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Frequency Stabilization of Argon Lasers at 582.49 THz Using Saturated Absorption in $^{127}I_2$

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Abstract

Preliminary results are given for the Allan variance of argon lasers stabilized on iodine saturation peaks in external absorption cells at 5145 Å (10^{-10} m). A prestabilization on Fabry-Perot transmission fringes was found necessary for the commercial lasers used in this work. A flicker floor at 8×10^{-13} is reached between 0.1 and 1 second. Line pressure broadening and shift are studied by the frequency offset locking technique. This new optical frequency standard appears to offer many advantages with respect to accuracy because of the high molecular weight of I_2 and of the possibility of using narrow three level resonances free of recoil structure.

1. Introduction

The development of frequency stabilized lasers in the visible appears as a reasonable goal in view of the widely shared philosophy that an optical length standard should be visible to the human eye and also because of the numerous efforts to extend the frequency measurements in that part of the spectrum. The first problem that we meet in this research is to make the best possible choice of the couple absorber–laser source. Let us recall the basic requirements for this double choice. The chosen molecules should present a strong absorption together with a long lifetime of the excited state: this implies a large Boltzmann factor and a small partition function to ensure a large population of the lower state. Also the absorber should have a high molecular weight to reduce the influence of the transit-time broadening, the curvature-induced shift [1] and the second order Doppler shift. The only problem due to high mass is a very small recoil splitting. The recoil doublet will therefore usually be unresolved and if the two levels have different lifetimes this will give rise to an apparent pressure shift. Fortunately there is a very elegant way out of this difficulty offered by the three level resonances (Doppler-generated level crossings or cross-over saturation peaks) which are free of recoil structure [2, 3]. Since the last requirement is to avoid differential saturation within an unresolved hyperfine structure, nobody will be surprised if we conclude that iodine is an ideal candidate. Next, let us examine what the conditions are for the light source: the laser should have a small amplitude and frequency noise when free running. It should also provide a good spatial quality beam, powerful enough to saturate the molecular absorption outside the laser cavity and to bring good signal-to-noise ratios. These requirements are presently met with an increasing level of technological difficulties by neutral atom lasers, ionized atom lasers and dye lasers. Finally it is, of course, essential for the laser emission to match the chosen molecular absorption line. This condition is easily fulfilled with a broadly tunable laser such as the dye laser, but on the other hand it is then much harder to be quite sure that one has picked up the right line in a complex spectrum such as that of iodine; in that respect a higher guarantee is provided by the naturally narrow and reproducible range of emission of gas lasers. The HeNe laser at 6328 Å (10^{-10} m) would have been perfect if the matched iodine transition had not involved a highly excited lower level ($v = 5$) with a small population and an upper level with a rather short radiative lifetime. A considerably better situation is that of the argon laser at 5145 Å which matches the hyperfine structure of two transitions P(13) and R(15) between the ($v = 0, X^1\Sigma$) and ($v = 43, B^3\Pi$) states of $^{127}I_2$, for which the natural width is of the order of 60 kHz. This attractive coincidence has been thoroughly explored by Ezekiel and collaborators [4], using linear spectroscopy in a molecular beam of iodine. These results are a challenge to nonlinear spectroscopy and promising preliminary results have been obtained using saturated absorption [5] and fluorescence [6] at Stanford and using saturated absorption and dispersion in Paris [7, 8]. Commercial lasers can be used for which the short term stability problem can be handled with the presently available high-speed servo-loop systems. These lasers are powerful enough so that one can use a low pressure iodine cell outside the laser cavity and control the laser beam geometry with expanding optics.

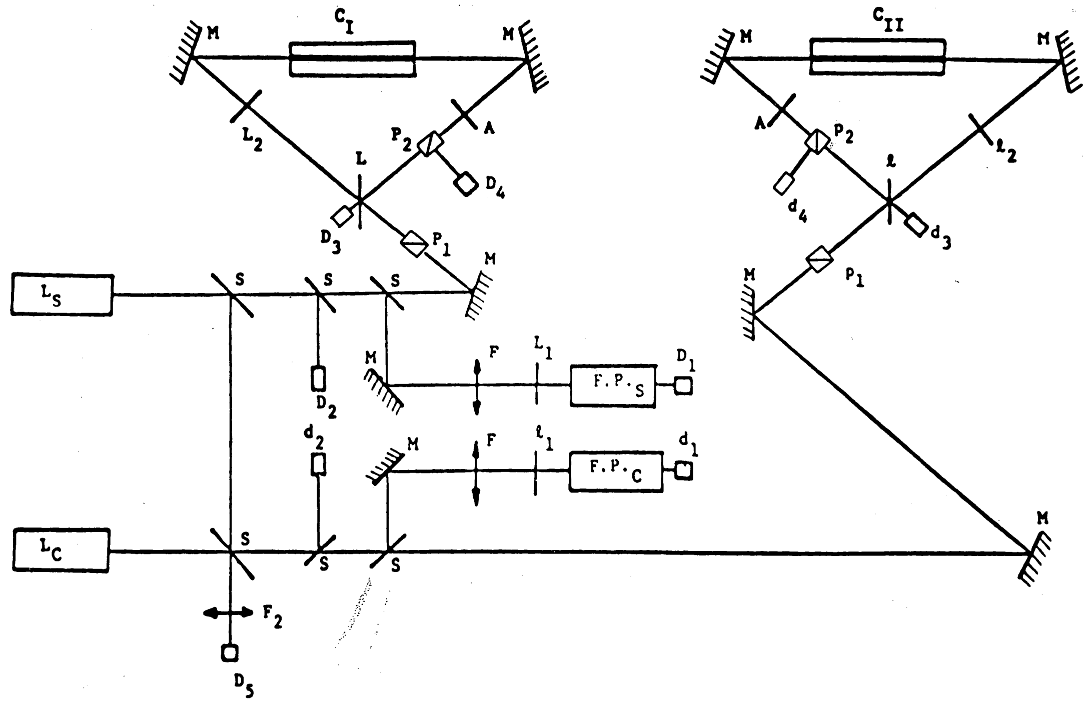

Fig. 1. Schematic diagram of the optical set-up. L_S, L_C: lasers; S, L, l: beam splitters; M: plane mirrors; C_I, C_{II}: iodine cells; l_2 L_2: $\lambda/2$ waveplates; P, p Glan prisms; FP: Fabry-Perot interferometer; l_1 L_1: $\lambda/4$ waveplates; D, d: detectors

2. Description of the Experiment

The schematic diagram of the optical set-up can be found on Figure 1. Two commercial 2-watt argon lasers (from two different companies) illuminate two identical optical circuits. The essential part of these systems is the usual interferometric ring with perpendicular polarizations that can be used to monitor either the saturated absorption or, with a minor modification, the saturated dispersion [7] in the iodine cells C_I, C_{II}. These cells are about 40 cm long and the iodine pressure is set to a few millitorrs* by the temperature of a cold finger. No special effort has yet been made to enlarge the laser beams but this should certainly be done in the future. Glan prisms are used to steer the probe beams towards the detectors and to prevent the light from returning into the lasers. A further improvement in the optical isolation could be obtained with TGG Faraday rotators. Single mode operation is achieved by using internal Fabry-Perot etalons in the commercial lasers. Because of the lack of mechanical rigidity of the structure of these lasers, the cooling water flow induces a frequency jitter between 20 and 50 MHz at a few hundred hertz. A good improvement has been obtained by removing the cavity mirrors onto external mounts but the most useful concept seems presently to be that of prestabilization of the laser frequency on the side of the transmission fringe of a spherical Fabry-Perot interferometer [9]. Until now we have used rather low-finesse (50–100) and short (5–10) cm confocal cavities with invar spacers, essentially because of feedback problems. To avoid converting amplitude noise into frequency noise, it is essential to use the difference signal between the signals corresponding respectively to the transmitted beam and to a reference beam (at least for attack times longer than the storage time of the cavity [10]). Presently the correction signal is dispatched through a dividing cross-over bridge to a fast PZT ceramic for high speed corrections (up to 70 kHz) and to a slow one for slow-drift corrections. In this way the short-term jitter has been reduced to a value of the order of a few hundred kilohertz. The beat frequency of the two lasers is monitored on the last detector and sent to a Hewlett-Packard computing counter.

3. Line Shape Considerations and Frequency Stability Results

By simultaneously scanning and dithering the length of the Fabry-Perot which drives one of the lasers, one can record a derivative spectrum of iodine such as that shown on Figure 2. This spectrum exhibits not only the 42 expected hyperfine diagonal components $\Delta F = \Delta J$ but also a number of Doppler generated level crossings between these and the weaker $\Delta F = 0$ transitions. Precise measurements of the splittings between these various hyperfine resonances are being made to complete the picture of the hyperfine energy levels given by linear spectroscopy [4]. Line shape studies have also required our attention and have been started thanks to a well-known frequency offset locking technique. One of the two lasers (through its driving Fabry-Perot) is locked to one of the iodine line centers and the second laser is scanned over some other adjacent line by monitoring the beat frequency between the two lasers. (This could equivalently be done by creating a side-band at a tunable frequency distance of a

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* 1 millitorr = 1 μmHg = 0.13 pascal (Pa).

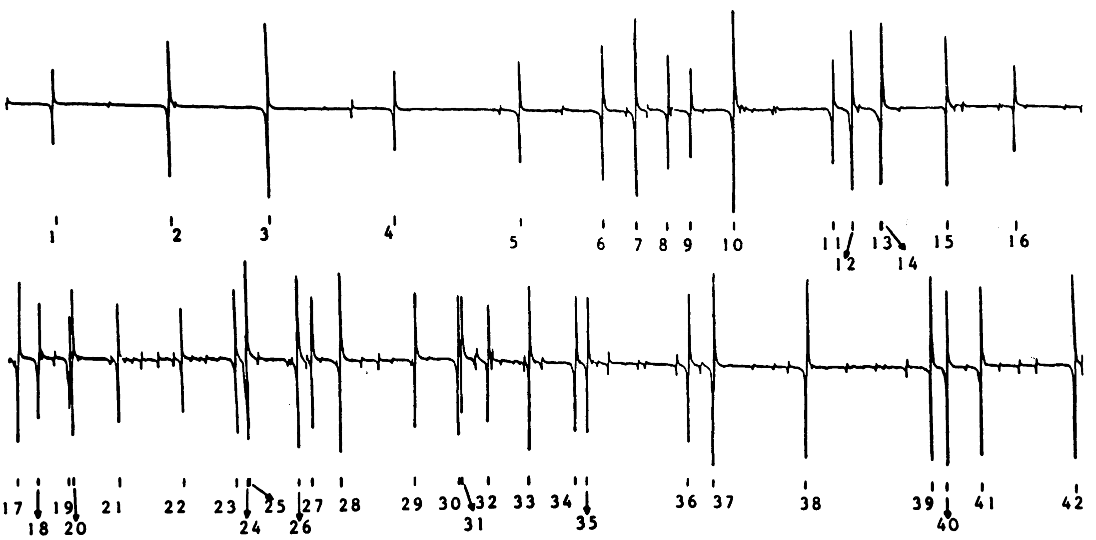

Fig. 2. Derivative spectrum of the iodine at 5145 Å showing the 42 main hyperfine diagonal components and the Doppler-generated level crossings. The frequency scale is such that there are 567353 kHz between line 1 and line 16

single laser carrier with a fast modulator.) A very preliminary study of the linewidth was performed, and the corresponding data are to be seen on Figure 3b. The extrapolated full peak-to-peak width at zero pressure (~500 kHz) is still much larger than the natural linewidth (~60 kHz) essentially because of the high saturation parameter and modulation amplitude used in these early experiments. Among other factors contributing to the linewidth the main ones are the residual laser frequency jitter and the transit-time broadening.

The next class of results of metrological interest is the Allan variance of the beat frequency. Figure 4 shows three σ versus τ plots for:

- A – Free-running lasers
- B – Lasers slaved to free Fabry-Perot cavities
- C – Lasers slaved to iodine peaks with prestabilization on Fabry-Perot cavities.

In case C it should be noted that the 10^{-12} level is reached very early (100 ms) essentially thanks to the very good signal-to-noise ratio. The flicker floor at 8×10^{-13} is also reached early and can be attributed to a number of uncorrected drift mechanisms of various but known origins (electronic offsets, iodine pressure changes, feedback towards the laser, etc.) against which a lot of progress should be possible. A preliminary study of pressure shifts (Fig. 3a) indicates shifts less than or equal to 1 MHz/mmHg. Besides instrumental shifts (baseline tilt, modulation shifts, etc.) and pure pressure shifts (due to collision physics only) line center shifts can be due to beam curvature, to recoil and to the second order Doppler effect. Curvature-induced shifts can be reduced to a negligible value by expansion of the beams and more care about the symmetry of the experiment. The recoil splitting (5.9 kHz) brings an apparent pressure shift in the low pressure region where collisions and spontaneous emission compete for the relaxation of the upper state. If the collisional decay constants of both levels are different, the differential saturation of the two recoil peaks can also give rise to a power dependent shift even at high pressures.

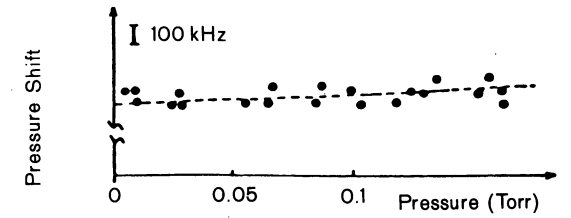

Fig. 3a. Pressure shift versus pressure for line # 23

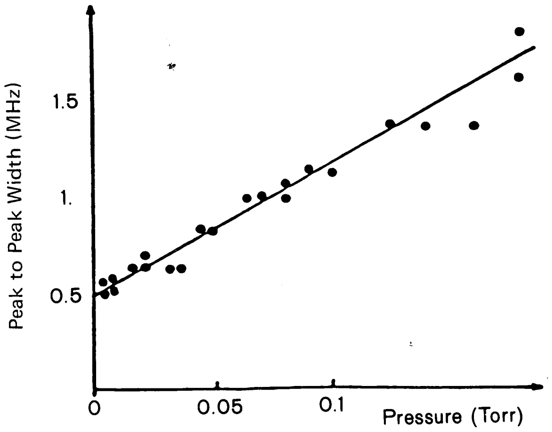

Fig. 3b. Peak-to-peak width of the lock-in signal versus pressure for line #23 (uncorrected for finite modulation and intensity)

A very good way out of this problem will be the use of the Doppler-generated level crossings. These resonances have only one recoil component blue or red shifted by a constant quantity according to whether they correspond to a lower or upper common level. As can be seen in Figure 2 these peaks are intense enough and well enough resolved to be used instead of the main peaks. The second order Doppler shift is only of the order of 3.6×10^{-16}/K, and it should be easy enough in this case to escape the transverse velocity selection effect [1] due to the finite transit time by a proper expansion of the beam.

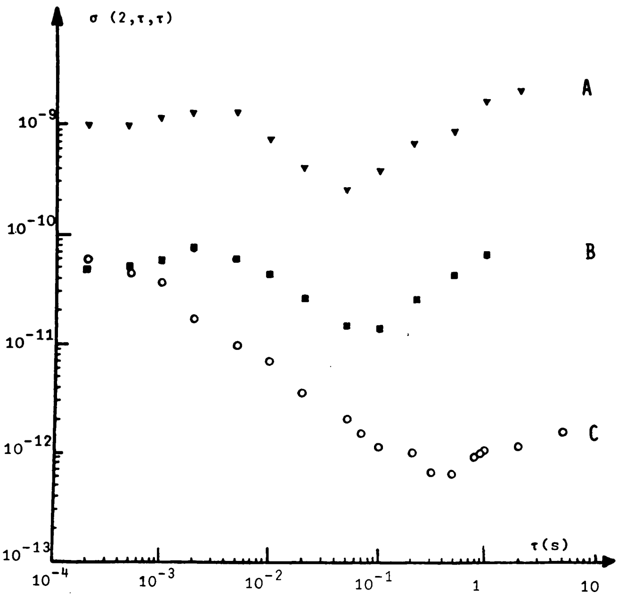

Fig. 4. Allan variance plots for the free-running lasers (curve A), the lasers stabilized on Fabry-Perot interferometers (Curve B), the lasers stabilized on iodine peaks with prestabilization on Fabry-Perot interferometers (curve C)

4. Conclusion

The argon/iodine couple appears to be one of the most promising systems for an optical frequency standard in the visible. All the conditions required to obtain a good accuracy are fulfilled: well-resolved hyperfine structure, very small and calculable second-order Doppler shift, small curvature-induced shifts, no recoil structure with the three level resonances, possibility to use very low pressure to reduce the pressure broadening and shift. Furthermore, the conditions to achieve a good stability in a short time are also there: strong absorption, possibility to use a long absorption cell outside the laser cavity to obtain an exceptionally good signal-to-noise ratio. It is possible to make considerable progress in getting a better short term stability of the argon laser, essentially by going to a better design of the mechanical structure for the laser and to super-fast frequency corrections with an electro-optic crystal in the cavity. It is not yet clear what the limitations due to the electric discharge will turn out to be. Some progress is also possible in the Fabry-Perot interferometer technology: higher finesse, greater length and smaller thermal drifts. To lower the flicker floor it appears necessary: 1) to use a temperature control of the cold finger defining the I_2 pressure, 2) to use an expanded beam, 3) to use longer cells with lower pressure, 4) to lock the lasers on Doppler-generated level crossing, 5) to improve the optical isolation, and 6) to reduce electronic offsets. In these ideal conditions the linewidth will be dominated by the natural broadening only. If narrower lines are then desired it should be possible to use the 5017 Å coincidence of the Ar^+ laser with the R(26) 62–0 transition of I_2 for which the upper state lifetime is between 15 and 30 μs (11). At last the power available from an argon laser may be a real advantage for a number of applications. For example it may be of interest to use a good optical frequency standard in many experiments involving non-linear optics. In this respect let us point out that the ease of doubling the argon laser frequency turns it into a convenient near U.V. standard. A powerful source capable of delivering several stable close frequencies should also be very useful in long-distance interferometry. Besides this good candidate, we certainly should not rule out other cw ion lasers such as the Krypton laser, which has overlaps with other good iodine transitions: R(77) 40–0 at 5208 Å (10^{-10} m), P(10) and R(13) 32–0 at 5309 Å, P(36) 17–0 and P(96) 18–0 at 5682 Å. Also, other heavy molecules like Br_2 or Se_2 should be carefully considered.

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Note added in proof: Recently the short-term stability of the Fabry-Perot slaved lasers has been improved by a factor ten yielding peak-to-peak linewidths of 170 kHz at one millitorr iodine pressure and the variance around one second, for the lasers locked on iodine peaks, has been lowered to 10^{-13}.