Indigenous Development of High-resolution Atomic Beam Fluorescence Spectroscopy Facility for Precision Measurements of Isotope Shifts and Hyperfine Structure

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Abstract
A compact and versatile ultra-high vacuum chamber in combination with a resistively heated graphite tube atomic beam apparatus for sub-Doppler fluorescence experiments has been designed and developed. The spectral resolution achieved in the developed system is more than adequate to clearly observe all the sub-Doppler hyperfine features in the D2 spectrum of rubidium. The compact interaction geometry, flexibility in changing the atomic beam source and simplicity of the entire set up makes it useful for the measurement of isotope shifts and hyperfine structure with a precision of <100 kHz.

The developed system is versatile enough and can be adopted to carry out high resolution atomic fluorescence experiments on radio-active isotopes after making minor modifications to ensure radiation safety. The spectral resolution and small sample size requirement would enable accurate measurements of nuclear parameters such as nuclear size, deformation, magnetic moments, quadrupole moments, spins etc. This finds applications in AVLIS and ADSS programmes.

Keywords: Atomic beam fluorescence, laser spectroscopy, isotope shift, hyperfine structure, sub-Doppler spectroscopy, laser isotope separation, radioactivity, AVLIS, ADSS, nuclear structure

Introduction
High-resolution optical spectroscopy has been used to determine isotope shifts and hyperfine structure for large number of isotopes, both stable and unstable, and this has led to valuable information about nuclear properties such as spin, magnetic moments, nuclear charge radii and nuclear shape. Measurement of isotope shifts and hyperfine structures of an isotopic chain is of interest for departmental applications. For example, measurement of isotope shift between $^{230}$U - $^{234}$U, $^{232}$U - $^{238}$U along with oscillator strength information of various transitions allow one to arrive at appropriate photoionization schemes for AVLIS [1-2]. Considering its importance, continuous efforts are being made to record emission spectrum of $^{232}$U – $^{238}$U for deriving isotope shifts [3].

Measurement of isotope shifts of stable isotopes; unstable / radioactive nuclides of an isotopic chain provide information on nuclear size, shape and spin. Such laboratories already exist worldwide in countries like Russia (FLNR), Korea (KAERI), Germany (GSI), TRIUMF (Canada), Finland (University of Jyvaskyla) to name a few.

In the Indian scenario, and more particularly in DAE context, the requirement of the nuclear data of various target materials (e.g., $^{209}$Bi, $^{208}$Pb, $^{207}$Pb, $^{206}$Pb, $^{185}$W, $^{184}$W, $^{183}$W, $^{182}$W, $^{181}$Ta, Zr, Sn, Hg, U, Pu, F, Cl, Na, Fe, Al), minor actinides ($^{237}$Np, $^{238}$Np, $^{241}$Am, $^{242}$Am, $^{243}$Am, $^{242}$Cm, $^{243}$Cm, $^{244}$Cm, $^{245}$Cm,
246\textsuperscript{Cm}, 248\textsuperscript{Cm}), long lived fission products, fuel materials, structural and shielding materials used for ADSS is very essential and is also well realized [4]. Measurement of isotope shifts using high precision laser spectroscopy is an excellent technique for the determination of few nuclear properties.

In fact, recently, the radius of the proton was re-measured using Lamb shift measurement of muonic hydrogen and it has been reported that the proton radius is 0.84184(67)fm (which is 4% smaller than the CODATA value of 0.8768(69)fm. This is the most accurate proton size measurement ever done [5].

High precision $2s$ $^2S_{1/2}$ – $3s$ $^2S_{1/2}$ Doppler-free two photon spectroscopy of natural ($^6\text{Li}$ and $^7\text{Li}$) and radioactive ($^6\text{Li}$, $^7\text{Li}$ and $^{11}\text{Li}$) has shown that the nuclear charge radii decreases continuously from $^6\text{Li}$ to $^7\text{Li}$, while a sudden increase in the nuclear size is observed in case of $^{11}\text{Li}$ resulting in the formation of "$^{11}\text{Li}$ halo nuclei" [6]. The above examples illustrate the ability of precision laser spectroscopy methods for deducing certain nuclear parameters.

Resonance fluorescence of atoms in an atomic beam allows one to precisely measure hyperfine splitting and isotope shift with high accuracy. The resolution and sensitivity of the atomic spectra depends on the laser parameters (frequency jitter, drift, power), atomic beam parameters (atomic beam diameter, mean free path, degree of collimation, Doppler shift, background radiation from source) and detection parameters (fluorescence spot size, signal collection efficiency, signal to noise ratio) etc [7,8].

The salient features of the atomic beam fluorescence setup developed at CCCM, Hyderabad, have been described briefly in the current article, addressing all the issues mentioned above.

**System description**

CCCM has procured cw ring dye laser and cwTi:Sapphire laser systems under the X\textsuperscript{th} plan project as part of RIMS programme. The lasers have a frequency jitter of $< 50$kHz when they are actively locked to the temperature stabilized reference cavity. The high resolution lasers can be tuned over a wide spectral range for carrying out isotope shift and hyperfine structure measurements of isotopes of several elements.

Usually the dominant contribution to the atomic spectral width is from Doppler broadening, which often limits the resolution. However, in order to measure isotope shifts and hyperfine structures of constituent isotopes for a given transition of an element, it necessary to employ sub-Doppler or Doppler-free high resolution laser spectroscopy techniques. To overcome the limitation set by Doppler broadening, primarily three techniques are used: a) the crossed-beam method; b) saturation absorption spectroscopy; and c) the two-photon spectroscopy. However, crossed-beam method is more versatile and allows one to record sub-Doppler spectrum with very small laser powers (in mW levels). In this technique, the laser beam intersects the atomic beam at right angles. A narrow vertical slit collimates the atomic beam to give a small angular spread. Collimation reduces the Doppler broadening by a factor determined by the angular spread. This technique allows one to unambiguously resolve and identify all hyperfine components of odd isotopes which enables precise measurements of isotope shifts and the hyperfine structure constants.

Development of sub-Doppler high resolution laser atomic beam fluorescence spectroscopy technique requires development and integration of a suitable atomic beam source, interaction chamber and fluorescence detection. A high-resolution atomic beam fluorescence spectroscopy facility has been designed and developed (Fig. 1). A resistively heated graphite tube atomic beam source with a collimation ratio of $-20$ was designed, tested and integrated into a compact interaction chamber for atomic beam fluorescence experiments (Fig. 2). The laser-atom interaction chamber and the source have been designed so as to achieve sub-Doppler resolution. The source chamber and the interaction chamber
are connected by a long tube which forms one of the six ports of the cuboidal chamber. This tube houses a collimator block. This collimator block additionally functions to establish a differential pumping between the source and the interaction chamber. A series of light baffles are used for minimizing the background radiation from atomic beam source. Additionally a light baffle comprising of graphite pipe has been inserted perpendicular to the atomic beam axis at the interaction region which...
further minimizes the back ground radiation from the source. Atomic beam interacts with the laser orthogonally to reduce the Doppler broadening. The incident laser beam is retro-reflected into the laser axis to eliminate the Doppler shifts due to residual angle (due to misalignment) between atomic beam and the laser. This ensures that the fluorescence peak lies within 100 kHz of the resonance center. Due to the elimination of systematic shifts, hyperfine splittings and isotope shifts could be measured with high accuracy.

The advantage of the source is that it requires very small amounts of sample (few mg) for achieving high signal to noise ratio (> 1000). The compact size of the source makes the turn-on and the turn-off times relatively less in comparison to the number of hours of operation of the atomic beam. The turnaround time is typically about 2 hours.

**Experimental**

Light resonant with the atomic transition was generated by a DPSS pumped cw-ring Ti: Sapphire laser having a frequency jitter of <50 kHz. A small fraction of the laser light is fed to a wavemeter with a resolution of 30 MHz to monitor the laser wavelength. The wavemeter is calibrated using a stabilized reference laser (frequency-stabilized He-Ne laser). The resonance fluorescence was collected using a Czerny Turner monochromator. This monochromator minimizes the amount of stray light reaching the photomultiplier tube and thus the S/N ratio of the fluorescence signal is significantly improved. The atomic hyperfine spectrum was recorded using a digital storage oscilloscope. The dimension of the fluorescence spot has been imaged through the view port and is found to be—3mm x 6mm.

**High resolution sub-Doppler spectroscopy of Rubidium and Potassium isotopes:**

**a) Rubidium**

To evaluate the spectral resolution and sensitivity of the developed system, experiments have been carried out on Rubidium and potassium atomic beam. The atomic beam of rubidium and potassium is produced by thermal evaporation of a mixture of rubidium chloride and potassium nitrate placed in a graphite crucible. Due to the resistive heating, the oven heats up to temperatures of about 1000 K. At this temperature a sufficiently dense atomic beam of Rb is produced. The atomic density is estimated to be $\sim 10^{10}$ atoms/cm$^3$ while diameter of the atomic beam is $\sim 10$mm. The atoms leave the oven through a small hole of 2.0 mm diameter. The laser light from a DPSS pumped Ti:Sa laser intersected the atomic beam orthogonally at about 20 cm downstream.

![Figure 3: Experimental (A) as well as the theoretical (B) laser induced fluorescence spectrum for the $5S_{1/2} - 5P_{3/2}$ D2 transition of rubidium spanning the entire range covering the hyperfine spectrum of $^{87}$Rb and the $^{85}$Rb isotopes. The sub-Doppler peaks of both the isotopes are seen whose FWHM is $\sim 26$ MHz.](image-url)
from the oven. The background pressure in the vacuum system was about $5 \times 10^{-7}$ mbar. The fluorescence light is imaged on to a low resolution monochromator and is detected using a photomultiplier tube (PMT) which is placed above the intersection region of atomic beam and laser beam. The detected laser induced fluorescence for $5^2S_{1/2} - 5^2P_{3/2}$ D2 transition of Rubidium is shown in Fig. 3A. Atomic beam fluorescence spectrum for the entire range of the $^{87}\text{Rb}$ and the $^{85}\text{Rb}$ isotopes has been recorded. The theoretical spectrum has also been simulated for comparison with the recorded spectrum (Fig.3B). The best spectral resolution that could be achieved after optimizing the frequency separation between the hyperfine components of D2 transition of Rb, the hyperfine separation between the components of K isotopes is much smaller ($30 - 55$ MHz). The entire fluorescence spectrum of $4^2S_{1/2} - 4^2P_{1/2}$ D1 transition of the $^{39}\text{K}$ and $^{41}\text{K}$ isotope has been recorded in two different frequency ranges.

Adjacent hyperfine components of the $^{41}\text{K}$ isotope could not be resolved (Fig.5) due to very small separation between the hyperfine components ($30$ MHz). To completely resolve the components, the spectral resolution needs to be further improved; however, it can be done only at the cost of sensitivity. The hyperfine components of the $^{39}\text{K}$ isotope could be partially resolved (Fig.5) due to relatively larger separation ($55$ MHz).

The experimental design and conditions for the production of collision free atomic beam have been optimized and the spectral resolution achieved clearly establishes the observation of all the hyperfine components of both the Rb isotopes.

**b) Potassium**

Laser induced fluorescence from atomic beam of potassium has also been recorded. The laser was operated at a power of 23 $\mu\text{W}$ and the temperature of the atomizer was maintained at $-950$ K for obtaining sufficient density of atomic beam. Unlike the power was $-16$ MHz (Fig. 4) and is limited by the angular spread of the collimator.

The hyperfine components of the major $^{39}\text{K}$ isotope originating from the lower $F=2$ hyperfine level have been shown in Fig.6. As it can be seen in the Fig.6, the hyperfine components, having similar intensities and frequency separation of $-55.5$ MHz could be resolved partially. This clearly shows the limit of the resolution of the system in the present configuration. It is possible to improve the resolution further by the control of angular spread of the atomic beam. Minor design modifications can be made on the system such as type of atomizer required, degree of

![Fig. 4: The hyperfine spectrum of the $^{87}\text{Rb}$ isotope for the $5^2S_{1/2} - 5^2P_{3/2}$ D2 transition. The FWHM of the transition is observed to be $-16$ MHz.](image1)

![Fig. 5: The hyperfine spectrum of potassium isotopes for the $4^2S_{1/2} - 5^2P_{1/2}$ D1 transition recorded for a laser power of 23 $\mu\text{W}$.](image2)
Fig. 6: The sub-Doppler peaks for the $^{39}$K isotope for the $4^2S_{1/2} - 5^2P_{3/2}D1$ transition which has been recorded for a laser power of 23 μW.

resolution desired etc on case by case basis and this would enable one to carry out precision isotope shift and hyperfine structure measurements on most of the isotopes relevant to the department.

Conclusions

We have developed a high-resolution atomic beam fluorescence spectroscopy facility for precision measurement of isotope shifts and hyperfine structure. We have achieved a spectral resolution of $16 \text{ MHz}$ for the $5^2S_{1/2} - 5^2P_{3/2}$ Rubidium D2 transition. We have also recorded the sub-Doppler spectrum of the $4^2S_{1/2} - 5^2P_{1/2}D1$ transition of potassium for the $^{39}$K and $^{41}$K isotopes and measured the hyperfine splittings.

The setup can be readily adopted for measurements on radioisotopes with adequate radiation safety measures, which can find applications in AVLIS and experiments to derive nuclear data of short lived isotopes relevant to ADSS program.

References


