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LASER/RF SPECTROSCOPIC TECHNIQUES IN FAST ION BEAMS*

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ABSTRACT

The perturbation free environment and kinematically compressed velocity distribution of ion beams are ideal for high precision spectroscopic measurements using laser techniques. The limitations in the optical techniques are explored and the evolution to laser-rf double resonance techniques is described. Using the double resonance technique a 150-fold improvement in precision is achieved. Problems in applying the double resonance technique are encountered at low rf frequencies. A novel method based upon stimulated resonance Raman spectroscopy is described which circumvents these problems.

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I. Introduction

The advent of the laser has revolutionized precision spectroscopy of ions as has been demonstrated by elegant experiments in traps [1] and beams [2]. However, often, the full resolving power of the laser (typically ~ 1 part in 10^9) is not utilized effectively. This is due in large part to the poor spectral overlap between the laser and the sample with an inhomogeneously broadened absorption profile. Early it was realized that an accelerated ion beam provided a favorable environment for precision laser spectroscopy of ions since, velocity compression results in narrow Doppler profiles; low ion densities yield a perturbation free environment; and mass selection simplifies isotopically rich spectra. Consequently, in the past decade, fast ion beam spectroscopy has been widely used, for example, to measure hyperfine structures [3] as well as Lamb-shifts and fine structures in one and two electron atoms [4-6].

In this paper, fast ion beam laser spectroscopy (FIBLAS) is first described using hyperfine structure of Sc II as an example. The limitations in precision using FIBLAS are then outlined. This is followed by a description of the natural evolution to the laser-RF double resonance (LRDR) technique to improve precision. Limitations to this technique are then presented, followed by the description of a novel technique, utilizing stimulated resonance Raman transition, which can be used to circumvent difficulties with LRDR.

II. Experimental

Figure 1 shows the experimental apparatus used for ion-beam laser interactions at ANL. Much of the apparatus has been described previously [7,8]. Briefly, ions are created, accelerated, mass analyzed and collinearly superimposed with a photon beam. Using this apparatus, all three types of

experiments outlined above (FIBLAS, LRDR and stimulated resonance Raman spectroscopy) are performed. All experiments used photon counting in the probe region for detection.

A. FIBLAS

In standard single photon fast ion beam laser spectroscopy, resonant absorption of a photon is detected. The frequency of the laser photon (ν_L) is then measured and the transition frequency of the ion in its rest frame, ν_0 , is deduced from

$$\nu_0 = \nu_L \gamma(\beta) (1 - \beta \cos \theta) \quad (1)$$

where $\beta = v/c$ and θ is the angle between the photon and ion beam propagation directions. It is, of course, not necessary to know the ion beam velocity in order to obtain a precise measurement of ν_0 , since

$$\nu_0 = (\nu_R \nu_B)^{1/2} \quad (2)$$

where ν_R and ν_B are the absolute frequencies for the red- and blue-shifted transitions, respectively. Unless ν_R and ν_B are measured simultaneously, the absolute error in the measurement will depend on drifts in ion velocity, i.e. acceleration voltage, source conditions etc.

The precision to which the line center can be determined is directly proportional to the linewidth. Ideally, upon acceleration, the width of the initial thermal distribution of velocities in the ion source is reduced by a factor of $R = 1/2 \sqrt{kT/eU}$ where T is the ion source temperature and U is the acceleration voltage. This ideal state is difficult to achieve due to ion source irregularities and the requirement of a highly stable acceleration voltage. Using active feedback, the high voltage has been stabilized to $dE/E \sim 2 \times 10^{-6}$, and has a negligible contribution to the observed width. However, ion source stability over long periods remains troublesome. While ion current may remain stable, the velocity distribution deteriorates markedly

with age, as is illustrated in Fig. 2.

In the measurement of hyperfine structure (hfs) splittings by FIBLAS, it is the difference between two optical frequencies which is important. Again, since these two frequencies are not measured simultaneously, but rather acquired through scanning the laser, the relative positions are subject to source and platform instabilities. At ANL, this limits the precision with which hfs splittings can be measured to ~ 2 MHz, for a typical optical transition of FWHM ~ 50 MHz.

B. LRDR

An energy interval is determined much more precisely by direct measurement than by differences. Hfs intervals lie in the radiofrequency regime and direct measurements can be made using LRDR. The basic principle [9] and apparatus [8] has been explained before, so only the briefest description will be given here. The population of a given hfs level is strongly depleted by optical pumping in the "pump" region (see Fig. 1). This causes little fluorescence to be induced in the probe region unless the depleted level is repopulated in the intervening rf region. The rf frequency at which the repopulation occurs in a direct measurement of the hfs splitting.

Figure 3 shows an example of a LRDR signal. The data is fitted well by the Rabi two-level model. Of special interest is the width of the transition ~ 700 kHz, which is determined solely by the transit time. This represents a ~ 100 -fold decrease in width compared with FIBLAS. In addition, since the LRDR measurements occur at lower frequencies the inaccuracies in the measured resonance position caused by voltage fluctuations are much less severe. As in FIBLAS, the line center is determined by measurement of the red and blue shifted frequencies.

However, difficulties are encountered when applying LRDR at low rf frequencies. Figure 4a shows the result of searching for an LRDR signal at low frequencies. These observations can be quantitatively explained [8] by a simple physical model: the ions are accelerated or decelerated at the ends of the rf-transmission line depending on the phase of travelling rf wave. This acceleration/deceleration effectively broadens the incident velocity distribution which results in a decrease in the number of ions in the velocity class sampled by the laser and thus causes the oscillatory behavior. Figure 4b shows the modelled behavior using measured beam and geometrical parameters. This effect, limits the applicability of LRDR at low rf frequencies.

C. Stimulated Resonance Raman Spectroscopy

It is possible to eliminate the bothersome effects shown in Fig. 4 through the use of stimulated resonance Raman spectroscopy. In this method, the rf is placed on the laser rather than on the ion beam. The principle is illustrated in Fig. 5a. The stimulated resonance Raman transition is a coherent two photon process in a three level Lambda (Λ) configuration. On Raman resonance, the population in the upper-fluorescence level is decreased due to the dominance of the two-photon process which traps population in the two lower levels. A dip in fluorescence is therefore expected when the coherent two photon process occurs. This is shown in Fig. 5b.

In our experiment, the two required optical frequencies are derived from the output of a single ring dye laser using an electro optic modulator (EOM). Since the relative position of the two optical frequencies is generated through a synthesizer, amplifier, EOM chain, it is possible to know the hfs splitting with rf precision. The observed width of the two photon transition, 4.5 MHz, is primarily determined by the transit time limitation (for the 10 cm

interaction length, the Rabi two-level width $0.8/T = 3.7$ MHz). The residual broadening is due to stray fields in the optical interaction region. A longer, better shielded interaction region would clearly improve the precision attainable by this method.

III. Summary

In this paper we have given a very brief overview of the experimental techniques currently used at ANL to make spectroscopic measurements in ion beams. The advantages and disadvantages of the three techniques; FIBLAS, RLDR and stimulated resonance Raman spectroscopy, have been outlined. Although precisions approaching 1 part in 10^9 can be attained using FIBLAS for optical frequencies, lower state hfs intervals are better measured using LRDR. At low rf frequencies, perturbations to the ion beam velocity distribution render LRDR ineffective. Stimulated resonance Raman spectroscopy eliminates the ion beam perturbations and still yields measurements of the lower state hfs intervals with rf precision.

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Figure Captions

Fig. 1. Schematic of the experimental apparatus.

Fig. 2. Laser induced fluorescence spectra of the hyperfine structure of the $3d^2 \ ^3F_4 - 3d4p \ ^3F_4^0$ transition in Sc II. (a) spectrum taken with a clean source having FWHM ~ 70 MHz, $\Delta\nu/\nu \sim 1 \times 10^{-7}$. (b) spectrum taken with a dirty source (anode insulating ring coated with a conducting material) having FWHM 110 MHz.

Fig. 3. Laser-rf double resonance spectrum of Sc II. The optical transition used for pumping and probing is shown in 3a. The rf resonance is shown in 3b. The solid line is a fit to the Rabi two level formula

$$P(t, \omega_0) = \frac{A(2b)^2}{(\omega_0 - \omega)^2 + (2b)^2} \sin^2 \left[\frac{t}{2} \left((\omega_0 - \omega)^2 + (2b)^2 \right)^{1/2} \right]$$

where ω_0 is the line center, A is the amplitude, t is the transit time and 2b the strength of the transition.

Fig. 4a Observed laser induced fluorescence in the probe region as a function of applied frequency to the rf section. rf=power = 1 W, step size = 100 kHz. Optical transition is $P_2(14)$ line of the (0,1) band of $B^2\Sigma_u^+ - X^2\Sigma_g^+$ transition in N_2^+ . Beam energy = 50 keV, beam current = 5 μ A.

b Model for observed oscillations. For details see reference 8.

$$Y = A / \left[1 + B \left(\frac{\sin ax}{ax} \right) \sin^2 (1 + a)x \right]^{1/2}$$

$a = \tau/T$, $x = \omega T/2$. A = unperturbed fluorescence intensity,

T = transit time, B = ratio of perturbed to unperturbed width,

τ = time spent in entrance/exit region.

Fig. 5a Energy level diagram of the hyperfine multiplet of the $3d^2 \ ^3P_2 - 3d4p \ ^3P_2^\circ$ transition in Sc^+ . The arrows represent the optical frequencies required for the two photon resonant Raman transition.

b Stimulated resonance Raman transition in Sc^+ . ν_L was fixed on the $11/2 - 11/2$ transition and $\nu_L - \nu_{RF}$ was scanned across the $9/2 - 11/2$ transition. The superimposed line is a fit of the resonance to two gaussians. The FWHM of the dip is 4.5 MHz.

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Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5