population of level F is partially depleted. The level F is repopulated by the rf transition in the region C and is probed at B by the laser-induced fluorescence.

Fig. 3 Schematic diagram of the two fluorescence regions (A and B) and the rf region (C) located between the two.

Fig. 4 Partial energy level diagram of Eu^+ showing the ground state and a few excited states. Laser excitations are shown by upward arrows and the fluorescent decays by downward arrows.

Fig. 5 Laser-excited hyperfine spectrum of $^{151}Eu^+$ at 1.65 keV ion energy. The hyperfine splittings and the transitions are shown in the upper part. The linewidth is ~45 MHZ.

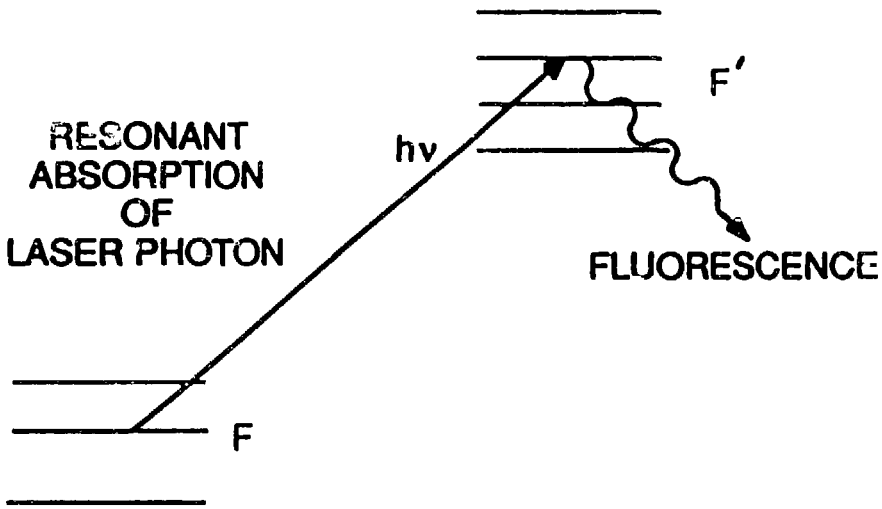
Fig. 6 Laser-excited hyperfine spectrum of $^{153}Eu^+$ at 1.65 keV ion energy. The rf transitions are indicated at the top.

Fig. 7 An rf-resonance spectrum of $^{151}Eu^+$ for the transition F = 7/2↔F=9/2 in the metastable level $^9D_2^0$. The two resonance peaks are due to the propagation of the rf wave parallel and antiparallel to the ion beam. They are symmetrically Doppler shifted in opposite directions from the true resonance.

Fig. 8 The Doppler Shift of the resonance peaks against the true resonance frequency. The slope of the straight line gives β and hence the acceleration potential can be calculated from it.

Fig. 1

LASER EXCITED FLUORESCENCE



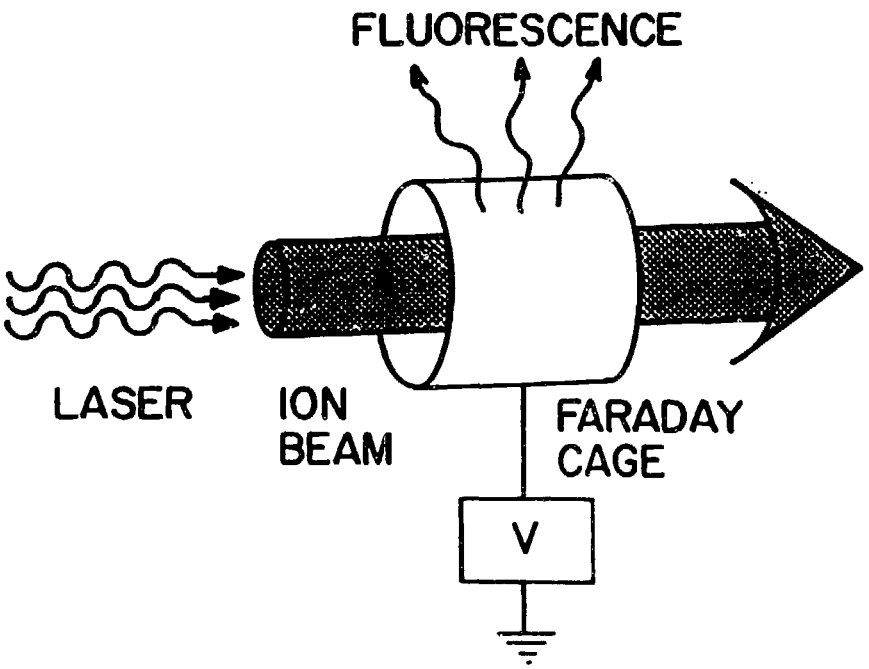
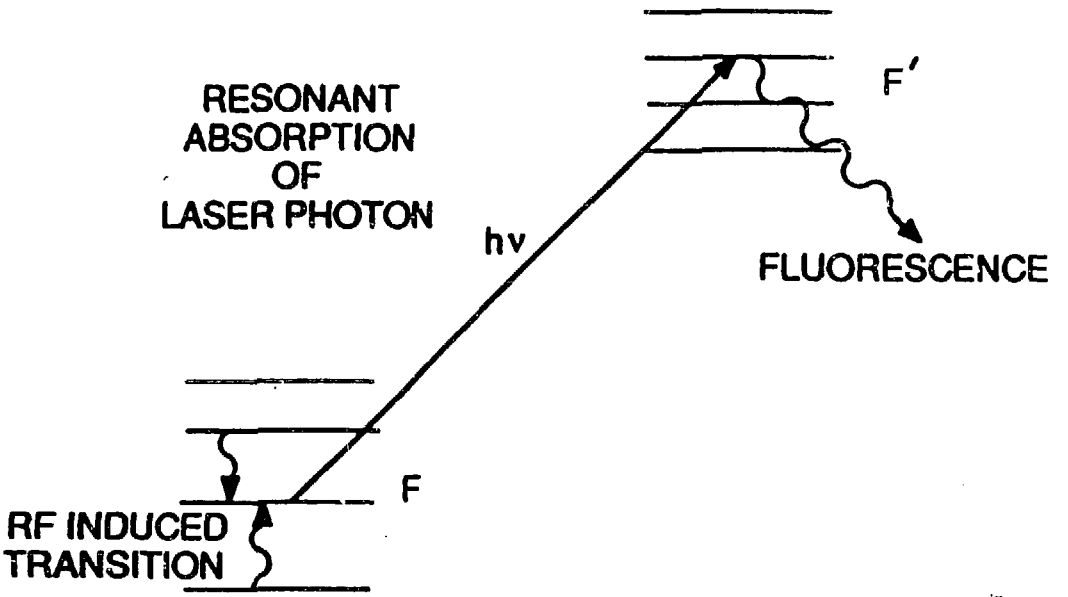
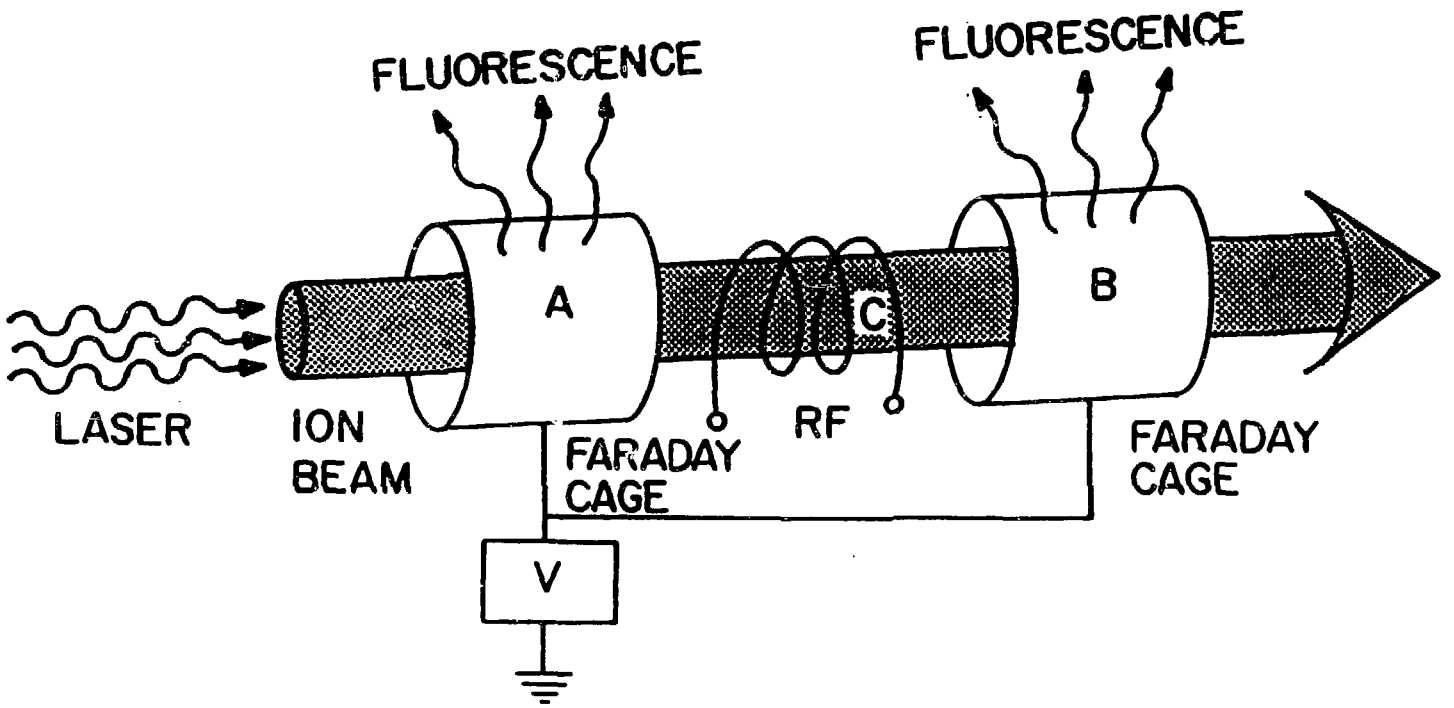


Fig. 2

LASER - RF DOUBLE RESONANCE





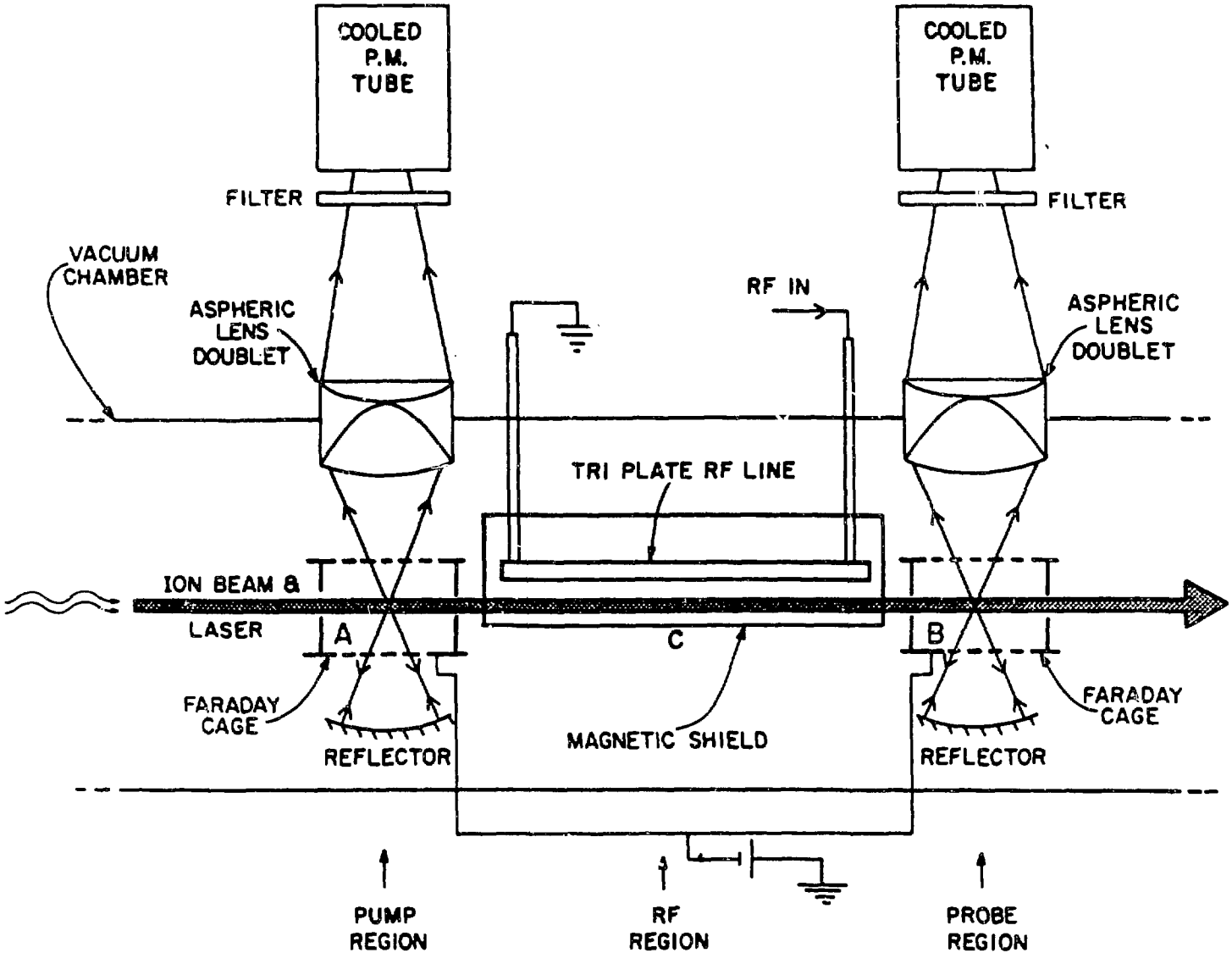

COOLED P.M. TUBE
COOLED P.M. TUBE
FILTER
FILTER
VACUUM CHAMBER
ASPHERIC LENS DOUBLET
ASPHERIC LENS DOUBLET
RF IN
TRI PLATE RF LINE
ION BEAM & LASER
A
B
C
FARADAY CAGE
FARADAY CAGE
REFLECTOR
REFLECTOR
MAGNETIC SHIELD
PUMP REGION
RF REGION
PROBE REGION

Fig. 3

Fig. 4

ANL-P-18,518

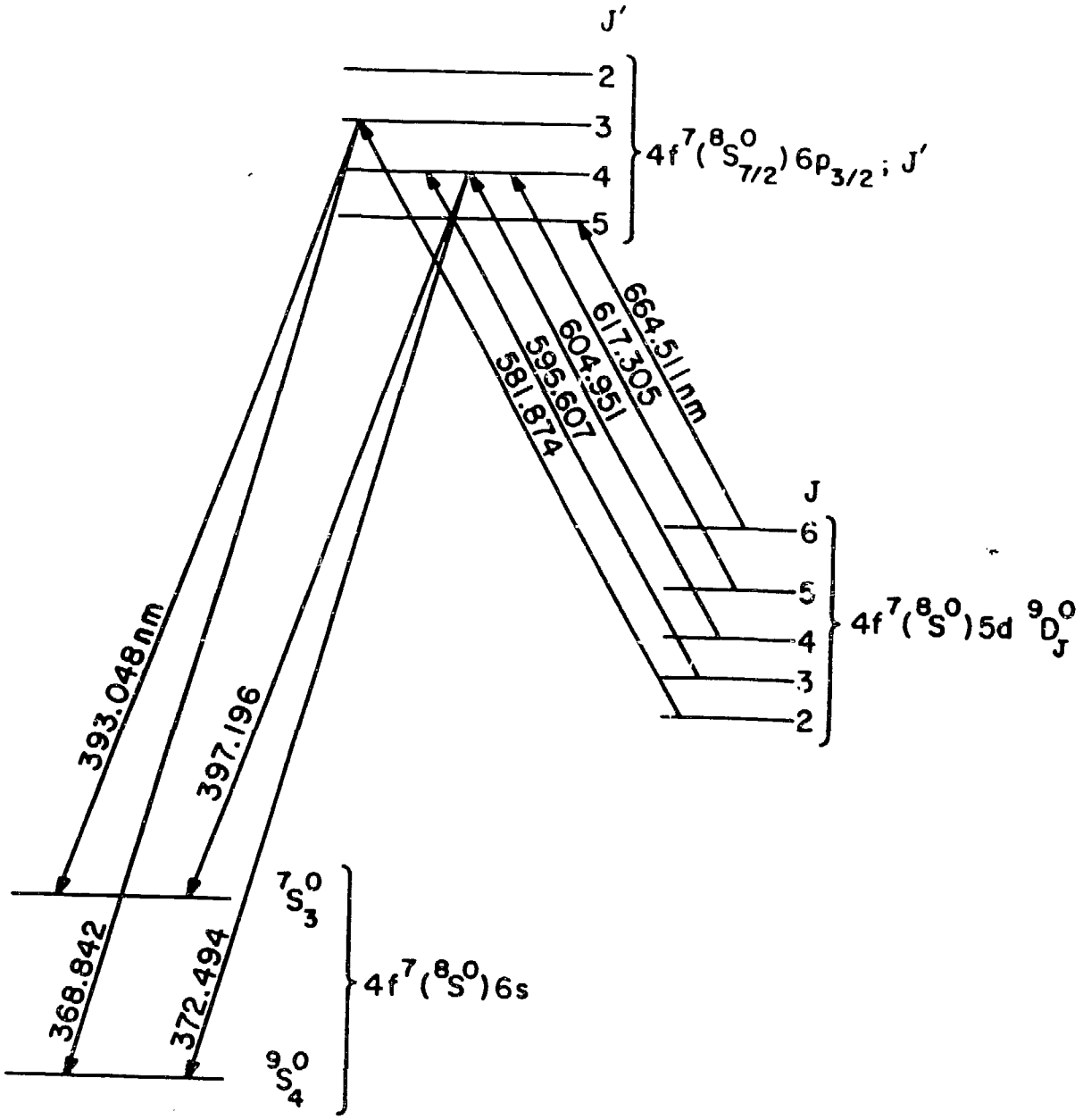


Fig. 5

ANL-P-18,818

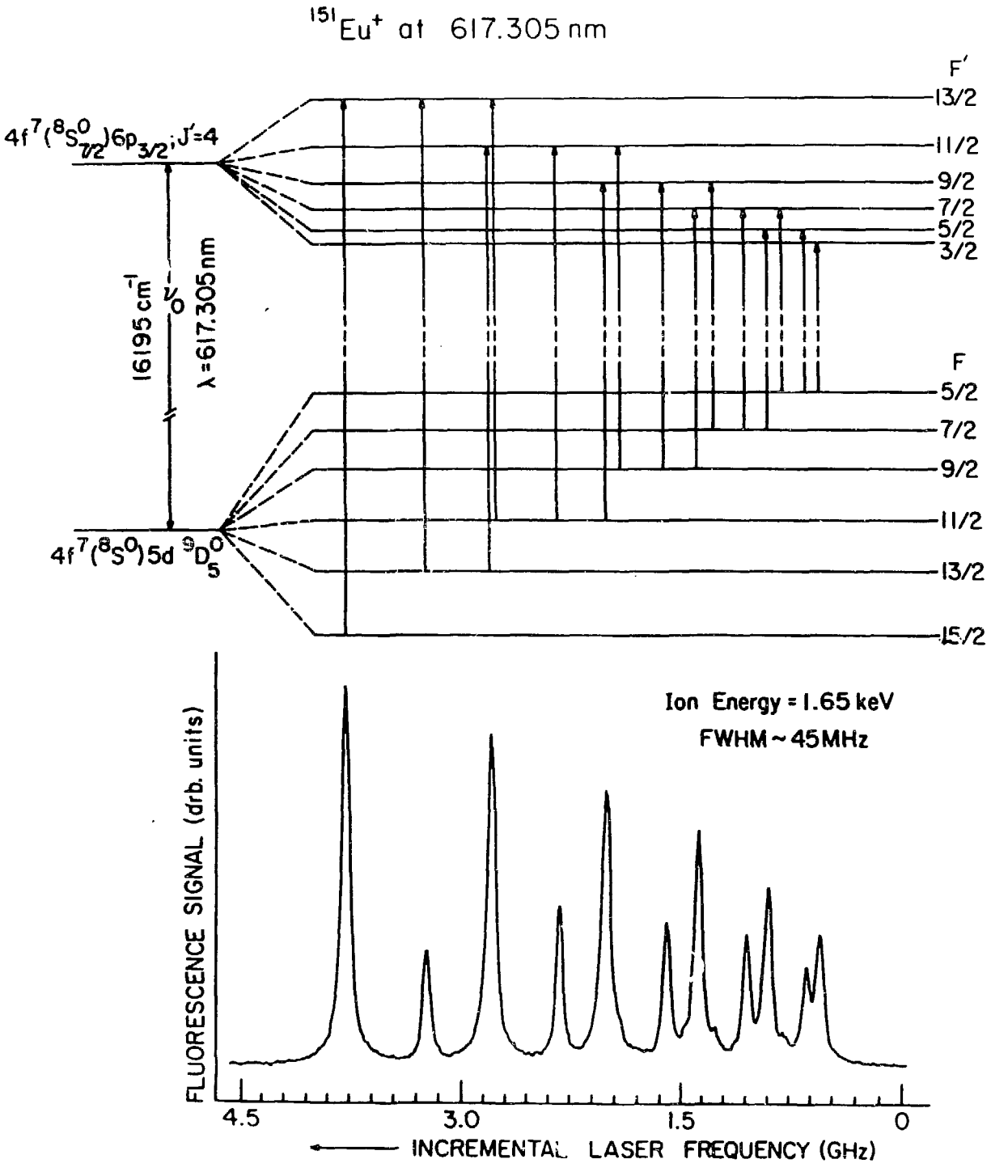


Fig. 6

ANL-P-18,817

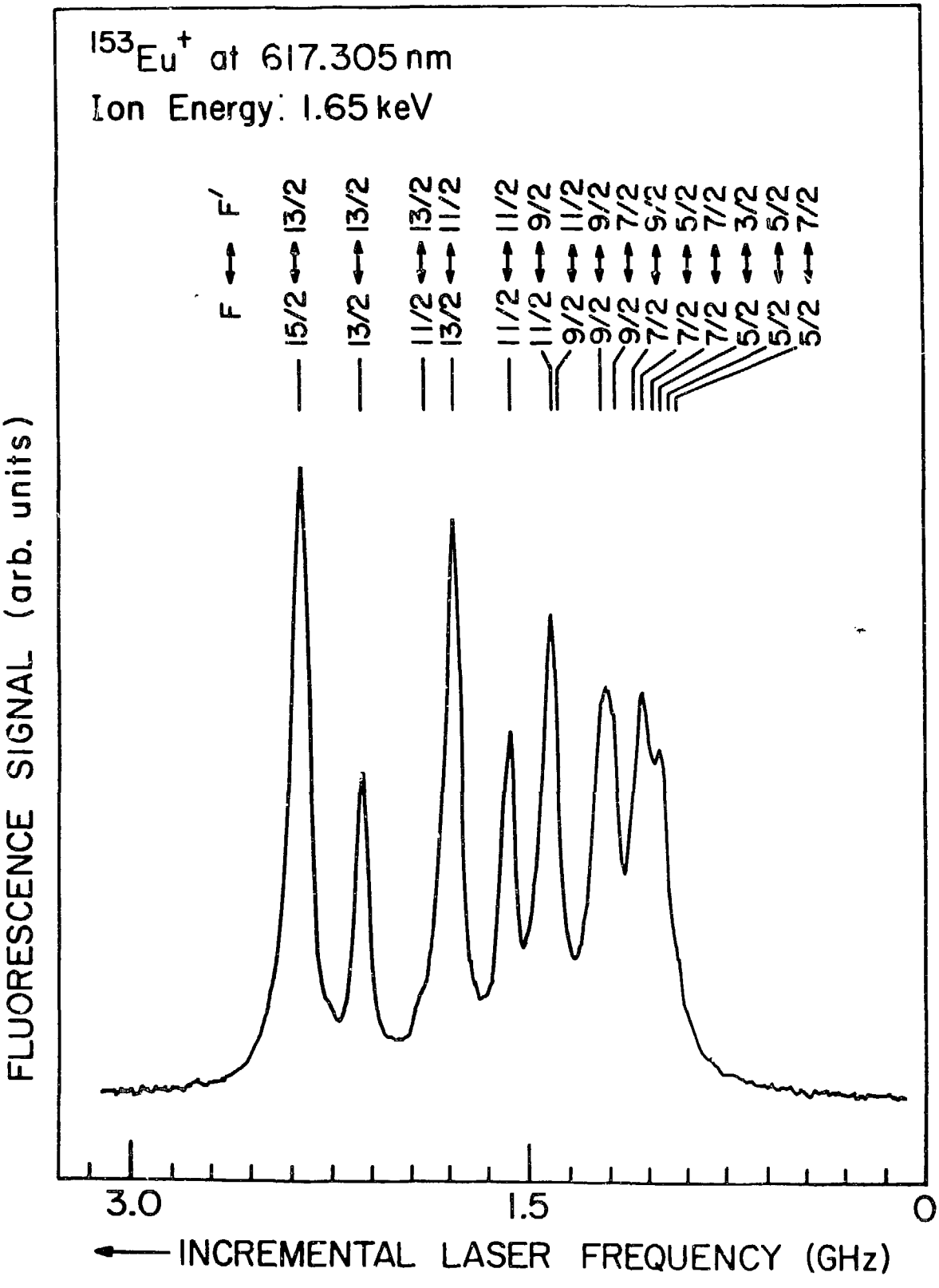


Fig. 7

ANL-P-18,502

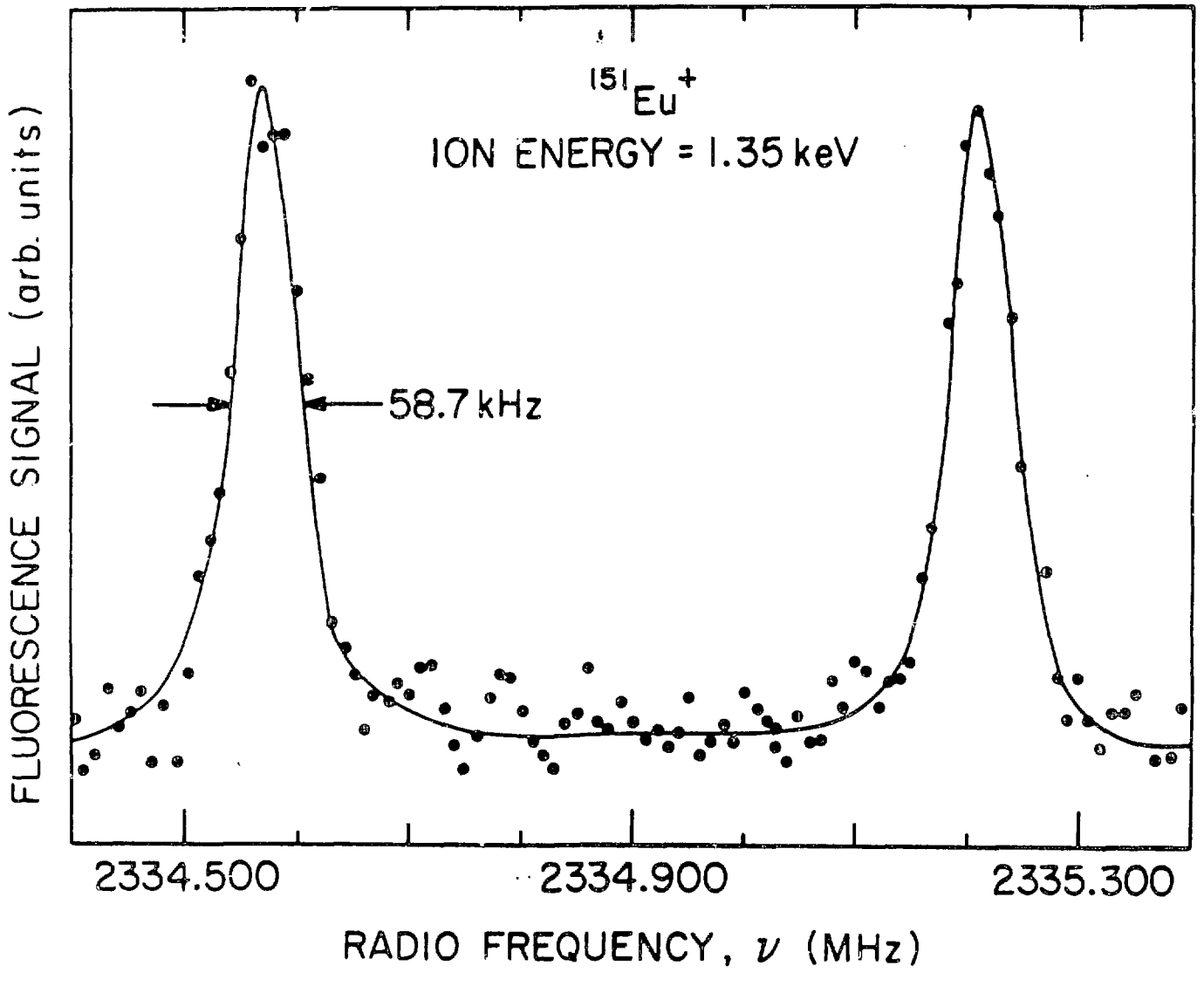
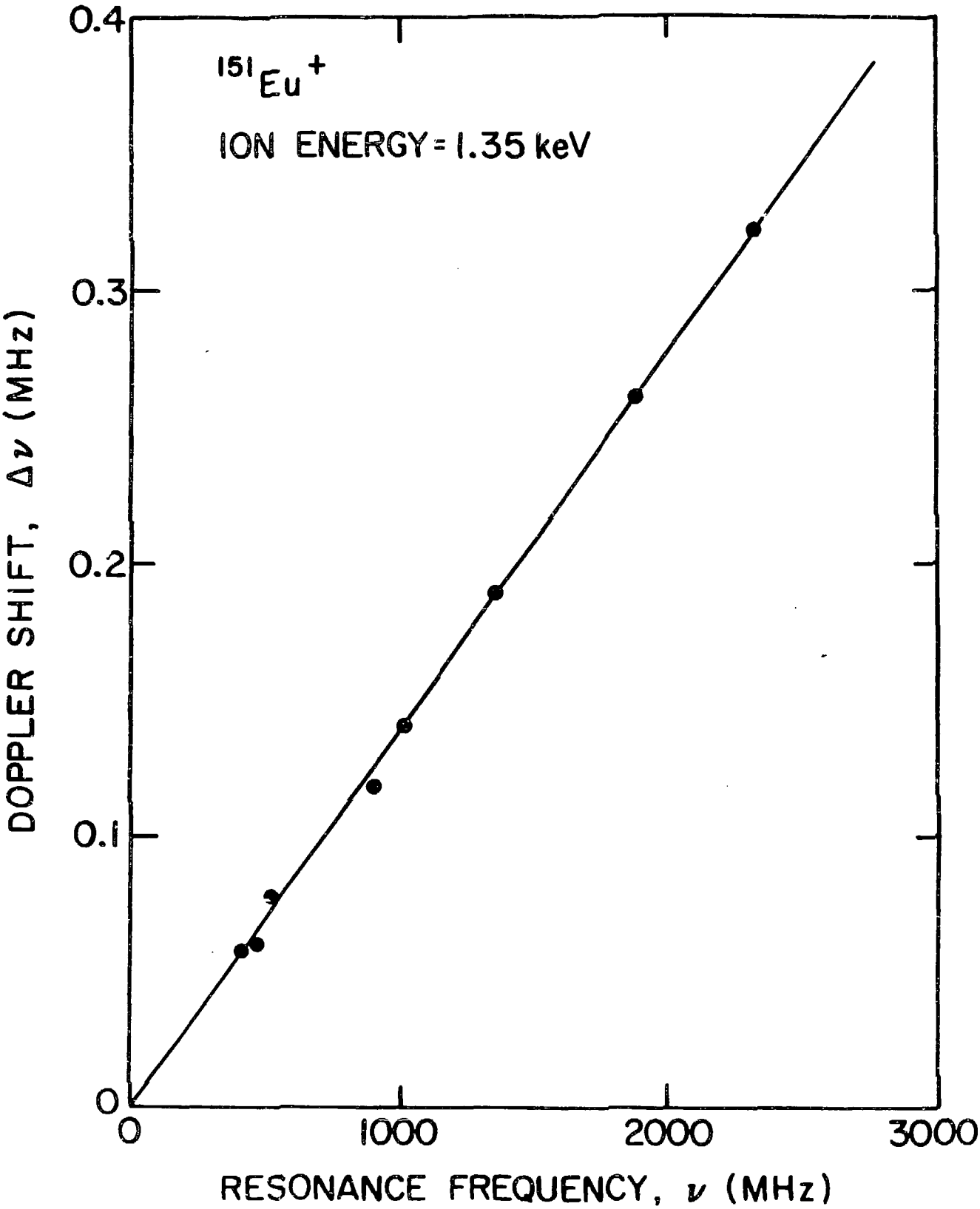


Fig. 8


$^{151}Eu^{+}$
ION ENERGY = 1.35 keV
DOPPLER SHIFT, Δν (MHz)
0.4
0.3
0.2
0.1
0
0
1000
2000
3000
RESONANCE FREQUENCY, ν (MHz)