

A new technique for high resolution emission spectroscopy of rare and radioactive isotopes

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Abstract : A technique for high resolution emission spectroscopy of rare and radioactive elements in normal cold laboratory conditions is described. This technique makes use of a sample size of $\sim 100 \mu\text{g}$, mixed with suitable carriers and loaded on a new design of an all metal and liquid nitrogen cooled hollow cathode discharge lamp. The technique has been successfully used for the high resolution spectroscopy of uranium isotopes.

Keywords : High resolution spectroscopy, hyperfine structure, isotope shifts.

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1. Introduction

In recent years there has been much interest in the high resolution spectroscopy of rare and radioactive elements [1–3]. These investigations are concerned with the measurement of isotope shift (IS) and hyperfine structure (Hfs) which provide a very useful information on the multipole moments of the nuclear ground and isomeric states. Such studies of nuclear physics *via* atomic physics have been immensely successful owing to the high sensitivity and high resolution provided by the optical spectroscopic techniques.

Implementation of a high resolution spectroscopic technique requires as a first step a stable source in the form of a collimated atomic beam or a hollow cathode discharge lamp [3,4]. Although these sources have been extensively used in the past, the conventional designs of such sources are inadequate for high resolution spectroscopy of rare and radioactive elements. The basic constraint faced in such efforts is the limitation on sample size. For rare isotopes the low production yields make it necessary to perform studies with very small quantities of the sample. In case of radioactive samples, such as hard γ -emitters, the radiological consideration

demand the use of minimum sample size to keep the shielding requirements at a manageable level. High resolution spectroscopy of such samples, therefore, calls for atomic sources operating at microgram levels of the sample. Recently collimated atomic beams working with microgram levels of samples have been developed for high resolution laser spectroscopic studies of rare and radioactive atoms [3]. These however, require expensive and elaborate equipments, especially for refractory elements. In this paper we report a new technique of high resolution emission spectroscopy based on Recording Fabry Perot Optical Spectrometer (REFPOS) [5] and on a novel design of a hollow cathode discharge lamp [6] that works with μg size samples. Using uranium as an example, we show that high resolution emission spectroscopy can be performed with less than $50 \mu\text{g}$ of a single even isotope and $\sim 100 \mu\text{g}$ of a single odd isotope. This method can be conveniently used for rare and radioactive elements.

2. Experimental

The basic equipment that was developed and used in the present work is a liquid nitrogen cooled all metal HC DL

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developed in our laboratory for high resolution spectroscopy of radioactive elements [6]. The design is based on the concept of a hermetically sealed device which can be loaded with a radioactive material in a protected facility and transported to an inactive area housing the spectroscopic equipments without any stringent safety requirements. Operationally this concept is superior to the atomic beam source since the active material remains confined within the HCDL and does not pose the usual problem of containment of radioactivity. The cathode of this HCDL is a copper cuvette which can hold the sample in a metal or oxide or nitrate form. Usually the sample requirement is in the range of 10 mg, which although is much smaller than that for a metal cylinder in a conventional HCDL [4], is prohibitively large for rare and radioactive samples. To operate the discharge at microgram levels of charge, we used carriers which provided a suitable matrix for a stable emission lasting for a couple of hours at ~100 microgram level. Typically, a 50/50 wt% mixture of silver and tantalum powder provided an excellent matrix for uranium [7]. Other matrices which we have used included mixtures of silver, graphite and titanium. The choice and the composition of the carrier was arrived at by a number of experiments aimed at optimising the emission from the HCDL. In the experiments reported in this paper, we mixed 100 microgram of uranium in oxide or nitrate form with 5 mg of matrix and coated this on the inner side of the copper curvette.

High resolution spectroscopic studies were carried out using REFPOS which provided resolution typically $\sim 10^6$. Such a resolution is sufficient for high resolution spectroscopy of actinides and lanthanides. In this work, the emission from the HCDL was focussed on the external slit of the 2 m monochromator which preselected a spectral line for high resolution study through the Fabry Perot interferometer assembly [5]. The Fabry-Perot etalons used were with $\lambda_{\text{max}} = 540 \text{ nm}$; the spectra were recorded with etalon spacers of 10 mm and 12 mm thickness which provided free spectral range of 500 and 415 mK respectively. ($1000 \text{ mK} = 1 \text{ cm}^{-1}$).

3. Results and discussion

The HCDL loaded with ~100 microgram of natural uranium oxide was optimised by varying the buffer gas (usually neon) pressure and discharge current. Usually, the HCDLs are operated under the conditions which ensure a high sputtering rate to obtain large intensity for atomic lines of interest. In the present case high sputtering rate would tend to limit the life of the HCDL due to sample at μg level. At the same time, the sputtering rate must be kept sufficiently high to obtain reasonable line intensities for high resolution emission

spectroscopic work. Role of the carrier matrix is extremely important in this regard. In order to obtain the optimum operating conditions, the discharge parameters were varied and the HCDL emission was recorded using the high resolution monochromator in 5910–5920 Å wavelength region. This spectral region included in strong uranium line at 5915 Å and a neighbouring neon line at 5913 Å. The enhancement of the intensity ratio $R = I(\text{U})/I(\text{Ne})$, was used as an indicator of the sputtering rate. Here $I(\text{U})$ and $I(\text{Ne})$ refer to the intensities of the uranium and neon lines respectively. As regards the lifetime of the HCDL, three hours of stable operation was set as a necessary condition. Results of these studies for uranium charge down to 100 microgram level revealed that optimum conditions corresponded to buffer gas pressure of ~3 torr and discharge current ~60 mA. In Figure 1, we show the ratio R , obtained as above, as a function of the uranium charge under optimum operating conditions. It may be noted from this figure that even at ~100 microgram levels, the intensity ratio is sufficiently large for high resolution spectroscopic work. Further the linearity of R with respect to the sample size is important in analytical applications.

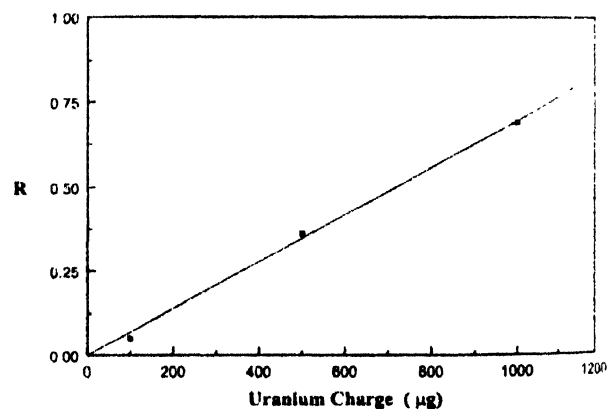


Figure 1. The ratio $R = I(\text{U})/I(\text{Ne})$, $I(\text{U})$ and $I(\text{Ne})$ being the intensities of U and Ne lines at 5915 Å and 5913 Å respectively, as a function of uranium charge (in μg) at buffer gas pressure of 3 torr and discharge current of 60 mA.

To demonstrate the applicability of the technique for high resolution spectroscopy, we loaded the HCDL with a mixture of ^{235}U and ^{238}U (100 microgram each) and under optimum operating conditions the spectrum of the 5915 Å line was recorded on the REFPOS. Figure 2 shows the high resolution spectrum of this sample where the spectral features corresponding to Hfs of ^{235}U ($I = 7/2$) are clearly resolved. The spectral positions of the Hfs components match very closely with those reported in the literature [8]. Figure 2 also demonstrates that for a single even isotope the high resolution spectroscopy can be done at sample size much less than 100 μg . We have confirmed this by recording the high resolution

spectrum of ^{238}U at 50 μg level. For single odd isotope, such as ^{235}U , however the distribution of line intensity over the Hfs components necessitate use of $\sim 100 \mu\text{g}$ sample.

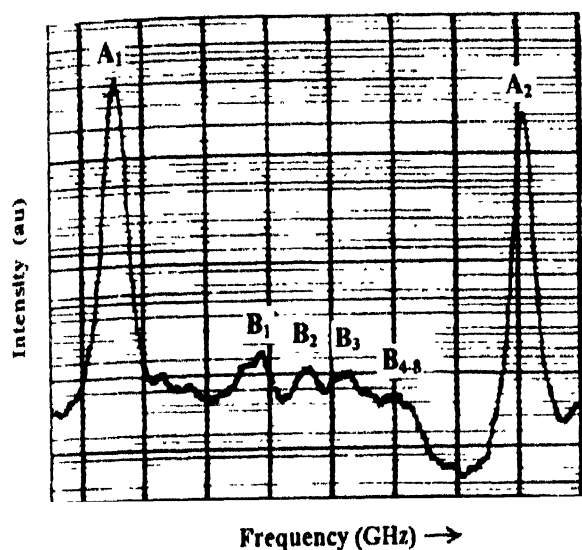


Figure 2. High resolution emission spectrum of the uranium line at 8015.3 Å obtained using 100 μg of ^{238}U and ^{235}U each and procedure described in the text. Spectral features of ^{238}U are marked as A_1 and A_2 (next higher order of the etalon). The separation between A_1 and A_2 is 15.02 GHz. The flag pattern of Hfs of ^{235}U is marked as B_1 - B_8 . Of these, the components B_1 ($F' = 21/2 \rightarrow F = 19/2$), B_2 ($F' = 19/2 \rightarrow F = 17/2$), B_3 ($F' = 17/2 \rightarrow F = 15/2$) are well resolved [8].

In conclusion, we have developed a new method of performing high resolution spectroscopy of rare and radio-active elements by devising a new design of a HCDL and using this as an emission source for high resolution spectroscopy on REFPOS. Since the total charge required for

such a high resolution work is only 100 microgram, this method provides a number of possibilities of high resolution spectroscopy of rare and radio-active elements. Extension of this technique for laser spectroscopy [9] is one such possibility; results of which will be reported elsewhere.

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