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Published in:
Optics Letters

DOI:
10.1364/OL.10.000597

1985

Laser spectroscopy using beam-overlap modulation

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Received September 12, 1985; accepted October 2, 1985

A new Doppler-free laser spectroscopy method is demonstrated that employs modulation of the position of a laser beam rather than the commonly used intensity or polarization modulation. The technique is applicable in saturated absorption as well as fluorescence measurements, as is illustrated in experiments on sodium and iodine lines. A particular feature of the method is that Doppler- and background-free fluorescence spectra can be recorded without using intermodulation techniques.

A large number of Doppler-free laser spectroscopy techniques have been developed that are based on using two laser beams propagating in opposite directions. These beams combine to produce narrow-band optical saturation of selected velocity groups. In the saturation absorption technique[1,2] the hole burned by a strong saturating laser beam is sensed at the line center by a weak counterpropagating beam derived from the same narrow-band laser. Since both beams then interact with the same atoms, chopping the probe beam modulates the absorption and hence modulates the intensity of the probe beam. This occurs only for those atoms that have zero velocity along the beam directions. Thus the signals that are free from the Doppler-broadened background can be recorded by using lock-in techniques. In the present work, the saturating beam, instead of being chopped, is displaced transversely by reflection from a vibrating mirror. The two beams then alternately overlap and can saturate the same population of atoms or travel through different regions of the gas and act on different atoms.

Spatial-overlap modulation is also applicable to many of the other techniques for Doppler-free laser spectroscopy. The limitations of the saturated absorption technique to rather strongly absorbing media are overcome in the saturated fluorescence method, in which two counterpropagating beams of essentially equal intensities are used. However, since all the fluorescence is monitored, the Doppler-broadened background cannot be removed by modulating the intensity of only one beam. Instead, the two beams are modulated at different frequencies f_1 and f_2, and the *intermodulated* signal at $|f_1 + f_2|$ or $|f_1 - f_2|$ is detected[3]. Such techniques have later also been used for intermodulated dc and rf optogalvanic[4,5] spectroscopy. Sensitivity can also be enhanced by using polarization effects for Doppler-free measurements. In the polarization spectroscopy scheme[6] optical anisotropy induced by a strong polarized beam opens up a crossed-polarizer arrangement at the line center.

In this Letter we describe a new class of Doppler-free experiment in which the degree of overlap between the beams, rather than intensity or polarization, is modulated. One of the beams is reflected from a high-frequency vibrating mirror to provide overlap modulation for measurements on the sodium D_1 and D_2 lines as well as on some molecular iodine lines. A specially valuable feature is that intermodulation techniques are not needed for eliminating the Doppler-broadened background that otherwise occurs in fluorescence or optogalvanic monitoring. The experimental complication of intermodulation can be avoided since both beams are present and individually interacting with the gas at all times, whereas beam interaction occurs only for spatially overlapping beams and only at the line center. Further, the technique allows the full laser power to be used in both beams.

The present experiments were performed on cells containing sodium or iodine vapor. A Coherent Radiation Model 599-21 single-mode dye laser operating on Rhodamine 6G was used. Experimental arrangements are shown in Figs. 1(a) and 1(b). In Fig. 1(a) the setup for fluorescence monitoring is presented. A fraction of the dye-laser radiation was split off for monitoring the single-mode operation of the dye laser

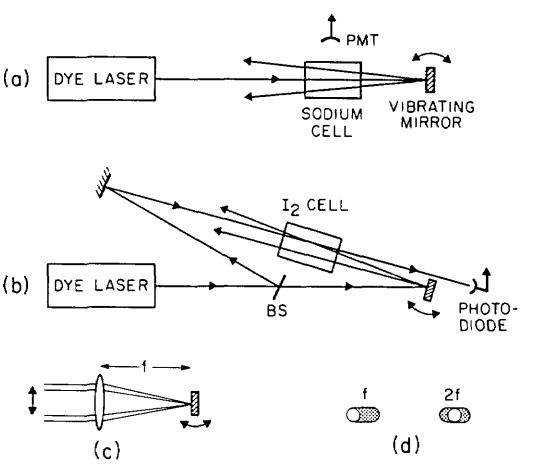

Fig. 1. Experimental arrangements for Doppler-free spectroscopy using spatial beam modulation. (a) Fluorescence detection, (b) absorption detection, (c) parallel translation of scanning beam produced by insertion of lens at distance f from vibrating mirror. (d) Signal is modulated at first or second harmonic of mirror vibration frequency according to position of stationary beam within the scan.

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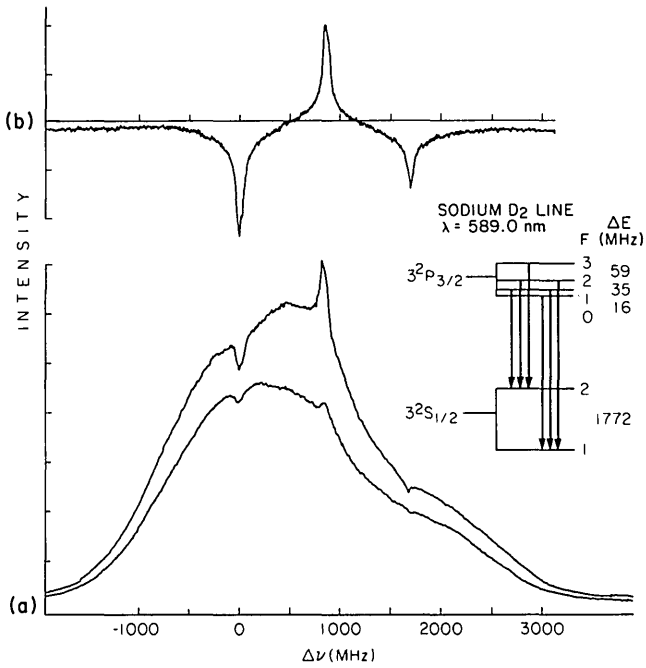

Fig. 2. Laser scans of the sodium $3\,^2S_{1/2}$–$3\,^2P_{3/2}$ line with fluorescence detection. A 3-cm-long cell at a temperature of 90°C was used. (a) Direct-current photomultiplier-tube recording showing the Doppler-broadened line profile together with saturation features that are due to counterpropagating cell-window reflexes and enhancement of these features by inserting a fixed retroreflecting mirror. For both curves a laser-beam power of 6 mW was used. (b) Background-free signals obtained by the vibrating-mirror technique. The lock-in time constant was 0.3 sec, and a laser power of 1 mW was used.

with a scanning Fabry–Perot interferometer and for wavelength measurements using an air-track wavemeter.[7] The main part of the dye-laser beam is transmitted through the cell and is incident upon the 4-mm-diameter aluminized mirror of a General Scanning Inc. Model S10418 resonantly vibrating scanner operating at 18 kHz. By placing the mirror at the focus of a lens [Fig. 1(c)] a parallel translation of the reflected beam in and out from the primary beam could be accomplished. A photomultiplier tube was used at close range from the cell without any light-collection optics. By using a small translation of the beam, just enough to ensure that the beams are brought in and out of spatial overlap, the light-collection efficiency of the photomultiplier was kept essentially constant for both beams at the same time as efficient lock-in detection was maintained (symmetric modulation). Depending on the position of the fixed beam in the swept beam area [Fig. 1(d)] lock-in signals at f or at $2f$ (f is the vibration frequency) are expected. That is, if the beams just overlap at one edge of the region swept out during vibration, a signal will be obtained at the scanning frequency. If the beams overlap in the middle of the scan, the moving beam will cross the fixed beam twice during the vibration, and the signal will appear at frequency $2f$. For optogalvanic detection the same optical scheme would have been used.

In Fig. 1(b) the setup used for saturated absorption measurements is shown. Here only a small fraction of the laser output is used for a fixed probing beam that is influenced by the swept strong saturation beam. This configuration could also be used for overlap-modulation polarization spectroscopy.

We will first describe our fluorescence experiments performed on the sodium $3\,^2S_{1/2}$–$3\,^2P_{3/2}$ (λ = 589.0-nm) and $3\,^2S_{1/2}$–$3\,^2P_{1/2}$ (λ = 589.6-nm) transitions. The starting point for the present work was the observation that Doppler-free features can be observed superimposed upon the normal Doppler-broadened fluorescence line profile in experiments in which the laser beam is just passed through a heated sodium cell with normal-incidence windows. Such an experimental trace is shown in Fig. 2(a) (lower trace). The Doppler-free features are due to the 4 + 4% backreflection of the beam into itself from the rear cell window. Two saturation dips, separated by the ground-state hyperfine-structure (hfs) interval 1772 MHz, are observed as well as the expected cross-over signal in the middle of the interval.[8] The signals, which are due to overlapping transitions to excited-state hfs levels, are enhanced by reflecting the whole beam back onto itself with a fixed mirror as illustrated in Fig. 2(a) (upper trace). Some care had to be exercised to avoid feedback into the laser cavity.

In order to lift off the Doppler-free signals from the background the technique with a vibrating mirror was used as illustrated in Fig. 1(a). A lock-in recording is shown in Fig. 2(b). With the fast beam modulation no perturbation of the laser caused by feedback effects was observed. Because of the slight movement of the fluorescence streak following the swept beam, a slight Doppler-broadened signal can be picked up if the detected light intensity varies because of solid angle variations, scattering, etc. Similarly, a wavelength-independent background that is due to slightly modulated stray light could occur if the detection follows at the laser wavelength. However, these effects were always small and could essentially be eliminated by geometri-

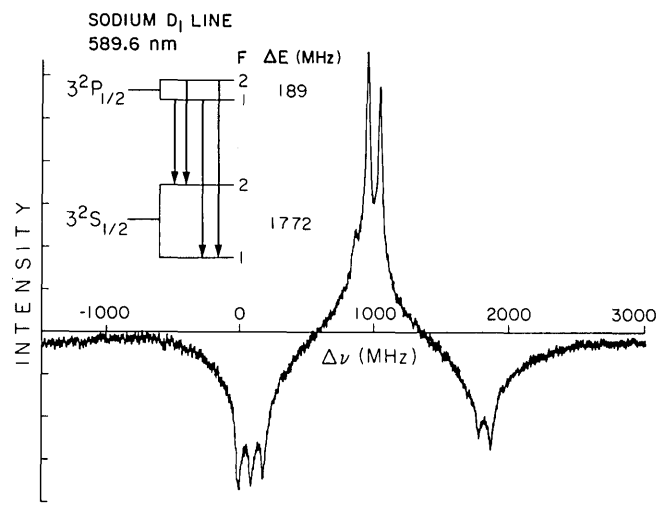

Fig. 3. Vibrating-mirror fluorescence recording of the $3\,^2S_{1/2}$–$3\,^2P_{1/2}$ transition in ^{23}Na.

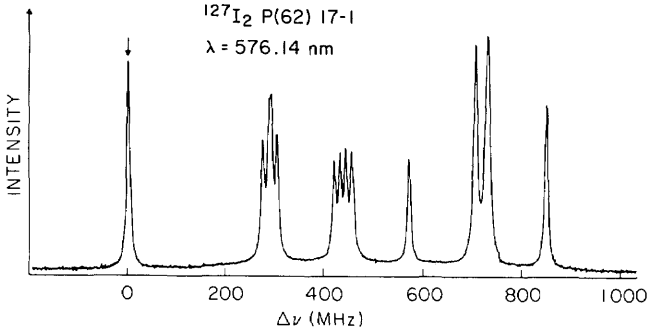

Fig. 4. Doppler-free spectrum of the rovibronic I_2 line 17-1 $P(62)$ at 576.14 nm with a hfs component (λ_{vac} = 576 294 760.27 fm) adopted as a secondary wavelength standard[11] indicated. The curve was recorded using saturated absorption spectroscopy using spatial modulation of a 22-mW saturating beam. A 10.3-cm-long I_2 cell at room temperature was used. The lock-in time constant was 0.3 sec.

cal adjustments. In principle, all three signals could have different lock-in phases.

The excited-state lifetime for the $3p$ states is 16 nsec, corresponding to a natural radiation width of about 10 MHz. With the known excited-state hfs intervals, given in Fig. 2, resolved components should be attainable. While the linewidth observed in our experiments varies with the laser power (saturation broadening), it was not possible to resolve the signals even at low powers. Further, by reducing the cell vapor pressure the signals could not be substantially narrowed. A pedestal with a half-width of about 600 MHz is also always observed below the sharp signals. These observations are evidence for the presence of residual gas in the old sodium cell used. Since the purpose of this Letter is to demonstrate a new measurement technique rather than to investigate the line shapes in saturation spectroscopy, we did not study the line broadening any further.

An experimental recording of the $3\ ^2S_{1/2}$–$3\ ^2P_{1/2}$ transition is shown in Fig. 3. With an excited-state hfs splitting of 189 MHz, resolved components are observed, including cross-over signals that are due to common ground-state or excited-state levels in the two-beam interaction.[8] A resolved linewidth of 50 MHz was observed.

As an example of the application of the spatially modulated beam technique applied in saturated absorption measurements, a recording of the 17-1 $P(62)$ line of I_2 (λ = 576.14 nm) (Ref. 9) is shown in Fig. 4. The setup illustrated in Fig. 1(b) was used. With a nuclear spin of 5/2 for the 100% abundant ^{127}I isotope, individual rovibronic lines of I_2, originating in the ground state of even parity, exhibit 15 or 21 hyperfine components, depending on whether the ground-state rotational quantum number is even or odd. The analysis of iodine hfs in terms of electric-quadrupole and magnetic-dipole interactions is discussed, e.g., in Ref. 10. We find an experimental linewidth of 8 MHz for the components in Fig. 4. In the figure a hfs component that is adopted as a secondary wavelength standard in connection with the recent redefinition of the speed of light[11] is indicated.

In this Letter a new technique for obtaining Doppler-free spectra has been demonstrated. By using spatial beam modulation rather than an intensity modulation, the Doppler-broadened background can be suppressed in experiments using fluorescence or optogalvanic signal detection without using intermodulation techniques. This permits system simplification and full utilization of the available laser power. Since one of the counterpropagating beams is spatially swept, it is important that the penetrated gaseous medium be homogeneous and that the detection efficiency be kept constant despite the displacement of the beam. Otherwise a Doppler-broadened signal will be detected.

For low gas pressures, spatial modulation could be used in a way such that the beams do not actually overlap but pass each other with a small separation. Cross saturation by the two beams would occur because of diffusion of atoms from one beam to the other, and that would be greatest when the beams are closest and less when they are apart.

The ease with which narrow-band saturation effect can be observed for lines with high oscillator strength by using just the cell-window backreflection was noted in connection with the present experiments. Here high-resolution laser spectroscopy has been reduced to its simplest form with utilization of neither mirrors nor choppers or other moving parts. Such signals can be used for marking line component center positions in studies of line shapes in absorbing gaseous media.

This research was supported by the National Science Foundation under grant PHY-83-08271.

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References

1. T. W. Hänsch, M. D. Levenson, and A. L. Schawlow, Phys. Rev. Lett. **27**, 707 (1971).
2. C. Bordé, C. R. Acad. Sci. **271**, 371 (1970).
3. M. S. Sorem and A. L. Schawlow, Opt. Commun. **5**, 148 (1972).
4. J. E. Lawler, A. I. Ferguson, J. E. M. Goldsmith, D. J. Jackson, and A. L. Schawlow, Phys. Rev. Lett. **42**, 1046 (1979).
5. D. R. Lyons, A. L. Schawlow, and G.-Y. Yan, Opt. Commun. **38**, 35 (1981).
6. C. Wieman and T. W. Hänsch, Phys. Rev. Lett. **36**, 1170 (1976).
7. F. V. Kowalski, R. T. Hawkins, and A. L. Schawlow, J. Opt. Soc. Am. **66**, 965 (1976).
8. T. W. Hänsch and P. Toschek, Z. Phys. **236**, 213 (1970).
9. K. M. Baird, K. M. Evenson, G. R. Hanes, D. A. Jennings, and F. R. Petersen, Opt. Lett. **4**, 263 (1979).
10. M. D. Levenson and A. L. Schawlow, Phys. Rev. A **6**, 10 (1972).
11. "Documents concerning the new definition of the metre," Metrologia **19**, 163 (1984).