Isotope Separation

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Atomic Vapour Diagnostics Using Tunable Diode Laser-based Absorption Spectroscopy

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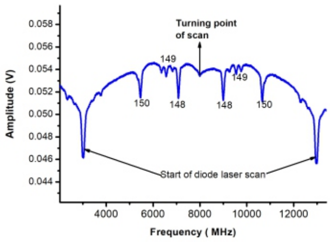


Absorption spectrum for 8^0 vapour divergence

ABSTRACT

In this paper we are presenting our recent results based on the diode laser-based absorption spectroscopy of weak atomic transition of Sm atoms carried out in the process chamber utilized for laser based medical isotopic separation. Identification of the isotopes, Doppler spectral width along with the level population of atoms involved in the process were successfully evaluated.

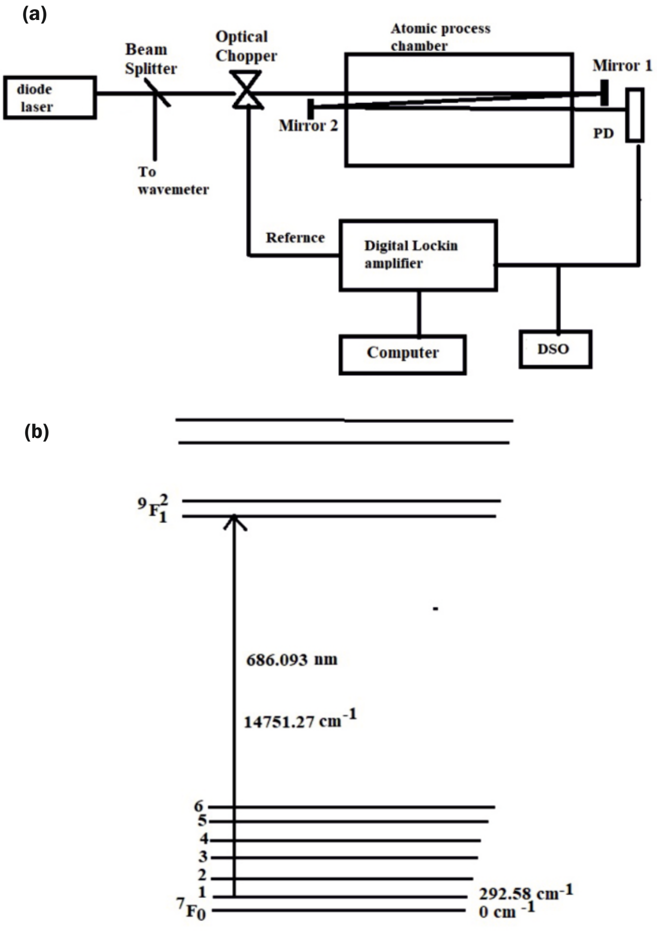
KEYWORDS: *Tunable diode laser absorption spectroscopy (TDLAS), Atomic vapour Characterization, Doppler width, Atom number density, Atomic vapour laser isotope separation*

Introduction

Collimated atomic and molecular beams finds applications in various research activities such as study of laser-atom interaction in laser spectroscopy, plasma physics and collision physics. Beam Technology Development Group, Bhabha Atomic Research Centre (BARC) is pursuing the development of the Atomic Vapour Laser Isotope Separation (AVLIS) process for separation of ^{152}Sm, ^{176}Yb and ^{176}Lu from natural feed for medical applications. The important parameters for characterizing the atomic beam are its atom number density and spectral width. Atom number density of atomic vapor has been estimated coarsely using techniques like gravimetric and source temperature based vapour density estimation. Diode laser-based absorption method is an elegant method to estimate the atomic vapour density, level population densities, spectroscopic information such as Doppler width of the transition and Isotopic shifts and HFS splitting of isotopes under study[1]. In this paper, we present tunable diode laser-based absorption spectroscopy of (spin forbidden) transition of samarium with weaker cross-section for the estimation of Doppler width, atomic level population and total density of samarium atoms in the process vapour generator. This technique can be used as a diagnostic tool for characterizing new atomic vapour source generator and online atomic vapour diagnostics for Samarium LIS process.

Experimental Setup

The experimental setup for the diode laser absorption study of samarium neutral atom is given in Fig.1(a). Commercial diode laser from Sacher Lasertehnik Littman series (TEC-500 model) at 686.093 nm was used as a probe laser for the absorption study. Samarium vapour are generated from a radiatively heated linear evaporating source. Three linear sources of 50 mm length at pitch of 70 mm are used together to generate vapour of ~200 mm length in the laser-atom interaction zone. Samarium Source in crucible was heated around 800°C in a high vacuum chamber and the emanating atomic vapor was collimated using a series of four collimators. External Cavity Diode Laser (ECDL) at 686.093 nm connects transition from metastable state at 292.58 cm^{-1} ($^7F_1$) above the ground state to $^9F_1$ transition of samarium atom as shown in Fig.1(b). The transmitted signal was measured with photo diode (PD) and lock-in amplifier was used to detect the phase-lock detection of the amplitude modulated PD signal. As the laser frequency scanned across the transition of Sm atom,



Samarium ground state multiplets and transition excited by Diode laser

Fig.1: (a) The experimental setup. (b)Transition for absorption study.

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absorption dips in the transmitted intensity were observed corresponding to transitions of isotopes and their HFS components. The percentage of absorption depends on the product of lower level population density, absorption cross section and length of interaction. The probe laser intensity is kept below saturation intensity ($h\nu/\sigma\tau$) of the transition to avoid intensity depended nonlinear absorption. Here symbol ν is the frequency of probe laser, σ and τ corresponds to absorption cross-section and radiative life time respectively.

ECDL wavelength was tuned by providing triangular pulses to PZT transducer attached to wavelength tuning mirror of grazing incidence cavity. Corresponding wavelength/ frequency scan without any mode hop or frequency jump was monitored with help of wave meter of WS-6 model from High Finesse. The population distribution of samarium atoms in ground state and its nearby metastable septets, at a particular source temperature, was evaluated using Boltzmann distribution. As the multiplets are closely spaced in energy, the appreciable population of higher levels at higher temperature is expected. For near operating temperature for Laser Isotope Separation (LIS) process, $^7F_1$ has more population followed by $^7F_2$ and ground state $^7F_0$. Samarium has seven isotopes with even isotopes of 154,152, 150, 148 and 144 and two odd isotopes of 149 and 147 with nuclear spin of 7/2. Diode laser experiments in the wavelength region of 630- 690 nm reported in literature were carried with higher number densities of ~10^{12} per cc using heat oven pipe of longer source length (~100 cm) for measurable absorption signal for Samarium [2,3]. In the process chamber of Sm isotope separation used in this study has nominal number density of ~10^{11} per cc and 21 cm of atomic vapour length [4]. The expected absorption is less than 1%, due to low value of absorption cross section (~10^{-15}cm^2) of the transition. Hence, the experiment was designed with three optical pass configuration to obtain good signal to noise ratio of the absorption signal.

Results & Discussion

Fig.2 shows the lock in amplifier output for the vapour source with half angle divergence of 8^o. Fig.3 shows simulated absorption cross-section spectrum for the transition at 686 nm for all isotopes along with various Doppler widths (100 MHz & 250 MHz) with available data on isotopic shift, Einstein's A&B coefficients, or Oscillator strengths (f_{osc}) [5], hyperfine splitting and the natural abundance. The peaks of two isotopes with even mass numbers i.e. ^{150}Sm and ^{148}Sm have been identified, along with well resolved HFS signal of ^{149}Sm with its three components in Fig.2 by comparing with simulated spectral signatures shown in Fig.3. The frequency separation between ^{150}Sm and ^{148}Sm (1616 MHz) is used as frequency marker for this spectrum and the time base was recalibrated to frequency and shown in the Fig.2.

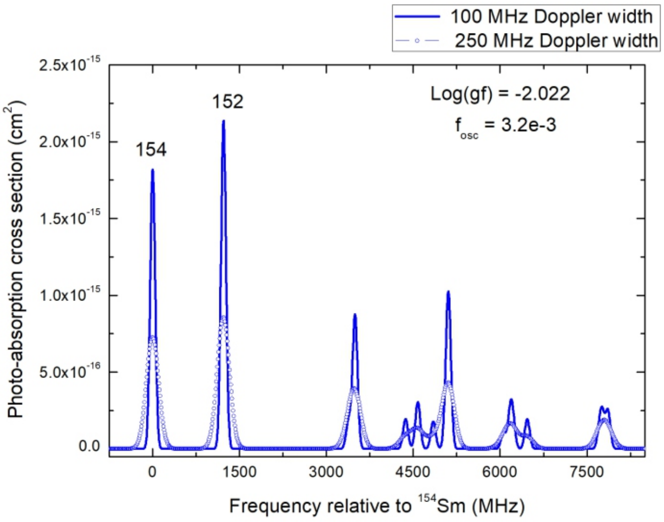

Fig.3: Simulated absorption spectrum at 686 nm.

Even isotope Sm-148 was fitted with Gaussian function and the spectral width of 92 MHz and peak absorption is ~4.89% was evaluated for three pass configuration. Here, Doppler width is dominant broadening mechanism over natural width of ~0.449 MHz as reported. Doppler spectral line broadening is an important process parameter for optimization of laser atom interaction[6] for high ionization yield of target isotope with high spectroscopic selectivity.

Diode laser absorption experiment was repeated for vapour source with half-angle divergence of ~18^o; which is expected to have higher vapour utilization of the evaporated vapour. Increased Doppler width is expected for atomic beam with 18^o half angle divergence which results in reduced peak absorption. Hence, the transition resonance is shifted from "Sm-150 & Sm-148" pair to "Sm-154 & Sm -152" pair of transition due to their higher abundance.

By red detuning the diode laser from centroid of the transition (Fig.3), we could capture three isotopic peaks with very good signal to noise ratio. Fig.4 shows the three pass absorption spectra with the vapour source at 835^{o}C and with half-angle divergence of ~18^o and at the interaction height 25 mm to 35 mm from the collimator. Comparing with the

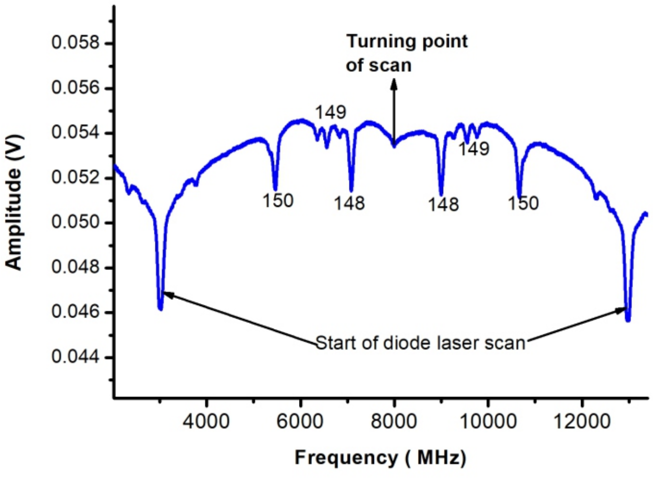

Fig.2: Absorption spectrum for 8^o vapour divergence.

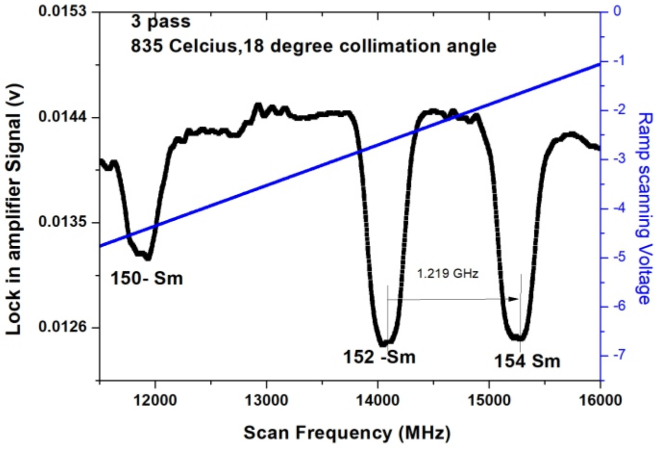

Fig.4: Three Pass Absorption spectra with 835^{o}C of half angle of divergence of 18^o.

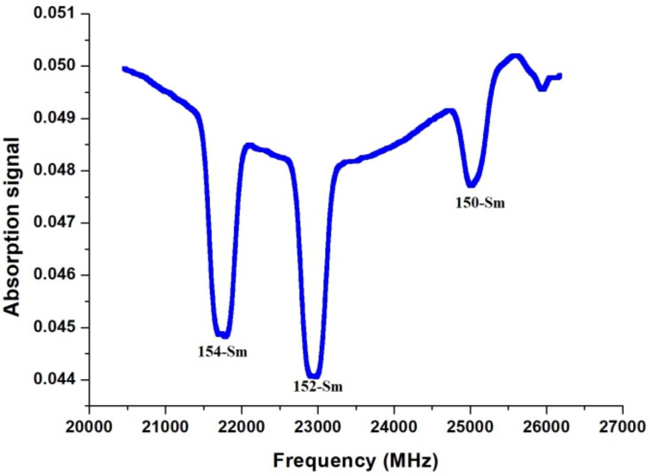

Fig.5: Two-pass absorption spectra at ~25 mm height.

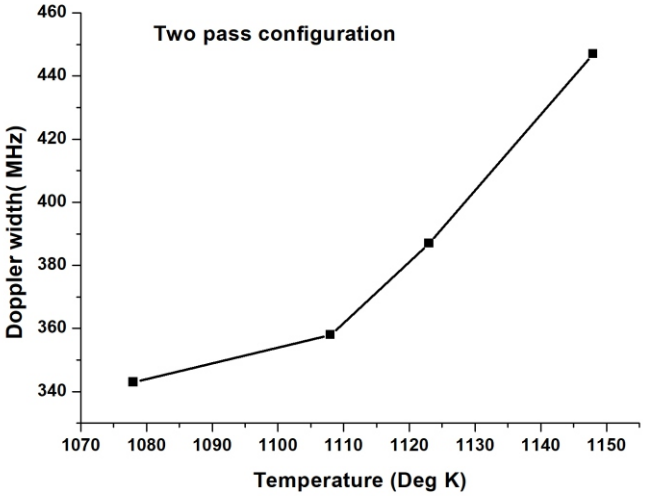

Fig.6: Doppler width Vs temperature.

simulated spectrum with position and amplitude, the peaks of 150-Sm, 152-Sm and 154-Sm were identified. The peak absorption of ~15 % for Sm-152 was obtained in a 3-pass configuration; which depends on the isotopic abundance, Doppler width and atom density. Considering all the contributions between Sm-148 and Sm-152, may be concluded that the increased peak absorption due to enhanced atom number density resulted in three-fold increase in absorption signal of Sm-152. Doppler width was estimated with Gaussian fitting from the absorption spectrum to be ~330 MHz. The peak absorption signal at Sm-152, in single pass configuration, was measured to be ~4%.

With 18-degree half angle for vapour, the absorption spectra were also obtained with two pass optical configuration, as shown in Fig.5 at 835°C of source temperature. Study of Doppler width and estimation of number density with four sets of temperature were carried out. The absorption signal, using two-pass optical configurations is measured to be ~10%. Effect of temperature on Doppler width was studied and is plotted in Fig.6 for Sm-152 isotope.

From the measurement of Doppler width and peak absorption (transmission), atom number density of the levels involved in the transition can be estimated using equation(1)[7].

$$k_{\vartheta 0} = \left(\frac{2}{\Delta\vartheta}\right)\left(\frac{\ln 2}{\pi}\right)^{\frac{1}{2}}\left(\frac{\lambda^2}{8\pi}\right)\left(\frac{g_2}{g_1}\right) N\ A \qquad (1)$$

Where K_{v0} is the absorption coefficient at the peak, which can be calculated from Beer-Lambert's law of absorption. Doppler width (Δv) and peak absorption estimated from the Gaussian fitting of individual isotopic (even) absorption profile. The constants g_1 and g_2 are degeneracy parameter of the lower and excited state for this J 1→1 transition. The constant A in equation 1 is Einstein's 'A' coefficient[3,5] for the transition is reported in the literature as $0.4487\text{x}10^5$ sec^{-1}.

From above equation, population at J=1 of ground state septet is estimated. From level population, isotopic populations derived and further based on the even isotope abundance, total population densities have been estimated. Fig.7 shows the variation of number density as function source temperature measured by DLAS method, which displays an increasing trend as expected.

Conclusion

Here, we have presented diode laser-based absorption technique as the diagnostic tool for samarium atomic vapour characterization for Samarium-LIS process chamber. As the absorption cross-section is very weak for samarium atom in the available diode laser wavelengths, we have used multi-pass configurations to achieve a good absorption signal. Important LIS process parameters such as Doppler width, available atomic level population and total atomic number density were

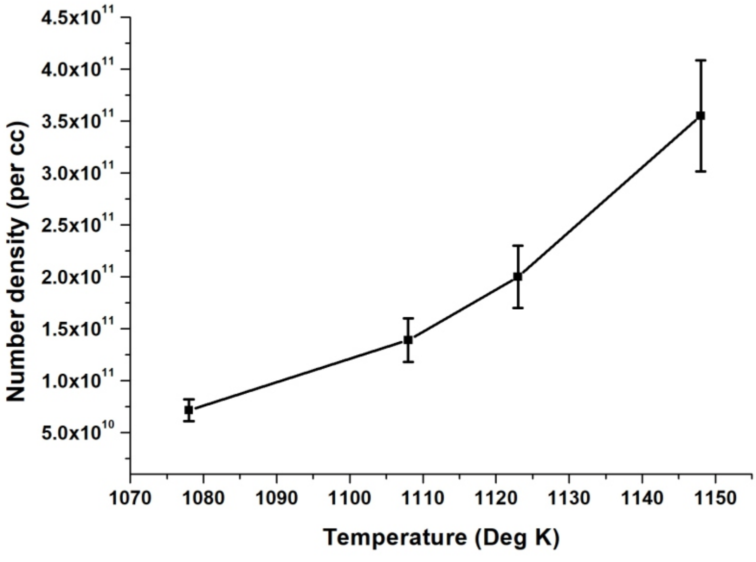

Fig.7: Total atom number density VS Temperature.

evaluated. With 18 degree half angle divergence of atomic vapour, absorption signal was obtained with Sm-152 in single pass configuration itself, firmly establishes that this technique can be adapted for online neutral density measurement for Samarium LIS process.

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