Molecular Beam Laser Saturation Spectroscopy

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Laser saturation spectroscopy on particle beams is demonstrated as a new sub-Doppler technique. It offers unique advantages regarding resolution and intensity.

A well-known method of achieving very high, sub-Doppler resolution in laser spectroscopy is the saturation technique, with its variants of absorption, intermodulated fluorescence or polarization detection [1]. Line widths of less than 10 MHz can be obtained. Experiments of this type are done using sample cells. They are therefore not suited to refractory species, such as most metal atoms or oxide molecules, or to radicals or excited particles which are rapidly quenched. In this note we describe a novel high-resolution laser spectroscopy technique which is also of the saturation variety but utilizes an atomic (or molecular) beam sample and has therefore almost unlimited applicability to any atoms and small molecules.

In this particular case the hfs of the 6265 Å Xe line was studied [2, 3]. Its lower level is the Xe $^3P_0$ metastable state. The principle of the experiment is shown in Figure 1. The Xe $^3P_0$ beam was generated by striking a DC discharge between the filament F and the nozzle N (Figure 1). It was crossed twice at precisely 90° by the unfocussed beam of a CR 699-21 ring laser (~ 60 mW, ~ 1 W/cm^2). The two crossing points, 20 cm apart, were viewed by an EMI G-26 E 315 UV solar blind and a Hamamatsu R 943-02 visible photomultiplier, respectively. The first was uncooled and located inside the vacuum chamber, the second, outside the vacuum, was cooled and coupled to the light collection optics M1, L via an optical fibre bundle, FB. Tuning the laser to 6265 Å excites the $5p^5[^2P_{1/2}]\,8p$ level (the brackets designate the ionic core). From there, with a mean lifetime of 250 ns, fluorescence occurs to the $^1P_1$ level. This LIF signal, isolated by a filter, is detected by the VIS PM. The UV multiplier detects the $^1P_1 \rightarrow {}^1S_0$ 1296 Å emission which also originates from the 8p excitation as a second cascade step. In addition, the 8p level emits at 4147 Å to $^3P_1$, followed by 1470 Å UV emission to the ground state. Both UV transitions contribute about equally to the UV-PM signal, the former being favoured by the branching ratio, the latter by the UV filter transmission curve. The UV and VIS signals were displayed simultaneously on a dual channel chart recorder, the visible one by direct DC amplification, the UV one via photon counting and a rate meter.

Figure 2 shows a typical result. The bottom trace is a straight LIF measurement of the isotopic shifts of the electronic transition in question for (from left to right) ^{136}Xe, ^{134}Xe, ^{132}Xe, ^{130}Xe (as a shoulder) and ^{128}Xe. The resolution (20 MHz FWHM) is here far inferior to that demonstrated in [3] (6–8 MHz) due to the use of a wider atomic beam and some power broadening. The fact that here a UV signal was used instead of the 6678 Å line as in [3] is merely accidental: In the present case the apparatus was being set up for an experiment involving collision-induced Xe emission.

The important signal is the top trace in Figure 2. Being derived from the second laser/beam intersection (counting in the direction of the atomic beam), it exhibits the effect of optical pumping in the first (upstream) crossing. Just like in conventional saturation spectroscopy, tuning the laser precisely to line center allows both of the counter-propagating laser beams to operate on atoms having a zero velocity component in the direction of the light. These are the atoms in the central ray of the slightly divergent Xe beam. Optical pumping at the first crossing then reduces the LIF signal at the second and produces a narrow dip in the line profile at line center. In Fig. 2 this dip is clearly developed for the ^{136}Xe, ^{132}Xe and ^{128}Xe components and has a FWHM of only 7 MHz. This suggests the present technique as a useful means of greatly improving the resolution. In fact, it should be superior to the classical saturation spectroscopy whose resolution is mainly limited by the residual Doppler width due to the finite crossing angle between pump and probe beam [1]. Here the two laser beams can be made exactly antiparallel.

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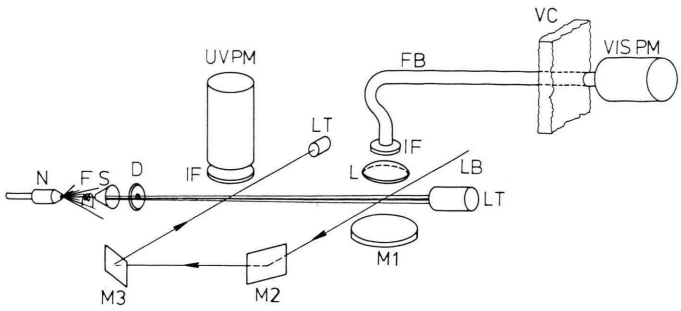

Fig. 1. Schematic of apparatus. N nozzle, F filament, S skimmer, D diaphragm, LT light traps, LB laser beam, M1 concave mirror, M2, M3 plane mirrors, L lens, IF interference filters, FB fibre bundle, VC vacuum chamber wall, VIS PM/UV PM photomultipliers.

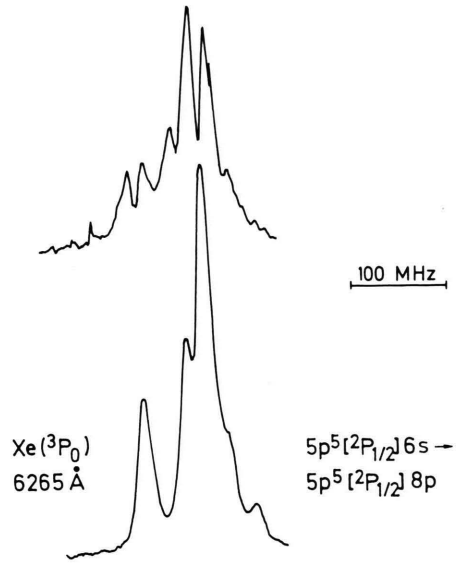

Fig. 2. Bottom: LIF signal from UV PM. The optical transition from Xe $^3P_0$ is indicated. Top: LIF signal from VIS PM, showing saturation dips.

A further advantage would result from using a divergent laser beam. With a well collimated laser beam, the saturation signal derives only from atoms on the axis of the atomic beam, equivalent to a conventional LIF signal from a very narrow atomic beam [3], with its attendant severe intensity sacrifice. It can be shown, however, that the full cross section of a divergent particle beam can be utilized with divergent countercrossing laser beams. This will widen the saturation dip, but only up to half the width of a conventional LIF signal (i.e. from a collimated laser plus divergent particle beam). Thus it should be possible to use the scheme suggested here to achieve an optimum compromise between intensity and resolution. Unfortunately we were unable to verify this experimentally, because the laser system was destroyed by fire immediately after the present results were obtained. – After submission of this note we learned of ref. [4], where a similar technique was used, involving lock-in detection.

Acknowledgement

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[1] W. Demtröder, Laser Spectroscopy, Springer, Berlin 1982.
[2] For preliminary results, see H. Geisen, T. Krümpelmann, D. Neuschäfer, and Ch. Ottinger, 18th EGAS, Marburg 1986, Europhysics Conference Abstracts 10 F, p. 39–41. The experimental method proposed here was first presented at this conference.
[3] H. Geisen, T. Krümpelmann, D. Neuschäfer, and Ch. Ottinger, submitted to J. Phys. B.
[4] T. Kröckertskothen, H. Knöckel, and E. Tiemann, Chem. Phys. **103**, 335 (1986).

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