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LASER SPECTROSCOPY OF U AND UF₆

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Laser Spectroscopy of U and UF₆

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In the laser isotope separation method, uranium atoms or molecules, usually UF₆, are excited by a laser beam with a wavelength tunability and narrow linewidth, before they are fixed by an appropriate procedure. Therefore it is important to know the basic laser photo-excitation process. From this point of view, spectroscopic studies were performed for uranium atom (U) and molecule (UF₆).

Highly resolved fluorescence and absorption spectra of U were obtained mainly for the energy level of 16900 cm⁻¹ by vaporizing uranium metal, producing an atomic beam and irradiating it with a narrow bandwidth laser beam. From these spectra, the isotope shift, the hyperfine structure and the photo-absorption cross section were obtained. And using a pulsed laser beam source, lifetimes of excited states of uranium atoms were measured.

Vibrational and vibronic states of UF₆ were also studied using a solid film of pure UF₆ or a mixture of UF₆ and buffer gas formed on a cold plate. Using a Raman spectroscopy, it was revealed that a peak of a vibrational spectrum is slightly influenced by the concentration of the buffer gas added in UF₆. From the measurement of the absorption and the fluorescence spectrum for the X- \tilde{A} transition, some progressions of the vibrational modes were observed.

Keywords: Laser Isotope Separation, Uranium, Excited State
Life Time, Spectrum, Energy Level, Atomic Beam, Laser,
Hyperfine Structure, Uranium Hexafluoride, Vibronic State
spectroscopy

ウラン原子および六弗化ウランのレーザースペクトロスコピー

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レーザー光によりウランの同位体を分離する方法においては、まず、ウラン原子やウラン分子 UF_6 を波長可変でライン幅の狭いレーザーにて選択励起し、これらを何らかの方法にて固定することが行われる。こうした原子分子の選択励起が本方法において基本的な過程を明らかとするため分光測定を実施した。U原子については、原子励起実験セルを用いてウラン金属を蒸発させ原子ビームを作りこれに単色性の良いレーザー光を照射することによりウラン原子の蛍光および吸収スペクトルを高い分解能で得ると同時にこれらのデータより吸収断面積も計算した。またパルスレーザー光源を使用することによりその蛍光減衰から励起状態の寿命を求めた。次に UF_6 については、低温でのラマン分光を行い $X - \tilde{A}$ 遷移の吸収特性を測定しバイプロニック状態が ν_1 プログレッションで生ずることを確認した。

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1. Introduction

Uranium isotope separation supported by LIS (Laser Isotope Separation) Method is based on the selective excitation of ^{235}U atoms or $^{235}\text{UF}_6$ molecules by an irradiation of a narrow linewidth tunable laser. Thus excited atoms or molecules are finally fixed through an appropriate process. Therefore the optical excitation is the key process in this method. Among others the following parameters are extremely important when uranium atoms are used: a spectroscopic isotope shift between ^{235}U and ^{238}U , a hyperfine structure (HFS) of ^{235}U which is closely connected with the enrichment efficiency of LIS, a lifetime of an excited state, and a photo-absorption cross section which also influences the separation efficiency.

For many years measurements on the isotope shifts and hyperfine structures have been carried out mainly by applying a highly resolved monochromator to the fluorescence emitted from arc or hollow cathode lamps. The atomic spectrum in a hollow cathode lamp, however, has a Doppler broadened spectrum or collisionally broadened spectrum caused by its high discharge energy, so that it is difficult to solve hyperfine structure with high accuracy ^{1), 2)}. Although this kind of broadening can be considerably restricted by cooling the lamp by liquid nitrogen,

the resolution is still insufficient^{3),4),5),6)}.

What is worse in the case of the hollow cathode lamps is that the electric discharge causes many transitions other than those from the ground state and/or the lower metastable states which are essential to LIS. Furthermore due to the weak fluorescence emission from the hollow cathode lamp and due to the adoption of highly resolved monochromators, there were difficulties in measurements.

In these several years, experiments using an atomic beam combined together with a laser spectroscopy have been performed, and so the measurement accuracy is extremely improved⁷⁾⁻⁹⁾. Among others Childs¹⁰⁾ reported ultrahigh resolution measurement based on the double resonance of a laser and a rf. These data, however, do not verify all the allowed hyperfine transitions, because some of them are very weak. Although the lifetimes of excited states of uranium atoms are obtained from an atomic beam^{11),12),14),19)} or a heat pipe method^{15),16)}, measured values do not always agree with each other among several authors. The photo-absorption cross sections are rarely measured by means of the atomic beam¹⁷⁾. All the reports above mentioned are not necessarily aiming at the isotope separation. We made comprehensive measurements of four parameters described above from the viewpoint of LIS method, especially being concerned with

the excited level of 16900 cm^{-1} .

On the other hand, in the case of UF_6 molecule it is known that a highly resolved spectrum can be obtained only when UF_6 is cooled down to a very low temperature, because a hot band, which rises from a thermal excitation of relatively low vibrational levels, can be eliminated, and the densely populated ground state is obtained. A matrix cooling method²⁰⁾⁻²⁶⁾ and a nozzle jet cooling method²⁷⁾ are commonly used for this purpose. The matrix cooling uses a gaseous mixture of UF_6 with the buffer or the matrix gas which has no spectrum interference and does not react with UF_6 . The mixture gas is solidified on the surface of a cold plate.

In this work stresses are put on the observation of peak position changes of the Raman spectrum according to the mixture gas ratio, and the measurement of the absorption cross section based on solid UF_6 film as well as the identification of the vibronic states in the $\bar{A}-\bar{X}$ transition.

2. Experimental

Figure 1 shows a schematic diagram of the experimental setup of Atomic Beam Excitation Apparatus for measuring spectroscopy of uranium atom. It includes a vacuum chamber for producing an atomic beam, a single frequency CW (continuous wave) dye laser pumped with an Argon ion laser for the measurement of the isotope shift and the photo-absorption cross section, a nitrogen laser pumped dye laser for observing the lifetimes of excited atoms, and optical system. Each dye

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laser beam is tuned to a certain selected wavelength with the help of a monitor monochromator and irradiates the uranium atomic beam in the excitation chamber travelling from the bottom to the top. At this moment the resonance fluorescence emitted from excited atoms is detected through a photomultiplier and displayed by an oscilloscope. At the same time the incident laser beam intensity and the transmitted laser beam intensity are also detected. Both optical signals are processed by an electronic divider. Metallic uranium sample is supplied to the hearth and is heated by an electron beam bombardment. Atomic vapor thus produced is collimated through a number of apertures so that the Doppler width may become as narrow as the natural spectrum width. In the photoreaction zone, a well tuned dye laser beam with an extremely narrow bandwidth is introduced. The dye laser has a narrow bandwidth of 15 MHz with small jitter of 5 MHz. On the other hand the pulsed dye laser has a rather broader bandwidth of 1 GHz even with etalon inserted in the cavity.

Natural uranium and enriched uranium of 20% and 89.89 % are used as sample materials. The impurities contained are C 600 ppm, Fe 80 ppm, Si 45 ppm, Ni 30 ppm, N, Al and V each 10 ppm for the natural uranium sample, and C 310 ppm, Fe 90 ppm, Al 45 ppm, Mo and Ni each 20 ppm, Cr and Cu each 15 ppm, Si 10 ppm for the enriched uranium samples. The vacuum chamber is a reaction cell in which uranium atoms interact with the photons emitted by the laser. It consists of a vessel, a vacuum system, an electron gun, a water-cooled copper crucible, collimators, a mass filter, and a film thickness monitor. In the vacuum

system, two diffusion pumps of 1500 liter/s are used in parallel, and a pressure of 1×10^{-7} torr can be maintained during the operation.

Uranium metal placed in the water-cooled copper crucible is melted and evaporated by the bombardment of an electron beam emitting from the electron gun. The uranium vapor passes through the collimators so that the broadening of the spectrum may be reduced. The collimators consist of 4 plates which have 5 mm x 10 mm rectangular openings. As the uranium metal is heated over 2000°C, a relatively large number of thermal ions are produced, and in addition, stray electrons from the electron gun can be observed. Therefore, a positive and a negative voltage are applied to the collimators to eliminate these ions and electrons. Above the photoreaction part, there is a film thickness monitor which measures the uranium atomic beam flux. The windows of the chamber through which the laser beam is introduced are made of BK-7, both surfaces of which are coated with the multi-layer anti-reflection films. The transmission of the windows is over 99 % at 590 nm.

Figure 2 shows a schematic diagram for UF_6 spectroscopy measurement. UF_6 is mixed with buffer gas in the reservoir and ejected onto the surface of a cold plate by adjusting its flow rate by a leak valve. The cold plate is made from copper whose surface is smoothly polished and is equipped with a liquid nitrogen trap to cool down around 80 K. The cold plate is placed in a vacuum chamber where a solid film of the mixture, i.e. matrix, is formed by blowing the mixture gas of 50~500 torr on the cold plate. The mixing ratio ranges between 2 and 100 % UF_6 in Xe buffer gas. Temperature is monitored by a thermocouple implanted in the cold plate. Blowing speed changes from 0.01 to 1 mMol/h. For the purpose of absorption

spectrum measurement, dye laser beam with 6 nsec pulse is used in the wavelength range between 3550 Å and 3850 Å emitting from PBD dye pumped with a nitrogen laser of 400 kW peak power and 10 nsec pulse width. Fluorescence is observed in the wavelength region between 4000 Å and 4400 Å which is obtained when UF_6 is irradiated with 3638 Å and 3514 Å of Ar ion laser emission. Fluorescence spectroscopy is measured by a monochromator combined with a U-360 prefilter. The Raman emission generated when the Ar ion laser of 600mW irradiates the solid matrix film with 5145 Å or 4880 Å, is measured by a monochromator prefiltered by glass filters, i.e. two O-53 for 5145 Å, Y-48 and Y-49 for 4880 Å. To improve S/N ratio, the Raman signal is measured by a lock-in-amplifier synchronized with the Ar ion laser beam modulation signal.

3. Results on uranium atom

3.1 Hyperfine structure and isotope shift

Figure 3 shows the fluorescence spectrum from the enriched uranium atoms observed when the CW dye laser irradiates the uranium atomic beam. In this spectrum the lowest energy peak of the ^{235}U hyperfine structure (HFS) is shifted to the higher energy side by 5.04 GHz from the single ^{238}U peak, which is defined as an isotope effect for LIS. When measured in wave number unit, ^{238}U peak is assigned to 16900.387 cm^{-1} , and the lowest energy peak of ^{235}U spectrum corresponds to 16900.552 cm^{-1} . HFS of ^{235}U originates from

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the nuclear spin $I=7/2$ of ^{235}U , and spreads over the range of 5.54 GHz. This phenomenon can not be observed in the case of ^{238}U with zero nuclear spin. The ground state of uranium atoms is determined as the most stable state which corresponds to the greatest multiplicity of spins or the greatest angular momentum among many sets of electrons composed of (KLMN)⁶⁰ $5s^2 5p^6 5d^{10} 5f^3 6s^2 6p^6 6d^1 7s^2$ configuration.

The ground state is assigned to 5L_6 . The excited level of 16900 cm^{-1} corresponding to the wavelength 5915.38 Å is assigned to 7M_7 , and the transition occurs from $7s^2$ of the ground state to the excited level $7p^1$ according to the selection rule $\Delta J=+1$. Although transition of ^{238}U with nuclear spin $I=0$ solely dominated by ΔJ criteria, the transitions of ^{235}U with non zero nuclear spin $I=7/2$ must be considered on the bases of total angular momentum which is denoted by $F=J+I$ as a result of conjunction between J and I .

$$F=J+I, J+I-1, \dots, |J-I| \quad (1)$$

Therefore 21 transitions are expected in all, which are classified as follows;

$\Delta F=+1$Major transitions with strong intensity

correspond to peaks from A to H

$\Delta F=0$Transitions with moderate intensity

correspond to 7 peaks from a to g

$\Delta F=-1$Transitions with weak intensity correspond

to 6 peaks α to e

Energy levels of hyperfine structure of ^{235}U derived from its nuclear spin are calculated through Casimir's formula.

$$W = \frac{C}{2} + B \frac{3C(C+1) - 4I(I+1)J(J+1)}{8I(2I-1)J(2J-1)} \quad (2)$$

$$C = F(F+1) - J(J+1) - I(I+1)$$

This transition scheme is depicted in fig.4. In order to get hyperfine constants A and B in eq.(2), energy differences

ΔW_1 to ΔW_4 shown in the figure are calculated as follows.

For the excited state ($J=7$);

$$-\Delta W_1(21/2 - 19/2) = 10.5A + 0.3214B$$

$$-\Delta W_2(19/2 - 17/2) = 9.5A + 0.1416B$$

For the ground state ($J=6$);

$$-\Delta W_3(19/2 - 17/2) = 9.5A + 0.3393B$$

$$-\Delta W_4(17/2 - 15/2) = 8.5A + 0.1380B$$

One can get each value from the observed spectroscopic data.

$$\Delta W_1 = 0.960 \text{ GHz}$$

$$\Delta W_2 = 1.260 \text{ GHz}$$

$$\Delta W_3 = 0.820 \text{ GHz}$$

$$\Delta W_4 = 0.060 \text{ GHz}$$

Using these values, the constants A and B are determined, and they have relatively good agreement with those found by

other authors, which are shown in Table 1. The hyperfine constants obtained here can give the precise spectral positions for each HFS peak. Calculated energy positions are tabulated in Table 2, for each F value. Comparing theory with experiment indicates a considerably good agreement.

Figure 5 shows the fluorescence spectrum of 20 % enriched uranium from 22862.5 cm^{-1} to the ground level obtained when the nitrogen laser pumped dye laser beam with a linewidth of 5 GHz is used. As this linewidth is far broader compared with the linewidth of 20 MHz above mentioned, therefore the precise spectrum such as a hyperfine structure can not be observed which requires higher accuracy than in the case of isotope shift measurement. The ratio of ^{235}U fluorescence spectrum peak intensity to that of ^{238}U is read as 1/24, but this value is considerably smaller than that of isotopic ratio, 1/4. This phenomenon might be attributed to the fact that the superposition of a hyperfine structure, a Doppler width and a laser linewidth brings the ^{235}U the more complicated fluorescence spectrum than the ^{238}U atoms. This can be explained as follows.

Spectroscopic structure of uranium atoms is assumed that each peak derived from ^{238}U and ^{235}U absorption is separated by isotope shift and ^{235}U spectrum is splitted into several peaks due to the coupling of J quanta with a nuclear spin. The transition probability W is then described in the following way¹⁸⁾.

$$W = \frac{\sigma I}{h\nu_a} \Lambda \quad (3)$$

where $\Lambda = \int d\omega' \int d\omega g_d(\omega' - \omega_a) g_a(\omega' - \omega) g_l(\omega - \omega_l)$

I ; laser intensity

σ ; excitation cross section

h ; Planck's constant

ω_a ; resonant angular frequency (especially resonant to ^{238}U)

ω_l ; laser frequency

g_d ; Doppler distribution function

g_a ; spectral distribution function of an atom

g_l ; laser power distribution function

Assuming the distribution functions g_l and g_a to be Lorentzian, and g_d to be Gaussian, the following functions are obtained.

$$g_d(\omega) = \frac{1}{\Delta\omega_d} \sqrt{\frac{\ln 2}{\pi}} \exp\left\{-\left(\frac{\omega - \omega_a}{\Delta\omega_d}\right)^2 \ln 2\right\} \quad (4)$$

$$g_l(\omega) = \frac{\Delta\omega_a}{2\pi [(\omega - \omega_l)^2 + (\Delta\omega_l/2)^2]} \quad (5)$$

$$g_a(\omega) = \frac{\Delta\omega_a}{2\pi [(\omega - \omega_a)^2 + (\Delta\omega_a/2)^2]} \quad (\text{for } ^{238}\text{U}) \quad (6)$$

$$g_a(\omega) = \frac{\sum_i^{N_s} \xi_i \Delta\omega_a}{2\pi N_s \{(\omega - \omega_{a,i})^2 + (\Delta\omega_a/2)^2\}} \quad (\text{for } {}^{235}\text{U}) \quad (7)$$

where

$\Delta\omega_a$: FWHM of the natural linewidth of an atom

$\Delta\omega_d$: FWHM of the Doppler linewidth

$\Delta\omega_l$: FWHM of the laser linewidth

N_s : number of the hyperfine sublevels of ${}^{235}\text{U}$

$\omega_{a,i}$: position of i-th hyperfine peak of the ${}^{235}\text{U}$
counting from the position of ${}^{238}\text{U}$
absorption peak.

ξ_i : relative absorption of i-th hyperfine peak

The calculated spectrum agrees with the observed spectrum and are illustrated in fig.5, which shows us that the extraordinary peak ratio is originated from the hyperfine structures. Moreover the broader the linewidth of laser is, the less clear becomes the isotope shift.

3.2 Absorption cross section

Absorption cross sections can be calculated from absorption spectra. The density N of atoms is calculated in relation to the atomic beam velocity v and the measured beam flux ϕ .

$$N = \phi / v \quad (8)$$

Accordingly the absorption cross section σ is obtained by using the optical depth L of the reaction zone and attenuation ratio ρ of the transmitted beam intensity to the incident beam intensity.

$$\sigma = - \frac{\ln(1-\rho)}{NL} \quad (9)$$

It is calculated from the spectrum of ^{238}U shown in fig.6, which results in $1 \times 10^{-13} \text{ cm}^2$. For ^{235}U , the above value of the cross section would be obtained by summing up each cross section of HFS.

3.3 Lifetime

When the uranium atoms are irradiated with the laser beam which has the transition probability of W , the time dependent population of excited state atoms N_2 can be written as follows;

$$\frac{dN_2}{dt} = W(N_1 - N_2) - \frac{N_2}{\tau} \quad (10)$$

where τ is the lifetime of the excited atom.

Equation (10) can be solved and

$$N_2(t) = N_2(T) \exp(t/\tau) \quad (11)$$

is obtained, where

$$N_2(t) = [W N_0 + N_2(0) \exp\{-(2W + 1/\tau)T\}]$$

N_0 ; ground state population at the initial time

T ; laser pulse width

Therefore τ is calculated from decay of the atomic fluorescence after the laser beam is intercepted, and the lifetime of 270 nsec is obtained for the excited level of 16900 cm^{-1} .

Observed lifetimes other than this level are listed in Table 3 with the values measured by other authors. Our values seem to agree with them except for the short wavelength range.

4. Results on uranium hexafluoride

4.1. Absorption spectrum

The solid film thickness on the surface of the cold plate is measured using an optical interference between the solid film surface and the copper plate surface.

Figure 7 shows an absorption spectrum for 35.8 % UF_6 -Xe solid film which is formed by blowing the mixture gas for 30~40 min. Many absorption peaks are assigned to vibronic states originating from 24564 cm^{-1} and 25265 cm^{-1} , which are mainly ν_1 progression. Figure 8 shows an absorption spectroscopy of UF_6 for 100 % UF_6 solid film, from which absorption cross section of ca. $2.5 \times 10^{-20} \text{ cm}^2$ This figure is slightly greater than the one obtained from gaseous UF_6 (20).

4.2. Fluorescence spectrum

Figure 9 shows the fluorescence spectrum generated from the X- \bar{A} transition of UF_6 when UV beam, i.e. 3638 and 3514 Å of Ar ion

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4.3. Raman spectrum

The position of a Raman peak which is measured for ν_1 and ν_2 fundamental peak is obtained as a function of a concentration of UF_6 . Figure 10 shows the dependence of this wavelength position on the UF_6 concentration, from which it is found that ν_1 position shifts to a shorter wavelength region as the concentration of UF_6 decreases. This value is compared with the one obtained by other authors^{22,24}). The similar trend is seen for ν_2 peak.

5. Discussions and conclusions

In the case of uranium atom, spectroscopic investigations were performed by applying a tunable dye laser with narrow linewidth to an uranium atomic beam. From the fluorescence spectra the hyperfine structure of ^{235}U was obtained and also the isotope shift between ^{235}U and ^{238}U was found to be 5.04 GHz for the transition from the excited state of 16900 cm^{-1} to the ground state. And from the fluorescence decay the lifetimes of several excited states were calculated. The absorption spectroscopy measurements indicated that the absorption cross section for the above transition was around $1 \times 10^{-13}\text{ cm}^2$. Furthermore the theoretical considerations on the hyperfine structure explained that the calculated fluorescence spectra of uranium isotopes agrees with observed ones.

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In the absorption spectrum of UF_6 , although the measurement is limited by an optical interference of solid film and the cold plate, obtained absorption peak positions are found to be unchanged. On the other hand, it is observed that the Raman peak positions change with the UF_6 concentration. However, the change of vibronic peak positions of the absorption and fluorescence spectra were not detected.

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In the absorption spectrum of UF_6 , although the measurement is limited by an optical interference of solid film and the cold plate, obtained absorption peak positions are found to be unchanged. On the other hand, it is observed that the Raman peak positions change with the UF_6 concentration. However, the change of vibronic peak positions of the absorption and fluorescence spectra were not detected.

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Table 1 Hyperfine constants of the ground and the excited state(16900 cm⁻¹) for ²³⁵U

GROUND STATE (J=6)		EXCITED STATE (J=7)		REFERENCES
A(MHz)	B(MHz)	A(MHz)	B(MHz)	
-62.4 _{±1}	4155 _{±90}	-172 _{±1.5}	2685 _{±30}	Gerstenkorn 1973
-60.9 _{±0.4}	4159 _{±17}	-171 _{±0.6}	2692 _{±36}	Hackel 1979
-59.4 _{±0.9}	4107 _{±24}	-169 _{±1.5}	2593 _{±51}	Boehm 1978
-60.559 _{±0.003}	4104.15 _{±0.2}	-170 _{±0.3}	2636 _{±6}	Childs 1979
-61.3 _{±0.9}	4136 _{±32}	-171 _{±1.6}	2623 _{±34}	this work

Table 2 Hyperfine structure of ²³⁵U(0-16900 cm⁻¹ transition)

TRANSITION	PEAK INTERVAL	THEORETICAL VALUE (GHz)	EXPERIMENTAL VALUE (GHz)
ΔF = +1	A - B	1.77	1.78
	B - C	1.29	1.29
	C - D	0.93	0.94
	D - E	0.68	0.69
	E - F	0.50	0.51
	F - G	0.39	0.40
	G - H	0.33	0.40
	ΔF = 0	A - a	0.94
B - b		1.24	1.21
D - c		0.49	0.48
F - d		0.35	0.33
H - e		0.30	0.32
H - f		0.71	0.70
H - g		0.93	0.92
ΔF = -1	B - α	0.43	0.45
	D - β	0.46	0.47
	H - γ	0.13	0.12
	H - δ	1.15	1.16
	H - ε	1.72	1.68
	H - θ	1.96	1.95

Table 3 Radiation lifetimes of UI levels

Errors included are estimated $\pm 18\%$ at maximum
 a:Janes¹²⁾, b:Corliss¹¹⁾, c:Klose¹⁴⁾, d:Miron¹⁶⁾,
 e:Carlson¹³⁾, f:Carlson¹⁹⁾

level (cm^{-1})	τ (nsec)	τ from literature (nsec)
27324.5	55	
26920.7	72	23 ^b
26791.6	68	
26583.5	109	
26566.9	89	
26550.4	107	65 ^b
26540.3	74	
26225.6	63	9.8 ^b , 60 ^c
25825.4	53	14 ^b
25791.6	63	
25672.5	94	49 ^b
24066.6	101	57 \pm 6 ^d , 31 ^b
24051.1	45	
24026.2	124	
23993.4	124	
23914.9	140	
23878.1	101	
23848.6	153	
23812.9	77	
23779.2	132	
23702.6	152	
23600.3	289	
23572.1	100	60 \pm 6 ^a , 66 ^b , 80 \pm 8 ^d
23543.5	76	83 ^b
23432.8	157	155 \pm 15 ^a , 135 \pm 14 ^d
23420.6	106	
23057.7	156	
22951.8	117	
22940.3	197	
22918.6	95	75 ^b
22917.5	176	
22862.5	124	135 \pm 13 ^a , 130 \pm 13 ^d
22754.1	80	81 ^b , 70 \pm 7 ^d , 65 \pm 5 ^f
22582.7	130	
22368.5	131	
21584.7	229	130 ^c
20464	188	
19885.5	223	200 ^c

Table 3 (Continued)

level (cm^{-1})	τ (nsec)	τ from literature (nsec)
19647.5	388	
18932.8	152	120 ^c
18759.2	264	
17361.6	116	
16900.4	270	205 ^a , 330 ^c , 255 ^e
16741.6	417	410 ^a , 390 ^c
16505.8	451	400 ^a , 330 \pm 10 ^e
15720.7	188	
15638.3	452	
15631.8	594	

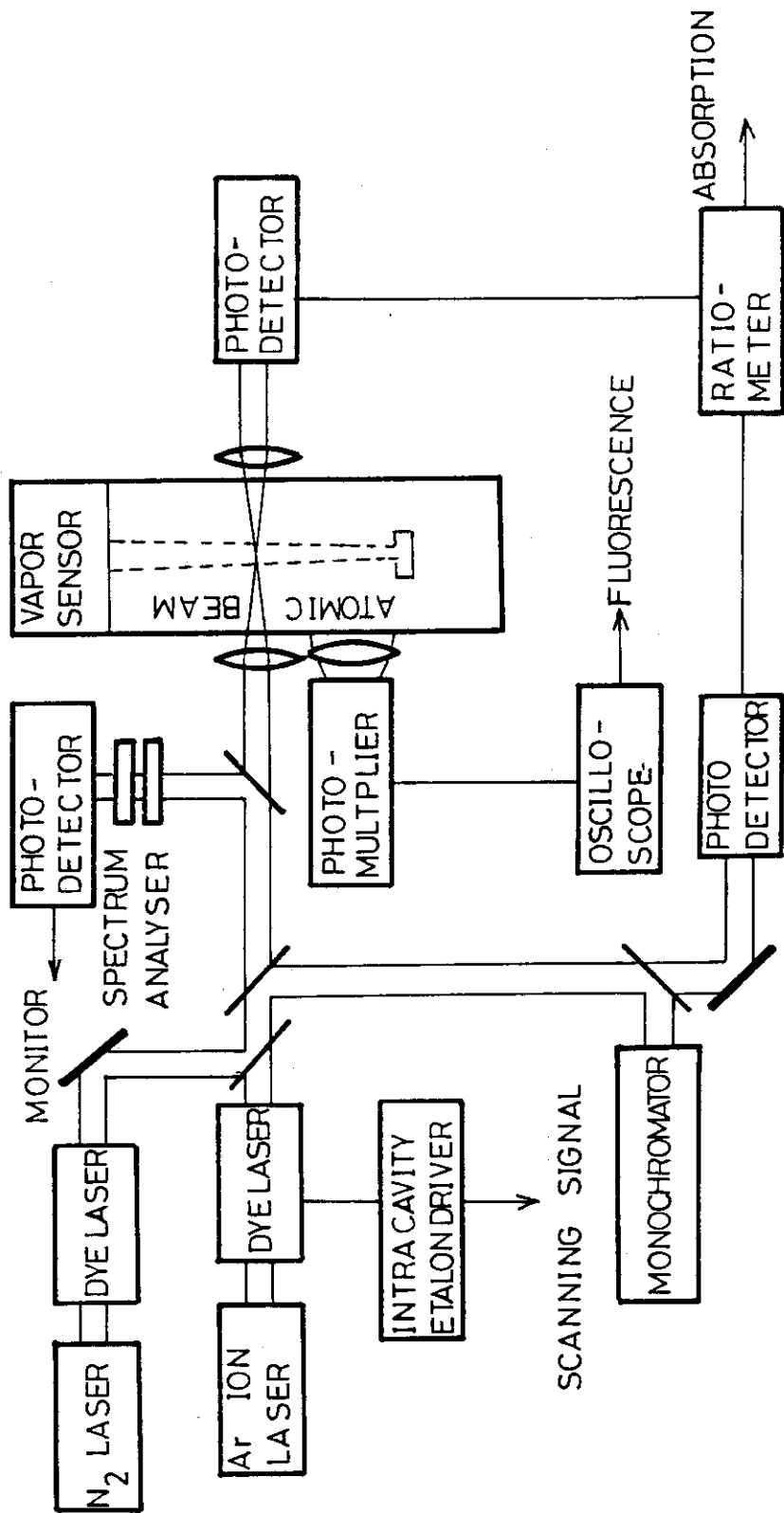
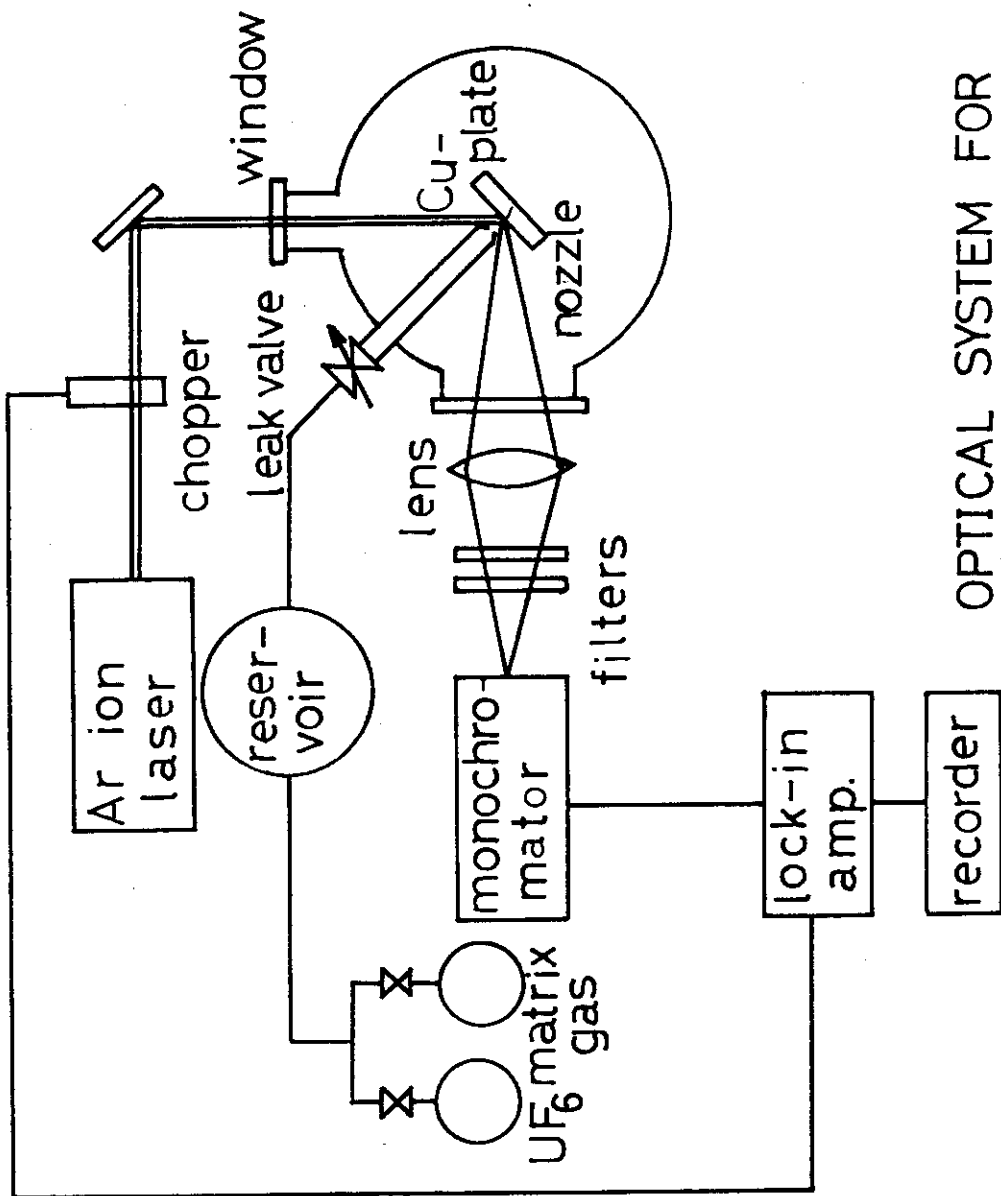


Figure 1 Experimental setup for atomic spectroscopy measurement



OPTICAL SYSTEM FOR RAMAN SPECTROSCOPY

Figure 2 Schematic diagram of UF_6 spectroscopy measurement apparatus

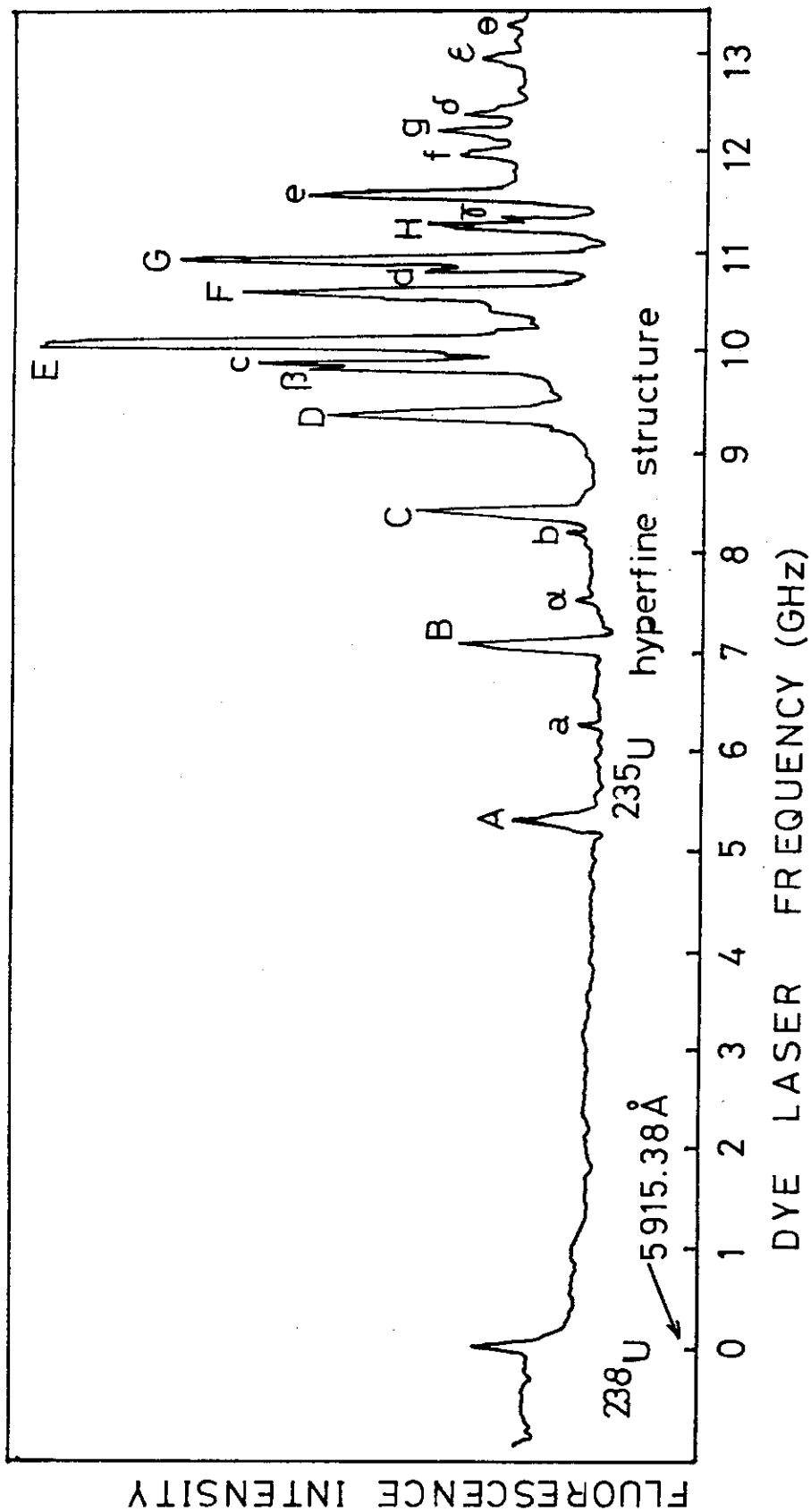


Figure 3 High precision fluorescence spectrum of uranium atom

Sample: 89.89 % enriched uranium

CW dye laser linewidth: 0.02 GHz

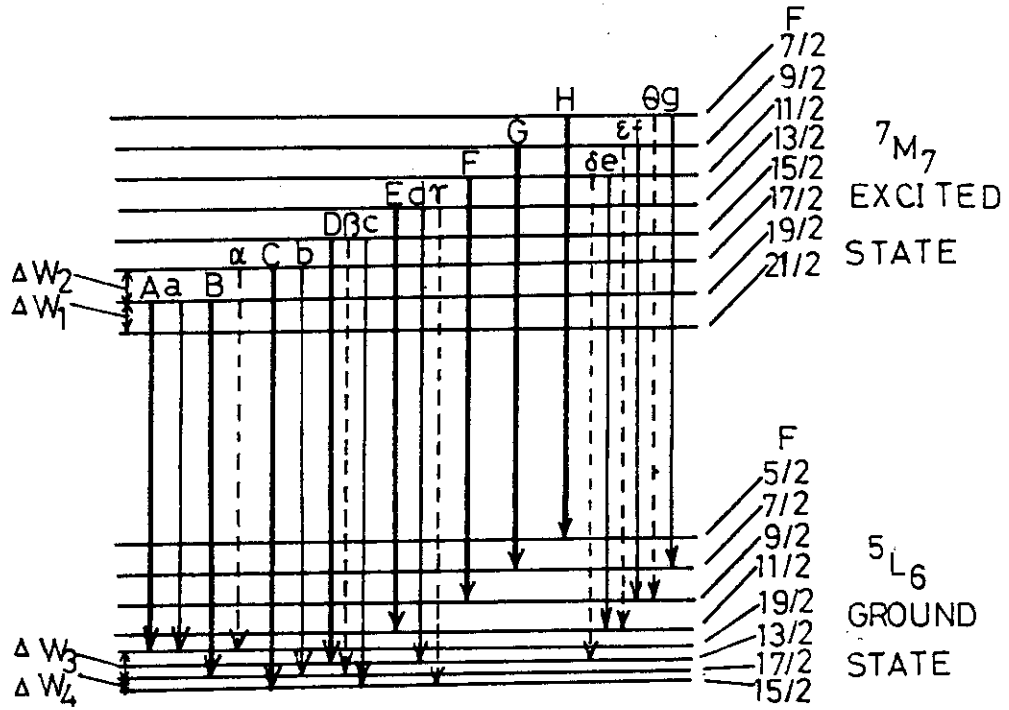


Figure 4 Schematic diagram representing hyperfine transitions of ^{235}U

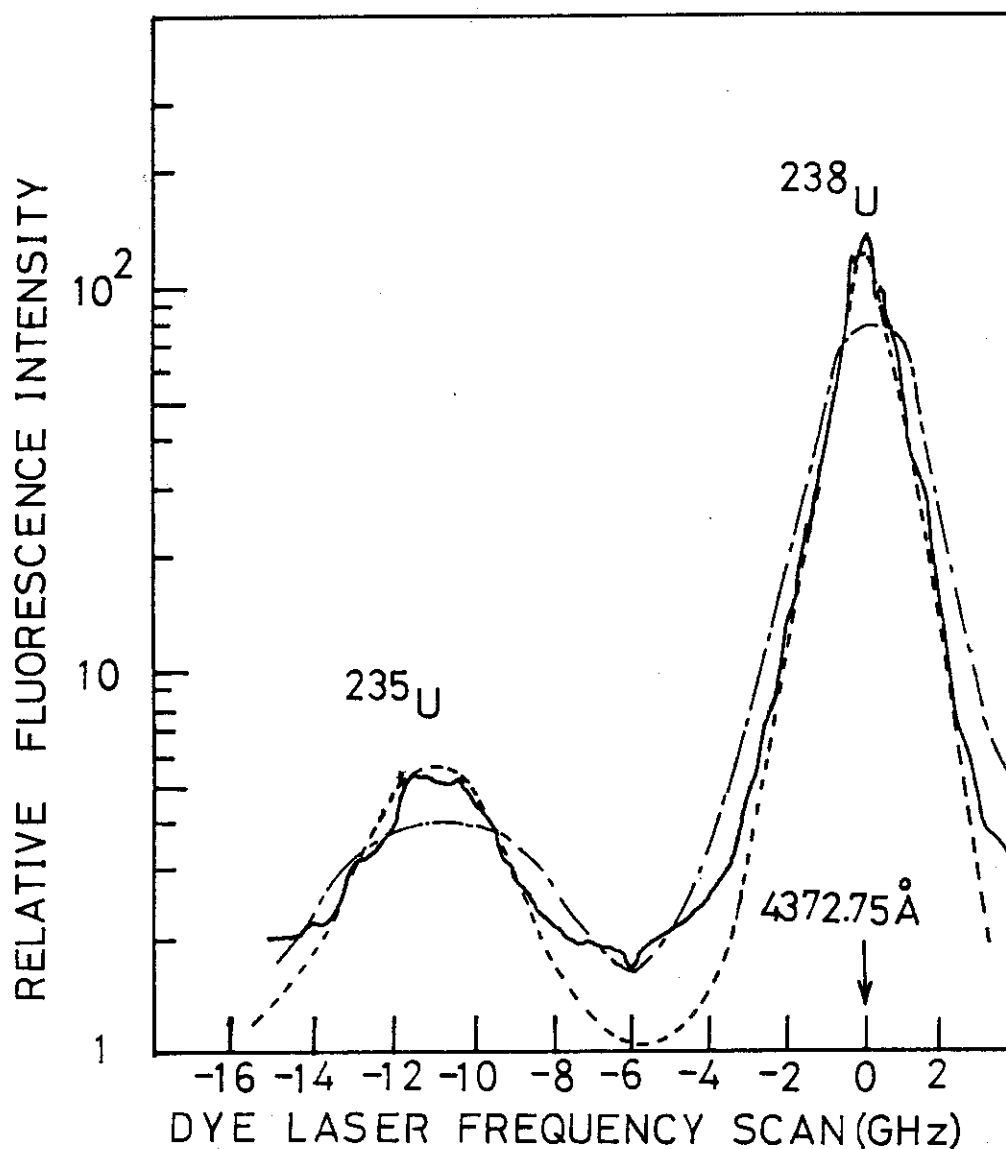


Figure 5 Broadband dye laser spectroscopy

Solid line(experiment):20 % enriched uranium,atomic beam density 2.6×10^{10} atoms/cm³,dye laser power density 76kW/cm²

Dotted line(theoretical calculation):isotope shift 10.5 GHz, Doppler width 0.05 GHz,natural width 0.04 GHz, laser linewidth 4GHz

Dot-dash-line(theoretical calculation):same as above except for the laser linewidth of 8 GHz

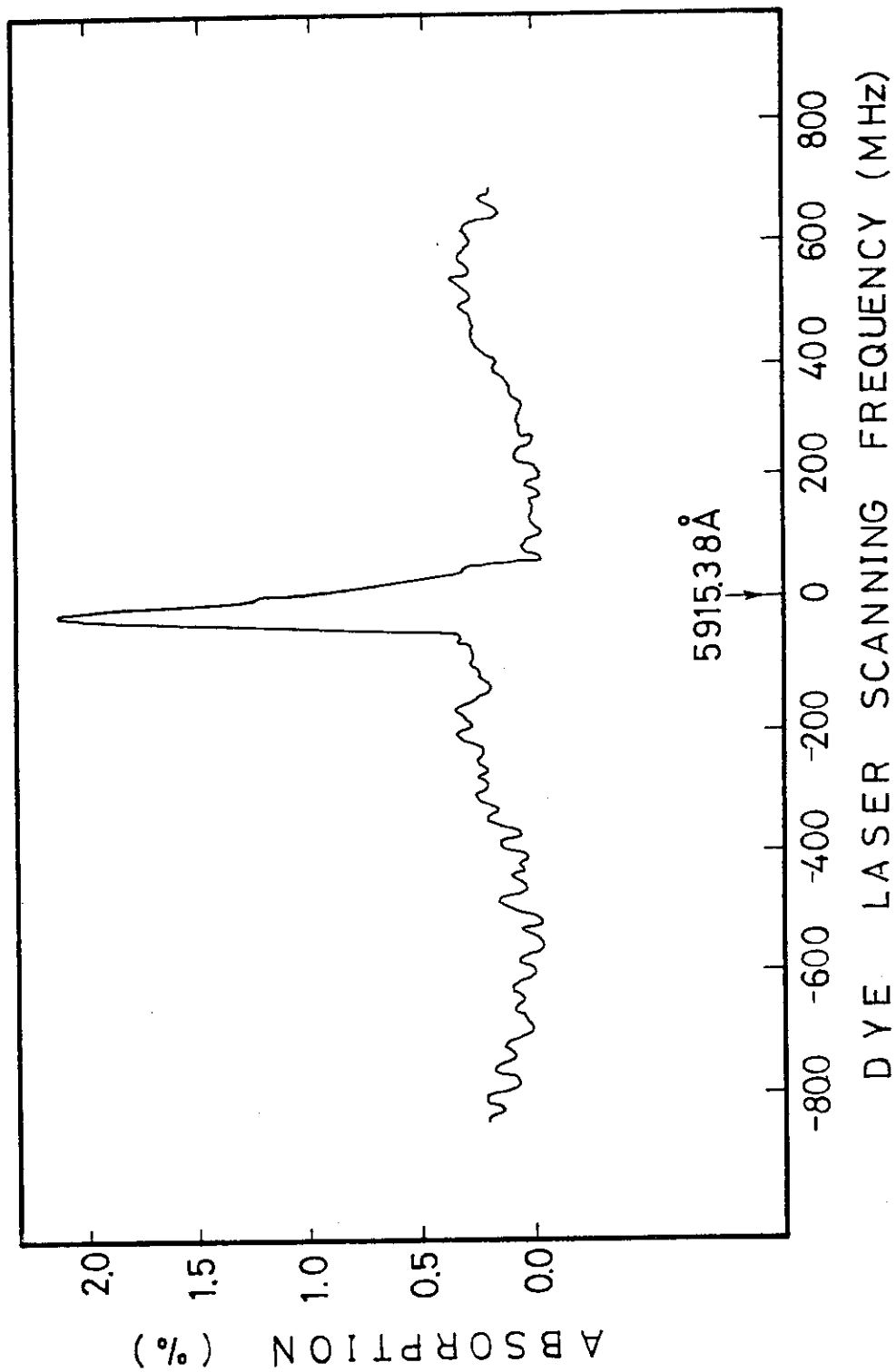


Figure 6 Absorption spectrum of uranium atom(^{238}U)

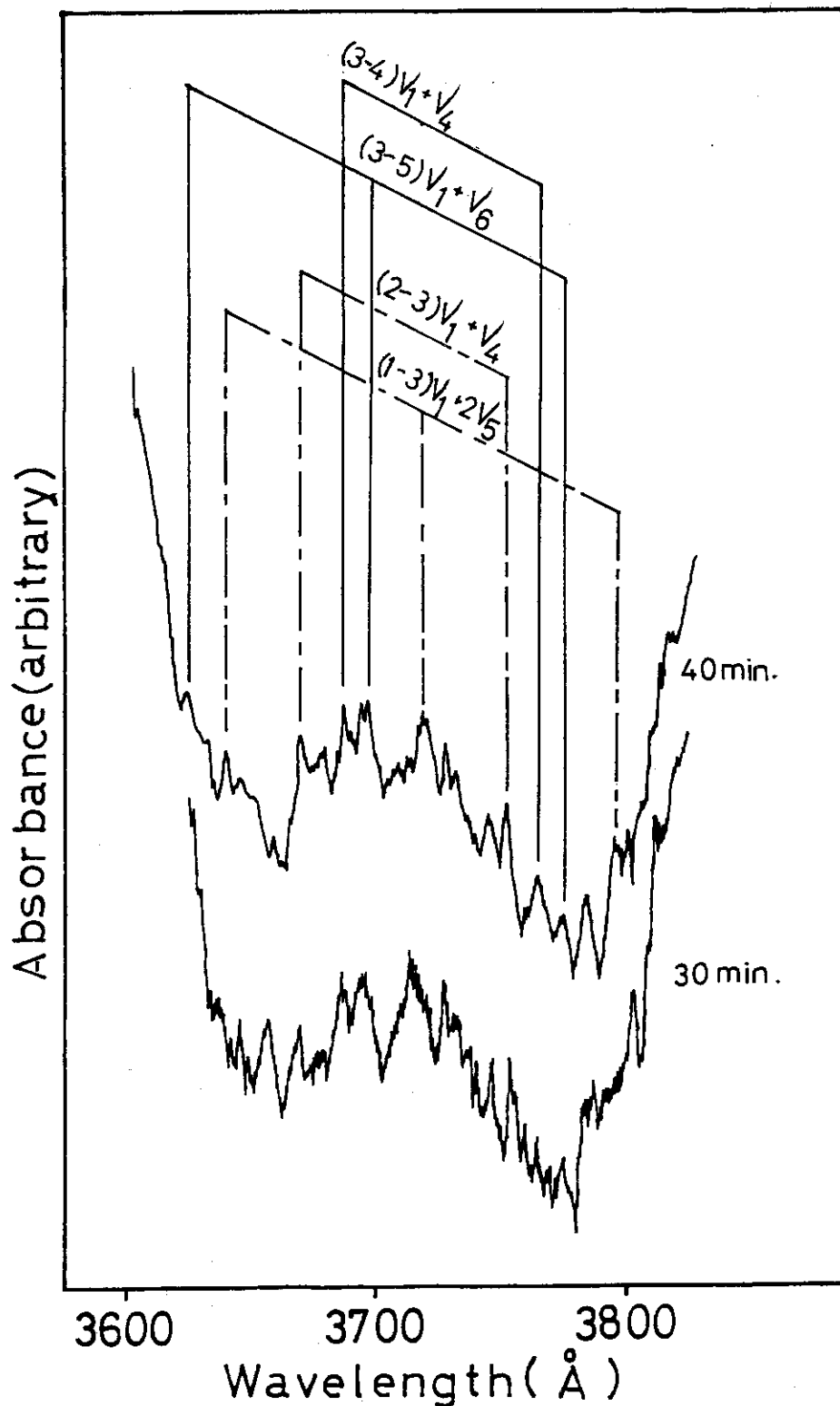


Figure 7 ν_1 progression of UF_6 measured by the absorption spectroscopy

UF_6 concentration: 35.8 %

solid line: origin at 24564 cm^{-1}

dotted line: origin at 25265 cm^{-1}

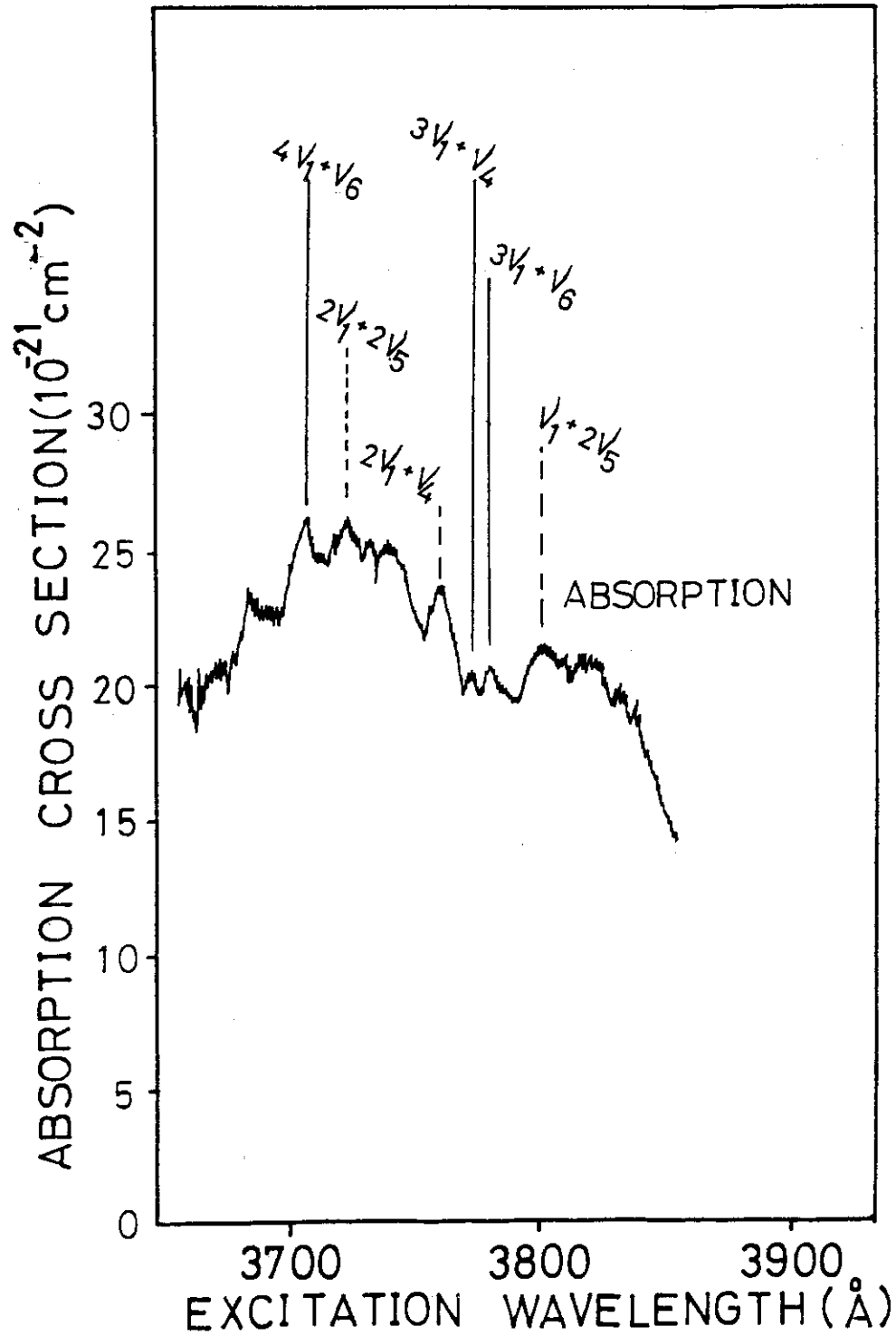


Figure 8 Absorption cross section of UF₆ in the UV range
 solid line:origin at 24564 cm⁻¹
 dotted line:origin at 25265 cm⁻¹

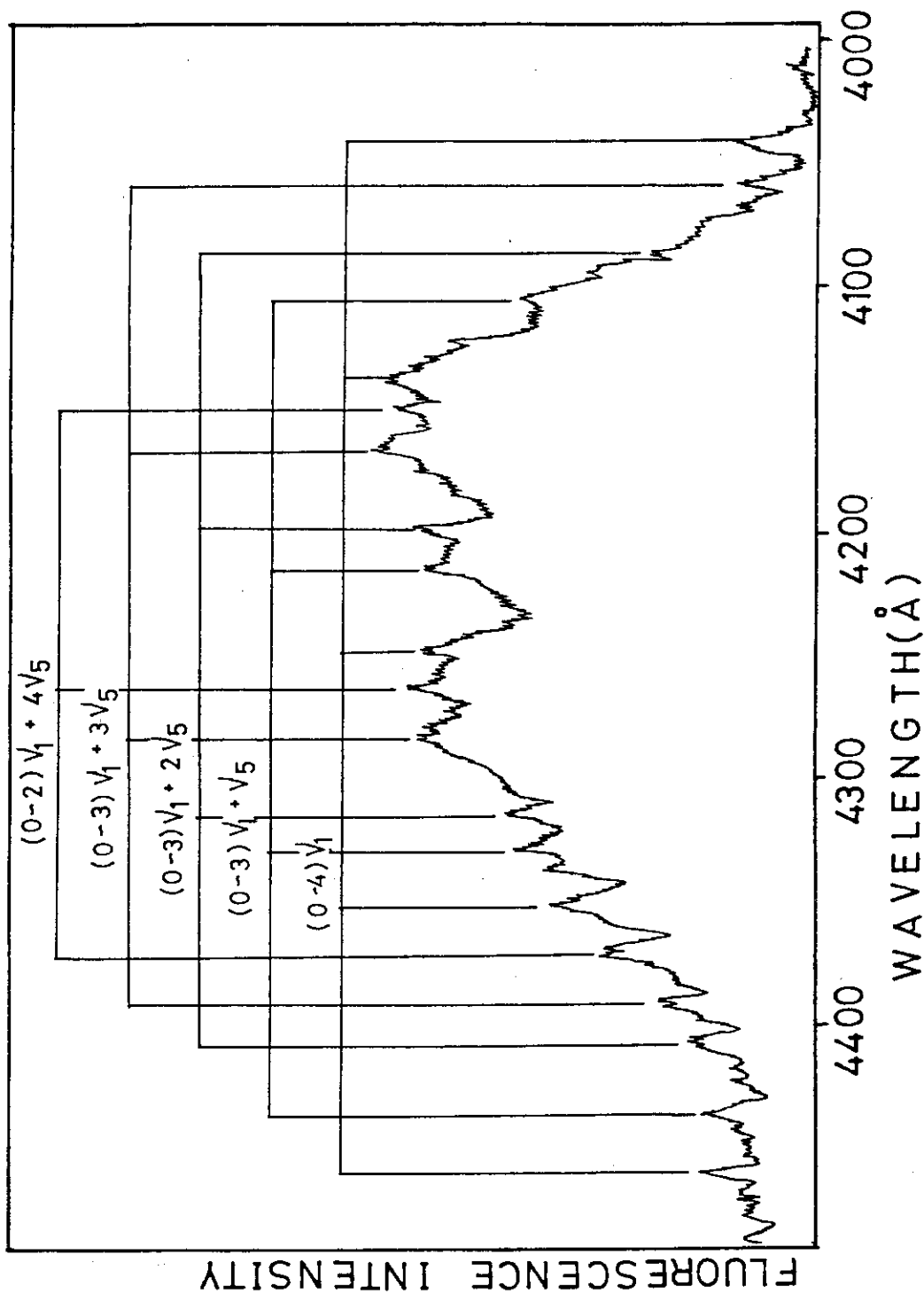


Figure 9 Fluorescence spectrum of UF_6
 excitation: 3638 Å and 3514 Å argon ion laser
 glass filter: U-360

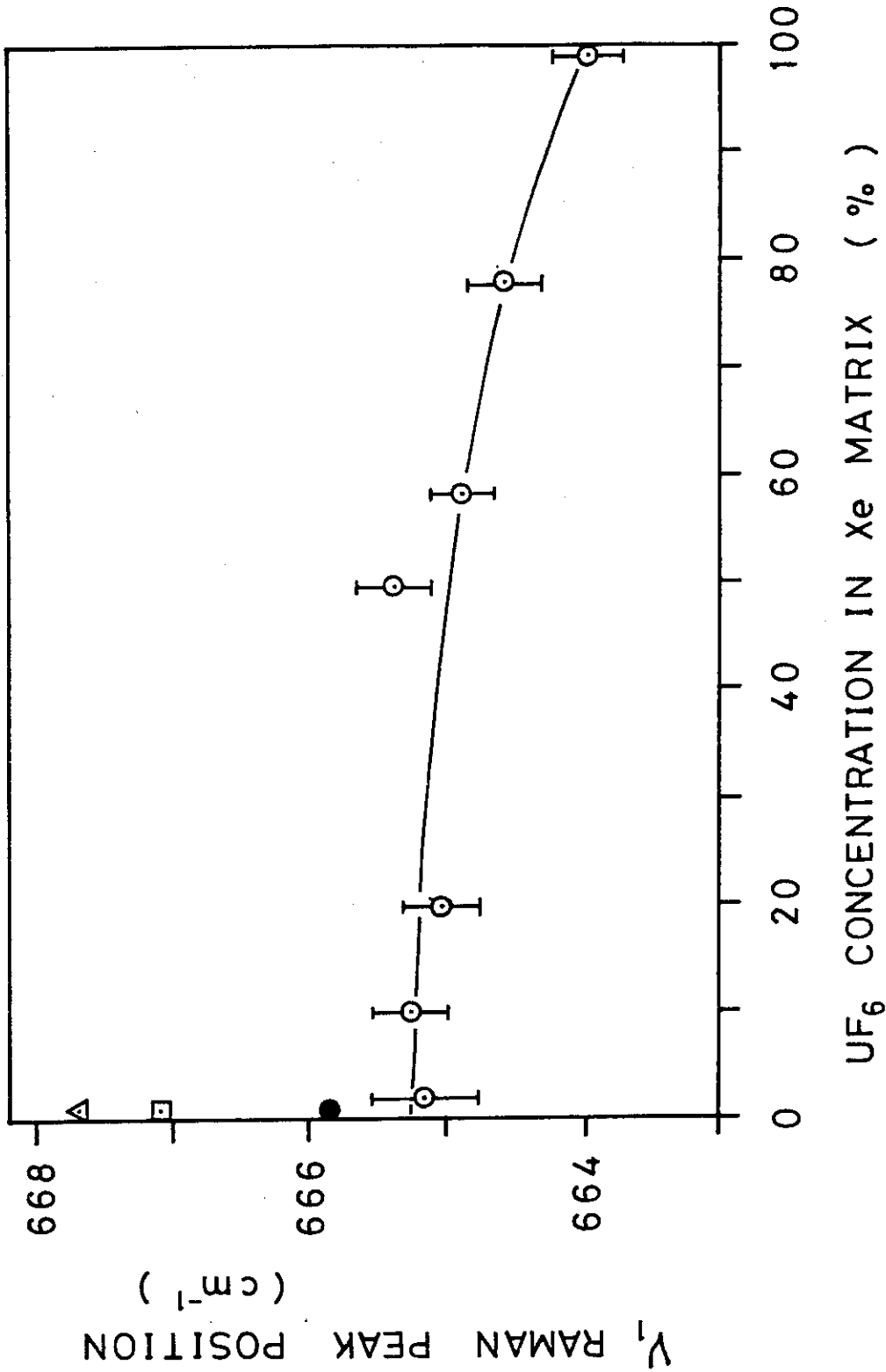


Figure 10 ν_1 position of UF_6 Raman spectrum
 triangle: 0.25 % UF_6 in Ne at 10 K³)
 square: UF_6 gas at 2 atm and 358 K⁵)
 solid circle: 0.25 % UF_6 in Ar at 10 K³)