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Isotope shift and hyperfine structure of the highly excited atomic uranium

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Abstract. The laser induced fluorescence method using atomic beam combined with Doppler-free two-photon absorption technique was applied for the measurement of isotope shifts and hyperfine structures of atomic uranium including ^{234}U, ^{235}U, ^{236}U and ^{238}U isotopes. The isotope shifts between ^{238}U–^{234}U, ^{238}U–^{235}U, ^{238}U–^{236}U, and the hyperfine structure of ^{235}U were obtained in the high lying odd levels around 4 eV.

PACS. 32.10.Fn Fine and hyperfine structure – 32.30.Jc Visible and ultraviolet spectra – 42.62.Fi Laser spectroscopy

1 Introduction

The isotope shifts and hyperfine structures are useful data for basic research in atomic or nuclear physics, and are important data for atomic vapor laser isotope separation (AVLIS) process. Since the atomic structure of uranium is very complicated, almost all configurations of levels are unknown theoretically and experimentally especially for high lying levels. It is expected that by accumulating the parameters such as J-values, g-factors, isotope shifts and hyperfine structures, the atomic structure of uranium may be clarified. Because of large neutron absorption cross-section, ^{236}U isotope contained in recycled nuclear fuel from nuclear fuel cycle processes is undesirable isotope for nuclear reactors. It is difficult to separate ^{236}U and ^{235}U by centrifugation or gaseous diffusion process because of their almost identical weight. It is indispensable to know the spectroscopic data of U isotopes in order to remove ^{236}U isotope in the uranium re-enrichment process using AVLIS. Many studies have provided high resolution spectroscopic data of uranium including ^{234}U, ^{235}U and ^{238}U, such as isotope shifts and hyperfine structures [1–17]. Some workers have reported isotope shift data including ^{236}U isotope [15]. However, sufficient data have not been obtained yet especially for high lying levels.

In the present work, we measured isotope shifts and hyperfine structures of atomic uranium including ^{236}U isotope in the 4 eV odd levels by laser induced fluorescence (LIF) spectroscopy. Near resonant two-photon absorption technique with two laser systems was applied [18–23]. Conventionally for hyperfine structure measurement in the 4 eV, resonant step-wise excitation technique using two laser systems (laser I and laser II) has been applied. The frequency of the laser I has been tuned to the one of the hyperfine components of the 2 eV level and the atoms have been excited resonantly to the 4 eV levels by the laser II [9]. In the same way, measurements must be performed for some components of the 2 eV level. However isotope shift data by this method could not been obtained. This method has been applied only to hyperfine structure measurement of atomic uranium.

The two-photon absorption method is a useful technique to obtain isotope shifts. The advantages of the two-photon absorption technique are that all isotopes and hyperfine structure components in the high lying levels can be measured by only one laser frequency scanning and Doppler-free spectroscopy can be applied. Two-photon absorption spectroscopy has been conventionally applied to metals with low melting point such as Na using a heat pipe in which a high vapor pressure can be obtained. The uranium metal has a high melting point and the vapor pressure is very low. For high melting point metals, the optogalvanic method using hollow cathode discharge may be applied. However, the optogalvanic method has not been applied for two-photon absorption so far. The LIF method is more sensitive than optogalvanic detection. So LIF is suitable for spectroscopic measurement of uranium using atomic beam. In this paper, the isotope shift and hyperfine structure of highly excited atomic uranium by two photon absorption LIF using atomic beam is reported.

2 Experiment

The experimental setup is schematically shown in Figure 1. The atomic uranium beam was produced in an oven

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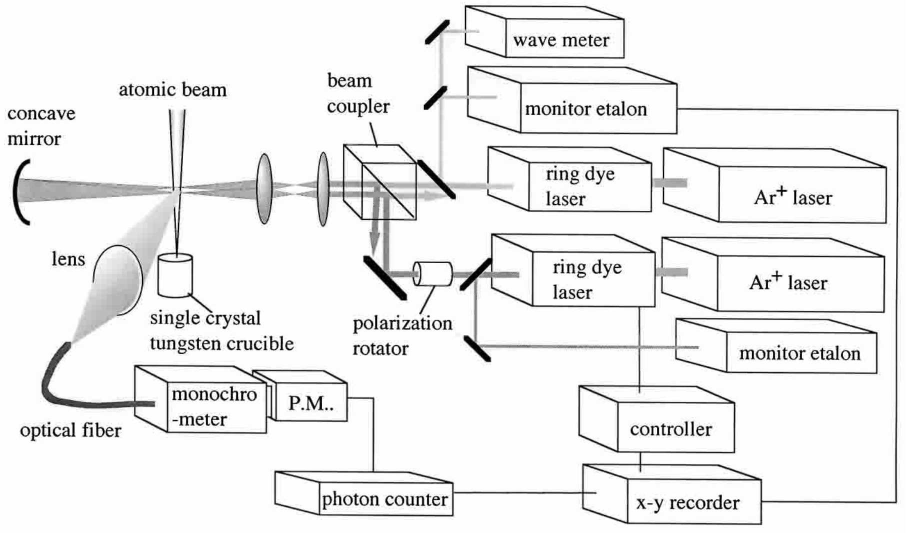

Fig. 1. Experimental setup.

in a vacuum chamber with a threefold crucible [24]. The isotopic abundance of uranium metal piece was 28.2% of ^{238}U, 15.2% of ^{236}U, 55.5% of ^{235}U, and 1.1% of ^{234}U. The sample was resistively heated up to 1 700 °C by a tungsten mesh heater, and the vapor was collimated by a 2 mm diameter hole. At the oven temperature of 1 700 °C, the vapor pressure is 10^{-5} torr. At the interaction region with laser beam, the atomic density is estimated to be 10^7 atoms/cc. Scattered light emitted from the oven is so bright that shields were placed around the interaction region.

Two ring dye lasers pumped by Ar$^+$ lasers were used (Coherent CR699-21 as laser I and Coherent CR699-29 as laser II). The dye used in this experiment was R6G. Two color near resonant Doppler free two-photon absorption technique with two laser beams propagating in opposite directions, was used [16–21]. The laser beams are focused onto the atomic beam by a lens, and reflected by a concave mirror which focused onto the atomic beam again. The two focusing points are very close, but not coincident. In this way, interaction area increases twice in a single pass.

Since the fluorescence intensity decreases rapidly as the detuning increases, the laser I frequency is tuned slightly to off resonance to the 2 eV level, but not overlapping on any spectral shape as shown in Figure 2. Then frequency of the laser II was scanned around the 4 eV level. At the resonance with transition from ground state to 4 eV level, two-photon absorption spectra can be observed. The two laser beams were propagated oppositely to reduce Doppler broadening. The wavelength of laser I was monitored by a temperature controlled confocal etalon with a FSR of 750 MHz, and that of laser II was monitored by 150 MHz as scanning marker. The laser beams cross the uranium atomic beam perpendicularly and then fluorescence is radiated from the excited atoms. Radiated fluorescence was fed into the optical fiber by a lens with an efficiency of 3%. Fluorescence signal through the fiber is fed into the monochromator to suppress the scattered laser light noise and the light from the heater. Fluorescence signal is detected by a photomultiplier cooled by a Peltier element and converted to electric pulse signals, then the pulses are counted by a photon counter.

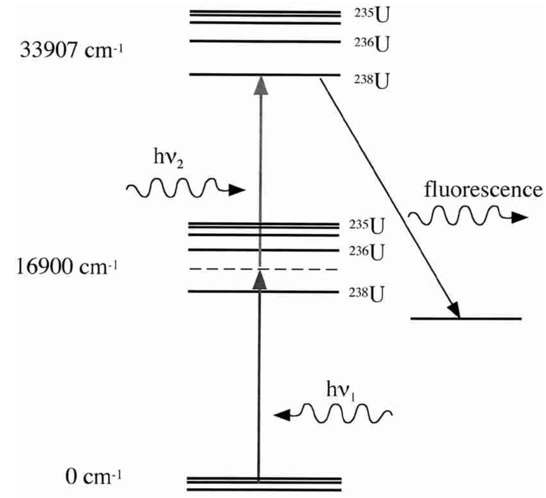

Fig. 2. Two-photon absorption scheme.

3 Analysis

The isotope shift is caused by mass dependent effects due to the finite mass of nucleus and field effects due to the finite size of nucleus. The hyperfine structure is caused by interaction between electric angular momentum and nuclear spin. For the atomic uranium, the isotope of ^{235}U has nuclear spin and the spectrum of ^{235}U has hyperfine structure.

The isotope shift can be described by sum of mass shift and field shift. The isotope shift $\Delta\sigma^i_{MM'}$ for line i between two isotopes with mass number M and M' is described as

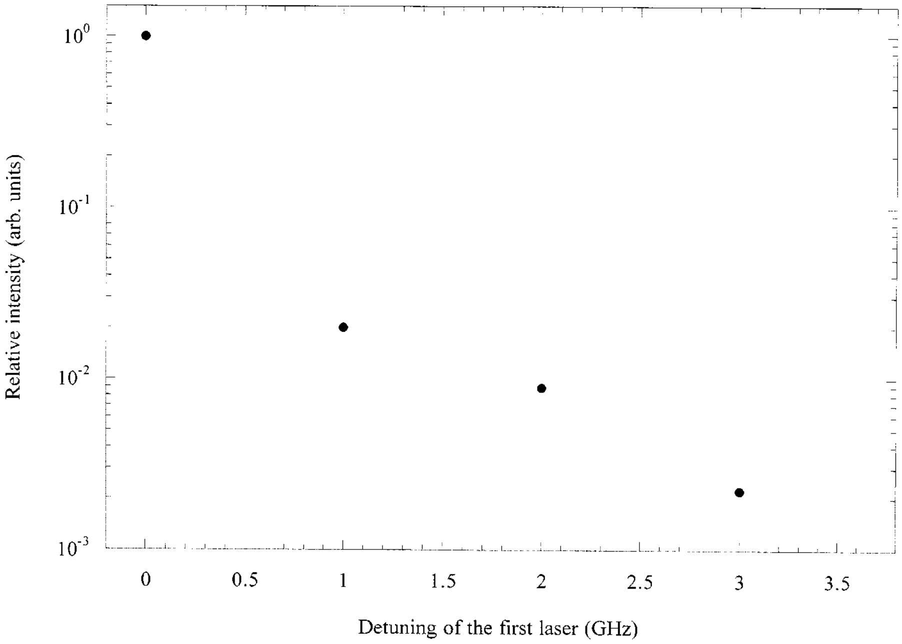

Fig. 3. Relative intensity of two-photon transition spectra as a function of detuning of the first laser.

follows,

$$\Delta\sigma^i_{MM'} = K^i(M-M')/MM' + E^i C_{MM'} \qquad (1)$$

where K^i and E^i are the electronic factor for the mass shift and the electronic factor for the field shift for spectral line i, respectively, and $C_{MM'}$ is the nuclear factor for the field shift so-called isotope shift constant [17]. The first term and second term on the right hand side are the mass shift and field shift respectively. If this modified shifts are plotted against another modified shifts of line j, the plotted point lie on the straight line, so-called King plot [17].

In the assignment and analysis, the detuning frequency of each component is taken into account in the calculation of the transition probability described as follows

$$P = \left| \sum_i \frac{\langle f|E_1\mu_1|i\rangle\langle i|E_2\mu_2|0\rangle}{h(\nu_1-\nu_0)} + \frac{\langle f|E_2\mu_1|i\rangle\langle i|E_1\mu_2|0\rangle}{h(\nu_2-\nu_0)} \right|^2 \qquad (2)$$

where P is the transition probability, f, i and 0 indicate the upper, middle and lower level respectively, μ_1 and μ_2 are electric dipole moments, E_1, E_2 are the electric fields of laser I and laser II with frequencies of ν_1 and ν_2, respectively, and ν_0 is the resonant frequency on the first step. The part indicated as $\langle\ \rangle$ is the electric dipole transition. For each component of the hyperfine structures, it is described using $6j$-symbol as follow,

$$\begin{aligned}\langle u|E_1\mu|l\rangle &= Q(IJ_uF_u,\, IJ_lF_l)\\ &= \frac{(2F_u+1)(2F_l+1)}{(2I+1)} \begin{Bmatrix} J_uF_uI \\ J_iF_iI \end{Bmatrix}^2 \end{aligned} \qquad (3)$$

where F is the total angular momentum, J is the electronic angular momentum and I is the nuclear spin, for ^{235}U isotope, $I = 7/2$. The energy levels of the hyperfine structure components of ^{235}U are calculated by Casimir's formula

$$\begin{aligned} W_F &= A(C/2) + B[3C(C+1)\\ &\qquad - 4IJ(I+1)(J+1)]/[8IJ(2J-1)(2I-1)]\\ C &= F(F+1) - I(I+1) - J(J+1) \end{aligned} \qquad (4)$$

where W_F is the hyperfine energy level, A is the magnetic dipole constant, B is the electric quadrupole constant. The J-values of the high lying levels were measured by two-step excitation Zeeman effect measurement [24].

4 Result

The relative fluorescence intensity when detuning the first step transition frequency is shown in Figure 3 taking notice of the ^{238}U peak. The intensity rapidly decreases as the detuning increases. At the detuning of 3 GHz, the intensity is 1 000 times smaller than on resonant transition. In this measurement, detuning of the laser I frequency was fixed at the center between ^{238}U and ^{236}U peaks of the 0–16 900 cm^{-1} transition whose distance has 5 120 MHz as shown in Figure 4. Laser I must be operated with power density lower than saturation intensity to suppress power broadening effect because of its large transition probability.

Figure 5 shows an example of the two-photon transition spectra from 0 cm^{-1} to 33 907 cm^{-1} *via* 16 900 cm^{-1}. Fluorescence of transition from 33 907 cm^{-1}

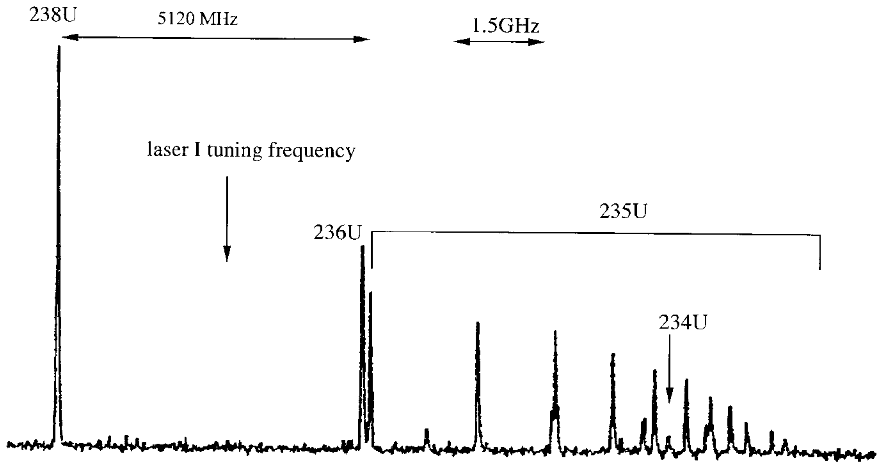

Fig. 4. Tuning frequency of laser I on the first transition of the atomic uranium transition: 0–16 900 cm^{-1}.

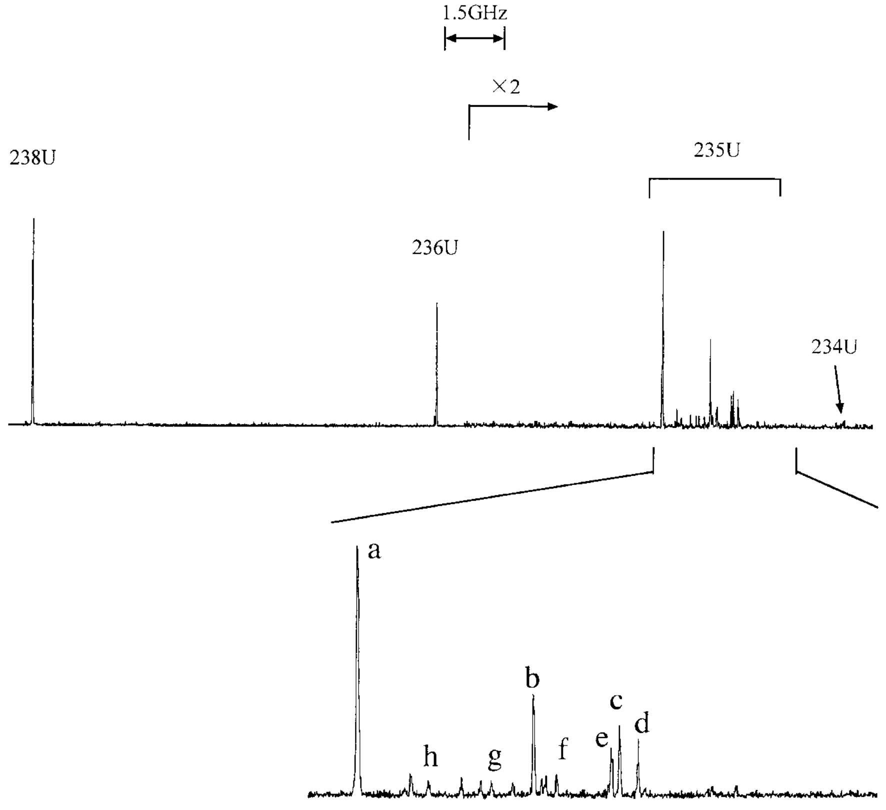

Fig. 5. Isotope shift of two-photon transition spectra of atomic uranium transition: 0–33 907 cm^{-1} *via* 16 900 cm^{-1}.

to 10 051 cm^{-1} that is different from laser wavelength, was observed to suppress the scattered laser light. By Zeeman effect measurement, The J-value of the 33 907 cm^{-1} level was determined as $J = 6$. Wort *et al.* have reported the hyperfine structure of the 33 907 cm^{-1} level. However its hyperfine constants A and B have not been clarified, and the isotope shifts have not been observed. The spectral line width is 20 MHz that is larger than the natural line width which is smaller than 1 MHz. This broadening might be due to the frequency jitter of the laser. Broadening of the frequency jitter is 10 MHz measured by spectrum analyzer. The reason of this frequency jittering is considered to be due to misalignment of the laser or noise.

The first peak from left is ^{238}U and second peak is ^{236}U. Spectral intensity ratio should be 2:1 which is the abundance ratio between ^{238}U and ^{236}U. The very small peak appearing on the right side is due to ^{234}U. The isotope shifts of ^{238}U–^{236}U and ^{238}U–^{234}U are obtained as 10 202 MHz and 20 302 MHz respectively. The spectral group lying between ^{236}U and ^{234}U is the hyperfine structure of the ^{235}U. The hyperfine structure levels are calculated by the least squares method. Hyperfine structure constants A and B of the 33 907 cm^{-1} are obtained as $-79.63(\pm 0.01)$ MHz and $1\,207(\pm 14)$ MHz respectively. The graphical scheme of hyperfine structure is shown in Figure 6. The hyperfine constants of the ground state are obtained as $A = -60.05(\pm 0.03)$ MHz

Table 1. Isotope shift and hyperfine structure constant of the 4 eV odd levels of atomic uranium.

transition (cm^{-1})	J-value (upper level)	Hyperfine structure constant (upper level) this work (MHz)	isotope shift (MHz) 238–236	238–235	238–234
0–33 907	6	$A = -79.63$ (0.01) $B = 1207$ (14)	10 202	17 053	20 302
0–34 434	7	$A = -59.67$ (–) $B = 1764$ (–)	8 481	14 191	—

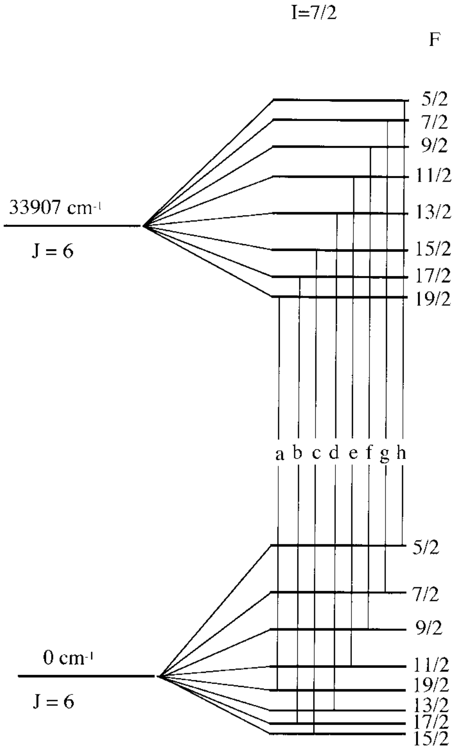

Fig. 6. Hyperfine structure of the ^{235}U at the transition of 0–33 907 cm^{-1}.

and $B = 4\,087(\pm 5)$ MHz which agree with the values obtained by previous worker within 1% [6]. Transitions between hyperfine structure sublevels are shown in Figure 6. The transitions for $\Delta F = 0, \pm 1, \pm 2$ are possible for the two-photon absorption transition. The transitions for $\Delta F = 0$ are stronger than the others. The values in bracket indicate statistical error in least squares calculation. The isotope shift of ^{238}U–^{235}U is obtained by deriving the center of gravity of the hyperfine structure, and is at 12 071 MHz.

The measured isotope shifts and hyperfine structure of the high lying levels are shown in Table 1. In the 34 434 cm^{-1}, J-value of 34 434 cm^{-1} was determined to be $J = 7$. Though the signal was small and only a part of the spectrum could be observed, the minimum number of peaks to analyze hyperfine structure constant could be observed. The peak of ^{234}U could not be observed.

In the isotope shift of the atomic uranium, field shifts are dominant and mass shifts have been investigated as 1~2% by previous work, which could be negligible [15,17]. Then the relative isotope shifts may not take into account the reduced mass that appears in the King plot. Relative isotope shifts between even isotopes with respect to the three transition lines, 0–33 907 cm^{-1}, in which the ^{234}U peak could be observed, could be obtained as,

$$\Delta\sigma(238\text{–}236)/\Delta\sigma(238\text{–}234) \approx 1.99 \qquad (5)$$

which agrees with the value in reference [15]. The isotope shift is one of the most significant parameters in the AVLIS. Even if the small isotope peaks such as ^{234}U were not found in the experiment, isotope shift can be estimated by the relative isotope shifts. The ratio of $\Delta\sigma(238\text{–}236)/\Delta\sigma(238\text{–}234)$ of the 0–33 907 cm^{-1} and that of 0–34 434 cm^{-1} is shown as follows

$$\frac{\Delta\sigma(238\text{–}236)/\Delta\sigma(238\text{–}234)_{34\,434}}{\Delta\sigma(238\text{–}236)/\Delta\sigma(238\text{–}234)_{33\,907}} = 1.001. \qquad (6)$$

This result indicates the King line should pass almost through the origin, and is consistent with the fact that the field shift is dominant in atomic uranium. However, to obtain confidence, more measurements should be performed.

5 Conclusion

The two-color Doppler-free two-photon absorption technique was applied for measurement of the isotope shifts and the hyperfine structures of atomic uranium including ^{236}U by atomic beam laser induced fluorescence spectroscopy with respect to high lying levels. New data of the isotope shifts and the hyperfine structures in high lying odd levels were obtained. In this paper, we showed a small sample of a larger levels in our research program. These data are useful for the AVLIS process. In order to clarify uranium atomic structure, more measurement should be required.

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