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Transition Intensity Relationship Between Different Isotopes of Atoms

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Abstract The total electric-dipole transition intensity originated from the same J- manifold in the small signal high resolution laser spectroscopy between different isotopes of atoms is proportional to their abundance providing that J- mixing from different J states into a hyperfine structure F state is negligible. The intensity distribution of single F-F' transition can be calculated from group theory. Experiments of uranium using intermodulated laser spectroscopy confirmed the theoretical prediction.

Key words transition intensity, isotopes.

Spectroscopic data of transition probability for certain isotope may be difficult to measure comparing to to another isotope of the same atom. For example, data of ^{235}U are important in atomic vapor laser isotope separation (AVLIS). Unfortunately the laser spectroscopic signal is normally very weak in the natural uranium sample because the abundance of this isotope is only 0.7 %. Furthermore, the nuclear spin $I = 7/2$ causes the transition intensity spreads into many hyperfine structures and that the intensity of each component is roughly equal to 10^{-3} of the corresponding transition intensity of ^{238}U which is with 99.3% abundance and no hyperfine structure ($I = 0$). Using ^{235}U enriched sample is one way to solve this difficulty, but such a sample is radioactive and expensive, and sometimes it is impossible for some laboratories.

This difficulty may be also solved by measuring the corresponding transition probability of ^{238}U and using Wigner-Eckart theorem to infer the relevant quantities of ^{235}U if it is mainly an electric-dipole allowed transition. Our experimental results showed that the agreement is very nice using a ^{235}U enriched sample and intermodulated spectroscopic technique.

Hyperfine structure in the atomic spectroscopy originates from the nuclear spin and the nuclear electric quadruple moment. These interactions are considered within a J-manifold in the first order calculation and within many J-manifold in a higher order calculation which is call J-mixing. The effect of J-mixing can be estimated to be

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(hyperfine splitting) / (energy spacing of different J level),

which is less than 10^{-3} in most levels located far beneath the ionization limit. So we shall neglect the J-mixing effect. The wave function of a hyperfine component $|FM_f\rangle$ can be expressed by its parents wave function $|JM_j\rangle |IM_i\rangle$ as

$$|FM_f\rangle = \sum_{M_1 M_j} (JM_j IM_i | FM_f) |JM_j\rangle |IM_i\rangle.$$

The total transition intensity from all the F components originated from J level to all the F′ components originated from J′ level is proportional to

$$\sum_{FF'} \sum_{M_f M_{f'}} |\langle FM_f | D_q | F'M_f'\rangle|^2$$

$$= \sum_{FF'} \sum_{M_i} \sum_{M_j M_{j'}} |(FM_f | JM_j IM_i)(J'M_j' IM_i | F'M_f')\langle JM_j | D_q | J'M_j'\rangle|^2, \quad (1)$$

where D is the electric dipole operator and q denote the polarization: q= 0 for linear polarization and q= ±1 for positive /negative circular polarization. According to Wigner-Eckart theorem[1, 2],

$$\langle JM_j | D_q | J'M_j'\rangle = \langle J \| D \| J'\rangle (J1M_j q | J'M_j')/(2J'+1)^{1/2}.$$

Using the orthogonal relation of Clebsch-Gordan coefficients

$$\sum_{m_1 m_2} (j_1 j_2 m_1 m_2 | jm)(j_1 j_2 m_1 m_2 | j'm') = \delta_{jj'}\delta_{mm'},$$

the eq. (1) can be rewritten to be

$$\sum_{FF'} \sum_{M_f M_{f'}} |\langle FM_f | D_q | F'M_f'\rangle|^2 = (2I+1) \frac{\langle J \| D \| J'\rangle^2}{(2J'+1)} |(J1M_j q | J'M_j')|^2, \quad (2)$$

which is just equal to (2I+ 1) times of the total intensity of transition from J'- manifold to J'-manifold. The measured intensity must account the abandance A_I and the populations on each hyperfine structures. Suppose that the J manifold is the ground state and the hyperfine splitting is much smaller than the thermal energy kT, then each M_F state has the same population. In another word, each M_I state has the population 1/(2I+ 1). The measred intensity for the isotope with nuclear spin I is

$$Intensity(measured) = A_I \frac{\langle J \| D \| J'\rangle^2}{(2J'+1)} |(J_1 M_j q | J'M_j')|^2, \quad (3)$$

where the bracket on the right side of (3) is independent on the nuclear spin I. Then we reach the impotant result: the measured intesity ratio of defferent isotopes of an element is equal to the ratio of their abundances.

The next step is to calculate the individual intensity of each F-F' transition. It is necessary to use the Clebsch-Gordan coefficient of practical values of J and I[3]. It is easy to show that these values are independent of the choice of polarization of light. The physical meaning is that the total transition intensity of F-F' levels for different polarization of the light is always the same due to the fact that there is no preferring direction in the free space.

A Spectra Physics 380D ring dye laser was used in the experiment with about 1 MHz linewidth. A home-made hollow cathode lamp with nickel cathode covered with several milligram ^{235}U enriched layer was used to produce the necessary isotopes. We used both Doppler-free intermodulated fluorescence and Doppler-limited laser induced fluorescence to record the spectra. The whole system was described in more detail in our previous paper[4, 5].

The experimental result of transition from $^5L_6^0$ to $^7M_7$ (0 cm^{-1} to 16900 cm^{-1}) was shown in

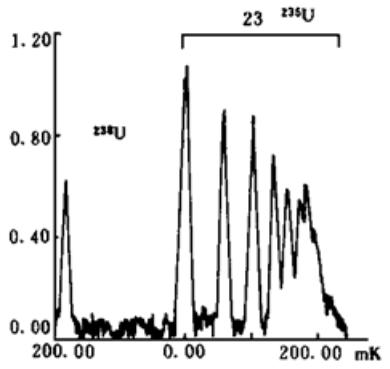

Fig. 1 Transition from 0 to 16900 cm^{-1}

Fig. 1. In accordance with the area ratio of ^{238}U to ^{235}U, it is deduced that the abundance of ^{238}U is 11%. Two similar experiments were performed for transitions from $^5K_5^0$ (620 cm^{-1}) to $^7L_6$ (17361 cm^{-1}) and from $^5L_6^0$ (0 cm^{-1}) to $^7L_6$ (17361 cm^{-1}) and values are obtained 10.4% and 9.4% respectively. For the latter two experiments, the hyperfine structures are hard to be resolved clearly. These values coincide with the actual abundance $(10\pm1)\%$ quite well.

In case of the resolved hfs as shown in Fig. 1, the abundance can also be found by comparision with the intensity of identified hfs component of ^{235}U to that of ^{238}U. The accuracy might be a little bit worse than that from the total area measurement where certain averaging process was performed against the fluctuation of laser system or uranium vapor density.

In order to model the laser isotope separation process, it is necessary to know the Rabi frequency of every M_f-M'_f transition. Such a measurement in isotope ^{235}U is extremely difficult not only from the reasons mentioned above but also to the fact that the interfering from the neighbor hfs component will introduce serious inaccuracy. On the other hand, to perform the measurement in an $I = 0$ system (^{238}U) is much simpler and much more accurate. The transition matrix element of each $M_f - M'_f$ transition can be easily deduced from the Clebsch-Gordan coefficients. For a complete modeling, it is still needed to measure the hfs constants A and B which has to be only performed in isotope ^{235}U. The high resolution spectroscopic technique was shown to be quite successful in such measurements[5~7].

The relationship of transition probability between different isotopes can be used to measure the abundance of isotopes. As shown in the above examples, the accuracy of such a measurement is about 10% provided the signal/noise ratio is fairly high. We used a Doppler-limited laser induced fluorescence technique for $^5L_6^0 - {}^5K_6$ (0 − 16505 cm^{-1}) transition and obtained the abundance of ^{238}U is 7.5% for the same sample. The accuracy is worse due to a smaller signal/noise ratio.

The above conclusion that the total transition probability from $J - J'$ manifold is independent of the value of I and that the total transition probability of any $F - F'$ is independent of the polarization of laser light are based on the homogeneity of free space. It is only correct for the equal occupation of the initial states. If the initial states are populated selectively, then both of the above conclusions will no longer hold. For example, a two step transition from $J = 1$ to $J' = 1$ to $J'' = 0$ for two parallel linear polarization laser lights will be equal to zero because at the second step the initial population of $M'_j = 0$ is zero which is the only possible state to make a transition to $J'' = 0$. If the polarization of the second laser is different from the first one or in a isotope with $I \neq 0$, there does be population on the last J manifold due to such a pumping scheme. This process was used as a method for laser isotope separation of Gadolinium.

The level density is very high for highly lying levels of uranium. It is expected that J- mixing effect would be important. Two experimental methods can be used to verify this J- mixing effect. The first one is to observe the existence of different life time of different F levels originated from the same J manifold[8], and the second one is to observe the transition intensities of different F-F'

deviated from the predicated ones from the Wigner-Eckart theorem. It must be very careful to avoid any nonlinear effects, such as the large signal saturation effect, in this measurement.

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原子各个同位素间跃迁强度的关系

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摘 要 在小信号高分辨激光光谱中, 当不同 J 态在超精细结构态 F 的混合可以忽略时, 起源于同一个 J 能级的原子各个同位素总电偶极跃迁强度正比于它们的丰度。在一个 F-F' 跃迁内的强度分布可以用群论计算出来。用内调制激光光谱方法测量铀的实验证实了理论的结果。

关键词 跃迁强度, 同位素。