Studies in Atomic Structure and Dynamics using Laser Spectroscopy

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Abstract
The thesis deals with investigations relating to the structure and dynamics of atoms using high resolution and laser spectroscopic techniques. The experimental and theoretical work reported here, falls into three broad domains, namely, the high resolution laser spectroscopy for the study of atomic hyperfine interactions, the pulsed laser photoionization spectroscopy to investigate two-photon transitions and highly excited atomic levels including autoionization levels, and finally the multi-step isotope selective photoionization of atoms.

Introduction
High resolution spectroscopy is a prerequisite for investigations on the complex atomic structure of multi-electron atoms and their dynamics. The thesis presents studies in atomic structure and dynamics, using high resolution laser spectroscopic techniques, performed on uranium and erbium isotopic systems, representative of the actinide and lanthanide group of elements respectively. The atomic structure studies were performed on uranium, wherein the region at ~ 2 eV was investigated to understand effect of hyperfine interactions from isotope to isotope. The levels at ~ 4 eV and above including autoionization (AI) levels were studied using photoionization spectroscopy. The photoionization dynamics of erbium isotopes was investigated, both theoretically as well as experimentally.

Atomic Hyperfine Interactions
The hyperfine interactions in uranium isotopic system (233U, 235U, 238U) were studied by investigations on Isotope Shifts (IS) and HyperFine Structures (HFS) of some transitions of uranium, starting from ground or lower meta-stable levels. These investigations were carried out by developing two independent high resolution techniques: - emission spectroscopy with a pressure scanned Fabry Perot spectrometer and scan averaged Opto-Galvanic (OG) spectroscopy, with a single mode tunable diode laser. A hermetically sealed, all metal, liquid air cooled hollow cathode lamp, specially designed and optimized to handle radioactive samples up to few milligrams, served as a spectroscopic source for both the techniques. The emission spectroscopic technique was extended to reduce the sample size to few tens of micrograms, to handle very rare / highly radioactive isotopes. The technique can be
gainfully utilized for off-line spectroscopic measurements of artificially produced isotopes of lanthanides and actinides, having moderate to long half-lives (> 1 day).

The recorded spectra were analysed using a spectral analysis code, developed on the basis of gradient minimization. The work generated new data on IS and HFS of $^{233}$U and $^{235}$U, hitherto unreported in literature. The Relative Isotope Shifts (RIS) and odd-even staggering (OES) of $^{233}$U and $^{235}$U were analysed using the IS values. Our data showed, that $\text{RIS}_{(233,235)} = 1.54 \pm 0.012$ and $\text{RIS}_{(234,235)} = 1.9 \pm 0.012$ and these values are in close agreement with the earlier reported work and the nuclear data of these isotopes. Further, the OES parameters for $^{235}$U and $^{233}$U based on our data are $0.368 \pm 0.012$ and $0.22 \pm 0.012$ respectively. Fig. 1 shows U I spectra recorded for the transition at 682.6 nm.

Photoionization Spectroscopy

For multi-step and multi-photon ionization studies, a versatile laser photoionization set-up was developed. It included both Resonance Ionization Mass Spectrometry (RIMS) and Photoionization Opto-Galvanic Spectroscopy (PIOGS) techniques developed from the first principles. For RIMS, a complete system consisting of a Time Of Flight Mass Spectrometer (TOFMS) of mass resolution of $\sim 300$, photoionization chamber, collimated atomic beam generator, thermal ion suppressor and ion optics was designed and developed.

This set-up was used for studies on U I levels, in the region $\sim 4$ eV and above including Autolization (AI) levels, which are of immense importance in the development of multi-step photoionization (PI) schemes for ultra-trace isotopic analysis and isotope selective photoionization experiments. In this work, two-photon transitions of U I were employed, to excite U atoms in ground or lower meta-stable levels to high lying two-photon resonant levels at $\sim 4$ eV and AI levels, connecting from these levels were studied, using two-colour three-photon $(2\lambda_1 + \lambda_2)$ ionization, by two tunable dye lasers of wavelengths $\lambda_1$ and $\lambda_2$. We investigated two schemes, involving two photon resonances, (a) $0$ cm$^{-1}$ ($J=6$) $\rightarrow$ $33801.05$ cm$^{-1}$ ($J=8$) and (b) $620$ cm$^{-1}$ ($J=5$) $\rightarrow$ $35886.88$ cm$^{-1}$ ($J=6$) and probed even parity AI levels in the energy region $51025-51760$ cm$^{-1}$. These two-colour schemes for probing AI levels, offered experimental simplicity as compared to the usual three-colour schemes used earlier in the literature. While the scheme (a) was reported earlier, we extended in this work the scope of its application to a larger energy region of AI levels. On the other hand, the scheme (b) was explored for the first time. The analysis of the PI resonances recorded using these schemes resulted in several new AI levels$^1$. Moreover, many highly excited odd parity levels in the region $33470-34800$ cm$^{-1}$ were confirmed.

An important criterion in the selection of the two-photon resonant levels in schemes (a) and (b), was their angular momenta $J = 8$ and 6 respectively. Consequently, the AI levels common to both the excitation schemes, were assigned $J = 7$, using angular momentum selection rules.

Isotope Selective Photoionization

Selective excitation and ionization of specific species is an important application of multi-step excitation and ionization spectroscopy. Isotope selective multi-step ionization was
studied in natural erbium, both experimentally as well as theoretically. This system with 5 even (A = 162-170) and one odd isotope ($^{167}$Er) offers an excellent example for investigating various issues, concerning isotope selectivity since typical IS between adjacent even isotopes is ~ 1 GHz while the HFS of $^{167}$Er extends over 3 GHz, thereby causing widespread spectral overlap. For experimental demonstration of isotope selective photoionization of erbium, first a three-step PI scheme was constructed with \( 4f^{12}6s^2 J = 6 \rightarrow 4f^{12}6s6p J = 7 \), as the first step (\( \lambda_1 = 582.7 \text{ nm} \)). The second and third step transitions were arrived at, by performing two- and three- colour three-photon PI spectroscopy to yield \( \lambda_2 = 559.8 \text{ nm} \) and \( \lambda_3 = 621.6 \text{ nm} \). Isotope selective photoionization of natural erbium was then demonstrated using this level sequence and invoking isotope selectivity only in the first step by use of a single mode CW laser of line width of ~ 1 MHz; \( \lambda_1 \) and \( \lambda_2 \) were provided by two Nd:YAG pumped dye lasers of line width 3-5 GHz. The experiments were conducted in the RIMS chamber, where natural erbium was evaporated and collimated, to yield residual Doppler width of ~ 60 MHz. Complete isotope selective photoionization of erbium was achieved in these experiments.

Theoretical studies were performed, based on rate equation formalism developed for the populations of various isotopes in various levels of the three-step photoionization scheme, incorporating the effect of all atomic, vapour and laser parameters on the photoionization dynamics. They were solved numerically, to obtain the fractional ionization (f) and selectivity (w) of the process. Further, the effect of important operating parameters, i.e., saturation of the first step (See Fig. 2), vapour collimation and magnetic field, on fractional ionization and selectivity of the even isotopes was investigated. These results provided quantitative estimates of these quantities, for a realistic three-step photoionization experiment, where the first step is made isotope selective. The variation of selectivity and fractional ionization as a function of the operating parameters was studied, in terms of the IS and HFS of the first step. The experimental and theoretical investigations are in general, applicable to isotope selective photoionization of lanthanide group of elements.

Conclusions

The work carried out in this thesis, has resulted in the development of sophisticated spectroscopic techniques, new IS and HFS data for uranium isotopes and their relation to nuclear parameters, identification of several new AI levels of uranium and demonstration of isotope selective photoionization of natural erbium. The total work opens up several new and interesting possibilities in atomic spectroscopy and laser-atom interaction processes.

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References