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Single Longitudinal Mode Interaction Between a Uranium Atomic Beam and a Modified Hänsch-Type Pulsed Dye Laser

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Abstract. Laser-induced fluorescence is used as a diagnostic tool for testing the tuning between a longitudinal mode of a nitrogen-pumped dye laser and a uranium spectroscopic level in an atomic beam.
According to a simple resonator model, the tuning instabilities can be ascribed to thermal drifts in the dye-laser system. Problems encountered in attempting a single mode scanning are also described.

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In the early seventies it was generally accepted that nitrogen-pumped Hänsch-type dye lasers did not show axial modes. The presence of these modes was later observed at the Lawrence Livermore National Laboratory [1] and more recently [2] axial modes have been observed also in a modified Hänsch design in which the enlarging telescope is replaced by a prism expanding system. In both works the potential effects of the mode structured laser output are foreseen and discussed, but no experimental data on these effects are reported.
In this work we present experimental results related to the interaction between the single axial modes and an atomic beam, in particular as far as it concerns the frequency drift of such cavity modes, which assumes a dominant role. In fact, if tuning between one of these modes and the atomic transition is assumed, it is necessary to allow for some integration time in order to compensate for the random leakage on the selected mode. Needless to say, this integration time is critical since, in the presence of drifts, the system does not present an ergodic behaviour and even continuous tuning and detuning conditions can easily occur in the measurement time.
Laser-induced fluorescence is the diagnostic tool in these measurements.

1. Experimental Apparatus and Results

The experimental set-up is shown in Fig. 1.
A uranium atomic beam is generated bombarding a cooled natural uranium rod with an electron gun [3]; by means of two slits the atomic beam has a calculated residual Doppler width of about 100 MHz (FWHM).
The nitrogen-pumped dye laser is a commercial Molectron DL-14P in the oscillator-only configuration, equipped with a magnetically stirred dye cell. The vacuum-tight box containing grating, beam expander and intracavity etalon (ICE), for narrowband operation, is nitrogen filled and pressure loop-controlled by an MKS-Baratron system (type 250 A controller and 315 BH 1000 sensor head). This system allows for a slow pressure variation when externally driven by a suitable ramp generator. The whole cavity is supported on an aluminium base plate.
The laser is operated at 10 pps repetition rate and the irradiance in the interaction region is about

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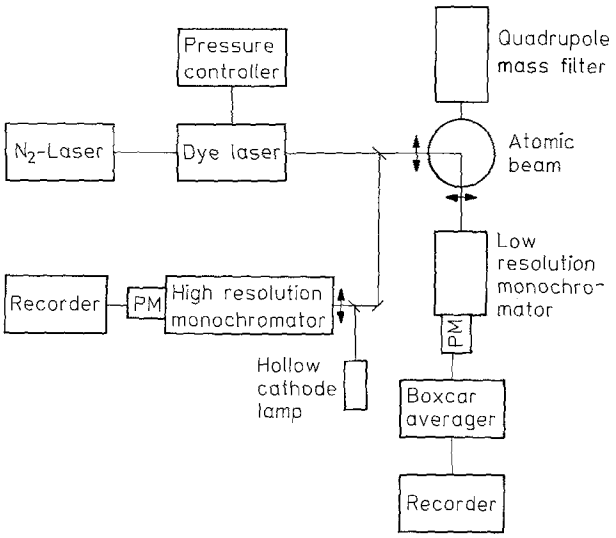

Fig. 1. Experimental set-up

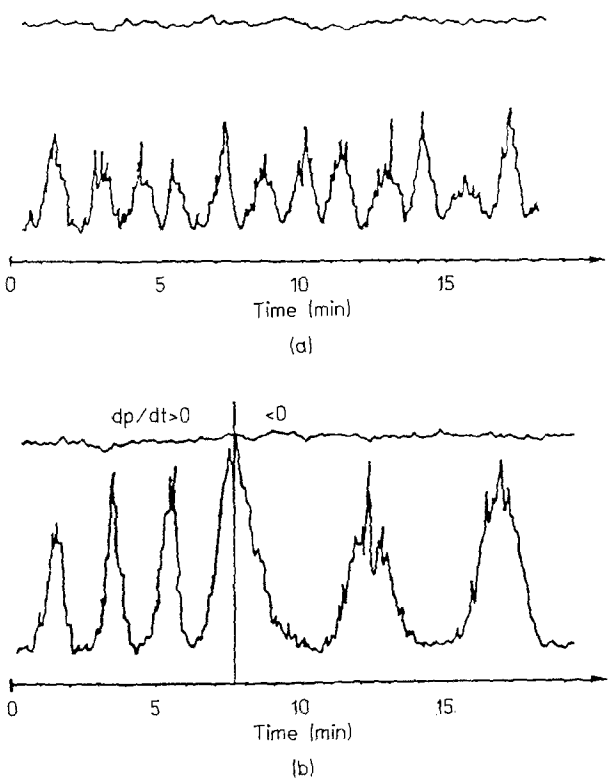

Fig. 2a and b. Fluorescence intensity vs. time. The dye laser was operated in broadband condition (without ICE). The pressure in the laser tuning box is (a) constant (±0.02 Torr), (b) swept at $|dp/dt| = 2$ Torr/min, positive *(left)* and negative *(right)*

150 kW/cm^2 (without ICE). Using the 7-diethylamino-4-methylcoumarin as lasing medium the measured pulsewidth is about 4 ns (FWHM).

The spectral linewidth of the laser beam is visually monitored through a Fabry-Pérot interferometer

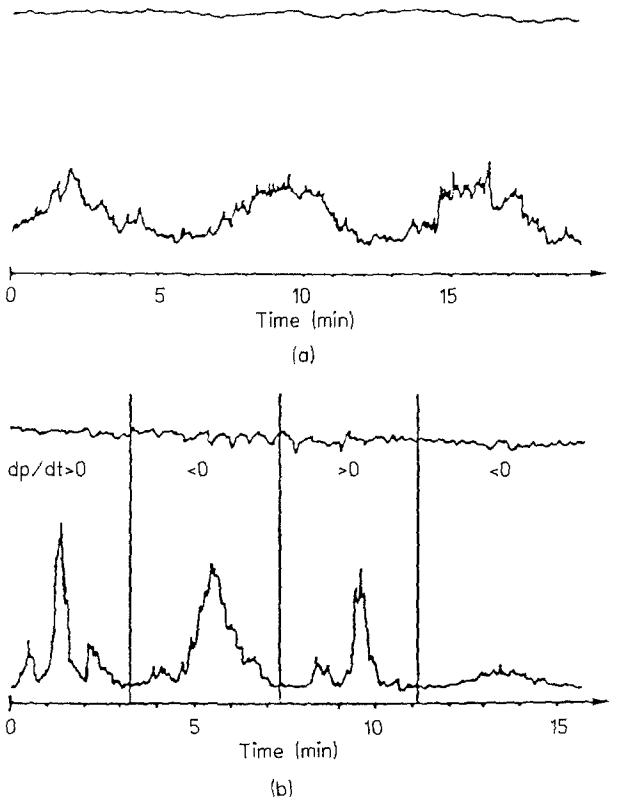

Fig. 3a and b. Fluorescence intensity vs. time. The dye laser was operated under narrow-band condition (with ICE). (a) Constant pressure; (b) scanned pressure at positive and negative *dp/dt*

(Tropel) coupled with a CCTV system; with ICE few modes (about 3–5) are emitted from the cavity and the time-averaged modes envelope is just about 1 GHz. Competition between modes and wavelength drifts are also observed (see later).

The exciting fluorescence wavelength is centered at 439.359 nm, corresponding to the 0–22754 cm^{-1} transition in the uranium atomic levels [4].

The tuning of the wavelength is achieved and controlled with a 350,000 resolving power grating monochromator (Sopra 2000) and a U/Ne commercial hollow cathode lamp.

Fluorescence coming from the uranium atomic beam is focused on the entrance slit of a $f/4$ double monochromator and the PM signal at the 451.6 nm non-resonant fluorescence is sent to a Boxcar averager (EG & G PARC).

In order to discriminate the laser tuning instabilities from the flow-density fluctuations in the atomic beam, a quadrupole mass spectrometer (Balzers QMG 511), is operated on line, settled on the 238 atomic mass.

Experimental data both without (Fig. 2) and with (Fig. 3) intracavity etalon have been collected, in turn, at constant (a) and at variable (b) controlled pressure in the vacuum tuning box of the dye laser. In Fig. 2a

strong fluctuations in the fluorescence intensity signal can be observed, as a result of subsequent cavity-modes tuning on the selected uranium absorption line. This effect could be ascribed to a thermal drift [5] in the laser housing that results in a change of the cavity optical length.

To support this hypothesis we generated a frequency drift by increasing and decreasing the pressure in the vacuum tight box, at constant rate $|dp/dt|$ (Fig. 2b). As expected, because of a monotone thermal drift modes are not equally spaced in the two cases.

Similar results have been obtained when the ICE was installed (Fig. 3a and b).

In all figures the upper trace indicates the atomic density stability.

2. Discussion of Experimental Results

We have tried to quantify the observed temperature dependence moving from the simple dye-laser resonator equation [6]

$$2\sum n_i l_i = q\lambda, \tag{1}$$

from which

$$\frac{\Delta\lambda}{\lambda} = \frac{1}{L}(\sum l_i \Delta n_i + \sum n_i \Delta l_i), \tag{2}$$

where l_i are geometrical path lengths in the dye cell, optical components, air and nitrogen ($i=1,\ldots,4$, respectively); n_i is the refraction index in the path length l_i; q is the axial mode number; λ is the vacuum operating wavelength; and $L=\sum n_i l_i$.

Equation (2) can be rewritten as [7]

$$\frac{\Delta\lambda}{\lambda} = -\frac{\Delta\nu}{\nu} = A\Delta T + B\Delta p_3 + C\Delta p_4, \tag{3}$$

where p_3 and p_4 are air and nitrogen pressures and

$$A = \frac{1}{L}\left\{\sum l_i \frac{\partial n_i}{\partial T} + \alpha_2(n_1 l_1 + n_2 l_2) + n[\alpha_s \sum l_i - \alpha_2(l_1 + l_2)]\right\},$$

$$B = \frac{1}{L} l_3 \frac{\partial n_3}{\partial p_3},$$

$$C = \frac{1}{L} l_4 \frac{\partial n_4}{\partial p_4},$$

in which $n = n_3 \simeq n_4$ and α_2, α_s are the thermal expansion coefficients for optical components and cavity support, respectively. Note that the optical path length in the active medium is increased by the dye cell thermal expansion.

In our case [7], at $T=293\,\mathrm{K}$, $p_3=760\,\mathrm{Torr}$, $p_4=200\,\mathrm{Torr}$ and assuming that all the components feel the same temperature changes, we obtain: $A=5.66\times10^{-6}\,\mathrm{K}^{-1}$, $B=8.10\times10^{-8}\,\mathrm{Torr}^{-1}$, $C=1.66\times10^{-7}\,\mathrm{Torr}^{-1}$.

Now, drifts involve time derivatives and (3) becomes

$$\frac{1}{\lambda}\frac{d\lambda}{dt}\Delta t = A\frac{dT}{dt}\Delta t + B\frac{dp_3}{dt}\Delta t + C\frac{dp_4}{dt}\Delta t. \tag{4}$$

During our experiments the room pressure did not change more than $\pm 2\,\mathrm{Torr}$ so that, in comparison with other terms of (4), we can disregard the component $B(dp_3/dt)\Delta t$. Under this assumption (4) reduces to

$$\frac{1}{\lambda}\frac{d\lambda}{dt}\Delta t = A\frac{dT}{dt}\Delta t + C\frac{dp_4}{dt}\Delta t. \tag{5}$$

An order of magnitude for the experimental coefficient dT/dt can be obtained from our data, both at constant pressure and at variable pressure. Let us consider first the case at constant pressure (Fig. 2a) where the following relation holds for $\Delta t_{q,q+1}$, i.e. the time elapsed between the tuning of two adjacent modes

$$\frac{|\Delta\lambda_{q,q+1}|}{\lambda} = \frac{\lambda}{2L} = A\left|\frac{dT}{dt}\right|\Delta t_{q,q+1} \tag{6}$$

from which

$$\left|\frac{dT}{dt}\right| \simeq 1\times10^{-3}\,\mathrm{K\,s^{-1}}. \tag{7}$$

On the other hand, referring to Fig. 2b, let us indicate Δt_1 and Δt_2 the time elapsed between adjacent modes at increasing and decreasing pressure, respectively. Since the mode spacing $\Delta\lambda_{q,q+1}$ does not depend from the sign of dp/dt, we can write

$$\frac{1}{\lambda}\left(\frac{d\lambda}{dt}\right)_1 \Delta t_1 = -\frac{1}{\lambda}\left(\frac{d\lambda}{dt}\right)_2 \Delta t_2, \tag{8}$$

or, from (5)

$$\frac{dT}{dt} = \frac{C}{A}\left|\frac{dp_4}{dt}\right|\frac{\Delta t_2 - \Delta t_1}{\Delta t_2 + \Delta t_1}. \tag{9}$$

From the experimental conditions $|dp_4/dt| = 2\,\mathrm{Torr\,min^{-1}}$ and $T=293\,\mathrm{K}$, we obtain

$$\frac{dT}{dt} \simeq 0.5\times10^{-3}\,\mathrm{K\,s^{-1}} \tag{10}$$

The two values (7, 10) are of the same order of magnitude and represent physically resonable temperature drifts in the experimental area at different times ($\simeq 0.05\,^{\circ}\mathrm{C\,min^{-1}}$); the positive sign of the thermal derivative can be attributed to the ambient heating from the uranium oven.

When the pressure is scanned in the narrow-band condition (Fig. 3b) the fluorescence intensity does not show the previous regular trend, but a more complicated behaviour. In fact, even if all but pressure parameters affecting the resonator frequencies are constant, from (2) we have

$$\frac{\Delta\lambda}{\lambda} = \frac{\Delta n_4}{n_4 + N} \tag{11}$$

with

$$N = \frac{\sum l_i n_i}{l_4} > 0 \quad i = 1, 2, 3,$$

whereas

$$\frac{\Delta\lambda}{\lambda} = \frac{\Delta n_4}{n_4} \tag{12}$$

drives grating and ICE band pass.
That is to say we are in the presence of two different tuning equations and a pressure scanning unevenly drifts the cavity modes and the envelope of the tuning elements. Of course, simultaneous and independent thermal instabilities generally cannot cancel this drift mismatching, but in the worst case, even enhance it.
As a striking consequence, it is not unusual that the simultaneous superimposition of one cavity mode and the ICE band pass upon the absorption line can be missed and during the whole scanning on the uranium absorption line we cannot detect any fluorescence signal at all (Fig. 3b right).

3. Conclusions

The presence of axial modes in a nitrogen-pumped dye laser has been confirmed by monitoring the fluorescence emitted from an atomic beam; moreover it has also been possible to investigate the temporal drifts of the mode frequency. The results obtained are consistent with a simple modelling of the dye-laser resonator and drifts can be mainly attributed to thermal effects. Consequences of the mode drift can be dramatic in several high-resolution experiments and applications, for instance, when multi-step laser excitation is required. In the scientific literature, some techniques have been described to circumvent this problem when utilizing pulsed lasers. Apart the expensive amplification of a cw dye-laser injection [8], in [9] a dithering was applied to the output resonator mirror in order to eliminate the spectral inhomogenity, but in a time-consuming average. For pulsed dye lasers operated at high repetition rate a feedback system has been successfully used [10]. Presently, this and other laboratories [11] are testing a feedback technique also for low repetition rate. The fluorescence signal itself, we have used as a probing tool, should be a helpful driver for an active stabilization of the dye laser resonator.

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