Novel Spectroscopy

Phase Modulation Laser Spectroscopy

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An elegant phase modulation method for detecting optical atomic resonances, particularly in the nonlinear regime, has been devised recently [1–3] and applied to phase locking a laser to a reference cavity with unsurpassed precision [2]. Laser phase locking, which resembles earlier methods at longer wavelengths, has produced a laser linewidth as narrow as 100 Hz in the case of a dye laser [2] and is also being used in an attempt to detect gravity waves with optical interferometers [4].

The basic spectroscopic arrangement considered here consists of a combination of optical pump and probe fields which appear either simultaneously or in sequence. The pump is typically a single frequency cw laser field that prepares an atomic sample, for example, by burning a hole within its homogeneous lineshape. The probe, which can be derived from the pump or another laser source, is phase modulated and therefore contains a Bessel function distribution of sidebands that appear symmetrically in pairs about the unmodulated laser frequency.

In the absence of attenuation, each pair of sidebands generates with the central frequency component at a photodetector a pair of heterodyne beat signals of opposite phase which just cancel. This balance is upset and a nonvanishing beat signal remains when the probe frequency is swept, bringing a sideband into resonance with the prepared hole. The background signal and its noise are therefore eliminated automatically. Moreover, since the beat frequency can be made arbitrarily high (radio or microwave), the desired beat signal can be detected in a spectral region where the residual noise spectrum is falling off. The resulting high sensitivity, which will ultimately be limited by shot noise, promises new levels of precision in laser spectroscopy.

In this paper, we investigate the theory [5] of phase modulation laser spectroscopy, extending the work of Bjorklund [1] and Hall et al. [3]. Our analysis treats the nonlinear optical response of a two-level atomic quantum system subject to copropagating or counterpropagating laser beams where at least one of the fields is phase modulated. The counterpropagating beam case resembles the Lamb-dip effect but is more complex due to the multimode character of the phase modulated probe. Solutions equivalent to a rate equation result are developed and the effect of *coherence* corrections are examined, revealing in certain cases new resonances. The delayed pump-probe measurement frequently encountered in solid state optical hole burning studies is analyzed where it is found that two and three-level quantum systems behave differently. The (linear) response of a Fabry-Perot cavity to a phase modulated light wave is also considered because of its relevance to phase locking a laser.

Finally, phase modulation spectroscopy will be illustrated in nonlinear optical spectra of low temperature zero-phonon transitions of dilute impurity ion crystals, such as Pr^{3+} in LaF_3, where kilohertz homogeneous linewidths appear using a highly stable phase-locked ring dye laser.

1. G.C.Bjorklund: Opt. Lett. **5**, 15 (1980)
2. R.W.P.Drever, J.L.Hall, F.V.Kowalski, J.Hough, G.M.Ford, A.Munley: Unpublished
3. J.L.Hall, L.Hollberg, T.Bear, H.G.Robinson: Appl. Phys. Lett. **39**, 680 (1981)
4. R.W.P.Drever et al.: In *Laser Spectroscopy V*, ed. by A.R.W.McKellar, T.Oka, and B.P.Stoicheff (Springer, Berlin, Heidelberg, New York 1981) p. 33
5. A.Schenzle, R.G.DeVoe, R.G.Brewer: Phys. Rev. (1982)

Subnatural Linewidth Spectroscopy by Phase Switching of the Optical Field

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We give a detailed discussion on the characteristics of the subnatural linewidth spectroscopy by phase switching based on our recent result on the laser induced fluorescence experiment of Na *D* lines [1]. This type of experiments relies on the detection of the coherent part of the response which lives longer than the average decaying time. To extract such information, the excitation field is modulated and the measurement is done some time later. Suppose the phase of the excitation field is switched by ϕ at time $t=0$, and the fluorescence intensity $I(t,\phi)$ is measured at time t. Then, the difference $I(t,\phi)+I(t,-\phi)-2I(t,0)$ contains the part which is driven coherently during this period. This expression is in the lowest order perturbation approximation

$$2\frac{\mu^2}{\hbar^2}|E|^2(\cos\phi-1)$$
$$\cdot\mathrm{Re}\left\{(e^{(i\Delta\omega-\gamma_{12})t}-e^{-\gamma_1 t})\int_0^\infty NP(t+\tau)\hbar\omega\gamma_1\right.$$
$$\left.\cdot\frac{e^{(i\Delta\omega-\gamma_{12})\tau}-e^{-\gamma_2\tau}}{(-i\Delta\omega+\gamma_{12}-\gamma_1)(-i\Delta\omega+\gamma_{12}-\gamma_2)}d\tau\right\},$$

where N is the number of atoms entering the interaction region in unit time, P the probability to stay in this region for $t+\tau$, $\Delta\omega$ the frequency detuning and γ's are relaxation times. The coherent part

appears in the factor $\exp\{(i\Delta\omega-\gamma_{12})t\}-\exp(-\gamma_1 t)$, which oscillates with Rabi frequency $\Delta\omega$. The integral over τ usually produces the envelope line shape of the width γ. For $\gamma_2=0$, as in the Na D lines, the line shape is approximately given by

$$\frac{\gamma_{12}^2}{\Delta\omega^2+\gamma_{12}^2}(\cos\Delta\omega t e^{-\gamma_1 t/2}-e^{-\gamma_1 t}).$$

It has a central peak of the width approximately $1/t$. The oscillation in the wing can be elliminated, if we further take the integral over t with the weightning function $\gamma_1\exp(\gamma_1 t)$. In a fortunate situation of $\gamma_1=\gamma_2=\gamma_{12}$ [2], this weighted average is automatically done through the decay of the lower state probability as $\exp(-\gamma_2 t)$. In this case the line shape is

$$\frac{\gamma_1^2}{\Delta\omega^2+\gamma_{12}^2}\left(\frac{\gamma_1}{\Delta\omega}\sin\Delta\omega t+\cos\Delta\omega t-1\right)e^{-\gamma_1 t}.$$

Our phase switching technique has many advantages over the intensity switching. Since the excitation intensity is constant and the spectral spread by modulation is smaller than γ, the standard natural-width-limited spectrum is retrieved by simply summing the time resolved signal without degrading S/N ratio. The above described operation can be done by recording fluorescence photon signal in a multichannel counter synchronous to the modulation. Since a complete temporal response is recorded by this method, a more complicated manipulation is possible. The same technique can be also applied for the absorption spectroscopy, if the counter is replaced by an appropriate device.

1. F.Shimizu, K.Umezu, H.Takuma: Phys. Rev. Lett. **47**, 825 (1981)
2. P.Meystre, M.O.Scully, H.Walther: Opt. Commun. **33**, 153 (1980)

Raman-Induced Phase Conjugation Spectroscopy

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We have demonstrated a new form of coherent Raman spectroscopy in which three beams F, G, and H (at frequencies ω_L, ω_L, and ν respectively) are mixed to generate a fourth beam E (at frequency ν). The beam geometry is that appropriate for phase-conjugation (with beam H nearly counter to beam G) so that the output optical field amplitude $\mathbf{E}(x,y)$ at the entrance plane is very nearly a constant times $\mathbf{F}^*(x,y)$ there. When $|\nu-\omega_L|$ is near the frequency of a Raman excitation of the medium, there is a resonantly enhanced or altered signal. Therefore, we refer to this form of four-wave mixing as "Raman-induced phase conjugation" or RIPC.

We use a frequency-doubled, Q-switched, Nd:YAG laser to produce the frequency ω_L, and also to pump a Rhodamine 6G dye laser which generates the frequency ν. We obtain spectra with a single shot by operating the dye laser broadband over the range of expected Raman lines, and analyzing the frequency content of the generated beam with a spectrometer. Typical photographic recordings of the spectrometer output are shown in Fig. 1. A 1 cm long sample of benzene is the Raman medium. The F beam is incident at 20 mr to the G beam. The F, G, and H beams are about 1 mm diameter, of 10 ns duration and 1 to 10 mJ energy. The phase conjugate reflectivity is of order 10^{-3} on resonance. In Fig. 1a the beam polarizations were arranged such that only the imaginary part of the third-order susceptibility tensor $\chi^{(3)}$ created observable mixing. In Fig. 1b the beam polarizations were such that the entire spectral range of the dye laser mixed to form an output mediated by the nonresonant real part of $\chi^{(3)}$. Here the

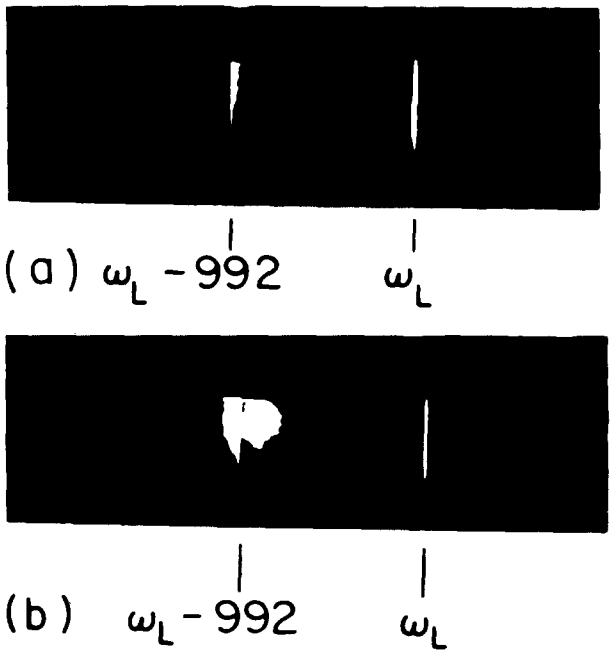

Fig. 1. Raman-induced phase conjugation spectrograms of benzene taken with a single laser pulse

Raman resonance interfered with the nonresonant background. This RIPC technique has the advantage over Raman-induced Kerr effect (RIKE) that four beam polarizations are available, rather than three, thus allowing determination of more elements of the $\chi^{(3)}$ tensor in crystals of low symmetry. The ability to record an entire spectrum with one laser pulse is an advantage over coherent antistokes Raman scattering (CARS). Precisely measured RIPC lineshapes are found to agree with theory.

Recent Progress in Frequency Modulation Spectroscopy

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Recently a frequency modulation (FM) spectroscopy technique has been developed which offers significant improvement in the sensitivity and time resolution achievable by absorption spectroscopy [1]. FM spectroscopy has been utilized to detect Doppler free saturation resonances [2], narrow photochemical holes in absorption lines of solids [3], and to lock lasers to interferometers [4].

We have now applied FM spectroscopy with nanosecond duration pulse pumped dye laser sources of relatively standard designs [5]. In addition to explicitly demonstrating time resolution on a nanosecond scale, these results show that it should be possible to perform FM absorption spectroscopy over the extremely broad spectral region covered by high power pulsed dye lasers and nonlinear crystals. In preliminary experiments with a non-optimized set-up, absorptions as small as 2% were easily detected with non transform limited 5 ns duration, 1 GHz bandwidth dye laser pulses having 30% to pulse fluctuations in energy.

To appreciate fully the advantages of FM spectroscopy with a pulsed laser it is worth reviewing the principle behind FM spectroscopy as illustrated in its cw application. A phase modulator is used to put sidebands on a single frequency laser beam of frequency ν_0. This results in two sidebands at frequencies $\nu_0 \pm \nu_m$, where ν_m is the modulator frequency. The modulated beam is passed through a sample cell and impinges on a square law detector, which gives a beat frequency signal at ν_m only if there is a difference in the absorption of the two sidebands or if the index of refraction for the carrier changes relatively to that of the sidebands. The absorption or dispersion signals arising from these two effects occur 90° out of phase, and it is thus straightforward to distinguish between them. Since with no absorption there is no signal, this approach offers the possibility of observing very small absorption signals with zero background.

In our experiment, a transversely pumped Rh 6G dye laser provided pulsed radiation with 12 GHz bandwidth, 70 μJ pulse energy, and 30% pp fluctuation in energy. The bandwidth was narrowed to 1 GHz by passing the beam through an external scanning Fabry Perot etalon with 30 GHz free spectral range. Sidebands with intensity equal to 10% of the carrier were produced using a $LiTaO_3$ phase modulator driven with several Watts of RF power with ν_m between 1 and 2 GHz. A 20 cm long I_2 cell with a coolable sidearm provided the absorption lines. After passing through the I_2 cell, the beam was attenuated by a factor of 1000 and focused onto a PIN photodiode. The photodiode signal was passed through a 800 MHz high pass filter and homodyne detected using a double balanced mixer. The phase of the local oscillator provided to the mixer was adjusted using a variable length air line. The intermediate frequency output of the mixer was amplified by 40 dB, detected with a boxcar averager with a 5 ns gate, and displayed on a chart recorder.

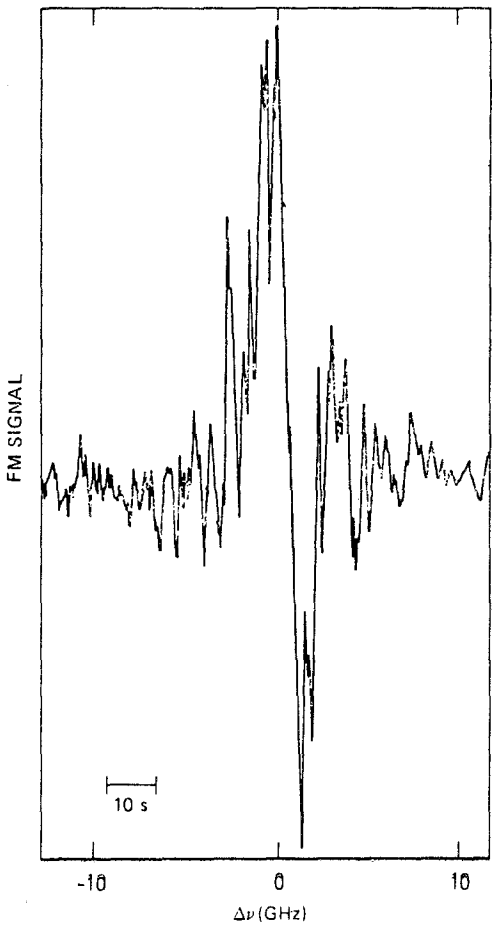

Fig. 1. FM spectroscopy signal resulting from a 2% I_2 absorption with a 1 s time constant. The time scale of the scan is shown

The laser was tuned to be coincident with an I_2 absorption line near 568 nm which provided a 2% deep absorption with the sidearm at 0 °C. The FM spectroscopy signals for $\nu_m = 2$ GHz were detected with a signal to noise of ~10. The FM signal lineshapes were recorded by monitoring the signal as the external etalon was scanned. Figure 1 shows typical results. These lineshapes were found to be in good agreement with cw results and exhibited the expected dependence on the phase of the local oscillator.

In another series of experiments, we have applied FM spectroscopy to detect Doppler free two photon absorption. Ordinarily the change in transmitted intensity due to such a multiquantum transition is far too small to detect. Previous authors have normally detected fluorescence or ionization due to the excited state population. Polarization spectroscopy and suppressed carrier acousto-optic modulation techniques have also proved successful. Frequency modulation with efficient high frequency electro-optic modulators leads to a more convenient and widely applicable technique, which is also capable of measuring the absolute strength of the two photon absorption.

The initial experiments were performed on Rb [8, 7] at a temperature of 187 °C. The strong vertically polarized pump beam ($\lambda = 7781$ Å) from a Coherent 599-21 laser passed through the sample and an electro-optic phase modulator driven at RF frequencies between 50 and 500 MHz, oriented to affect only the

horizontally polarized component. A quarter wave plate and retroreflector converted the pump beam into a horizontally polarized probe, which was then converted into an FM optical spectrum by the modulator, passed through the sample again and directed to a fast photodiode by a Glan-Thompson prism. This geometry maximized signal at the diode and minimized feedback into the laser cavity. When the sum of the frequency of one sideband and the pump frequency equals one of the hyperfine transitions in the $5S_{1/2}$–$5D_{5/2}$ two photon line, an amplitude modulated (AM) signal appeared at the photodiode due to the "unbalancing" of the sideband amplitudes. This absorption phase signal was homodyne detected with a double balanced mixer and a local RF oscillator of the correct phase. In addition, when the sum of the pump and a sideband or carrier frequency approached one of the two photon transitions, two photon dispersion altered the relative phases producing another AM signal at the detector. The dispersion shaped resonances due to this effect could be enhanced with a phase shifted local oscillator. In either case, the spectra are perfectly antisymmetric with respect to the true transition frequency. Figure 2 shows the experimentally obtained absorption and dispersion resonances.

FM spectroscopy permits quantum noise limited detection efficiency even with noisy or pulsed lasers. The optimum FM sensitivity compares with fluorescence detection, if about 0.1% of the absorbed quanta result in a detected fluorescence photon [8]. Such is often the case in the infrared or the VUV. Sum and difference frequency generation processes preserve the FM sideband amplitudes and phases, thus extending this technique beyond the visible. FM detected two photon spectroscopy techniques may prove especially useful for measuring the 12–2S two photon frequency of hydrogen under conditions where fluorescence from the nearby 2P states is difficult to detect.

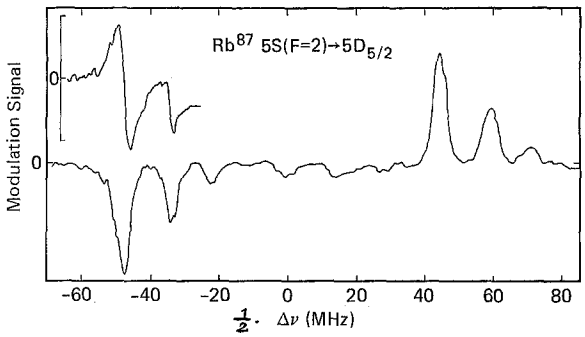

Fig. 2. Absorption and dispersion phase Doppler-free two-photon FM spectroscopy signals ($\gamma_n = 91$ MHz)

1. G.C.Bjorklund: Opt. Lett. **5**, 15 (1980)
G.C.Bjorklund, W.Lenth, M.D.Levenson, C.Ortiz: SPIE **286**, 53 (1981)
2. G.C.Bjorklund, M.D.Levenson: Phys. Rev. A **24**, 166 (1981)
J.L.Hall, L.Hollberg, T.Baer, H.G.Robinson: Appl. Phys. Lett. **39**, 680 (1981)
3. W.Lenth, C.Ortiz, G.C.Bjorklund: Opt. Lett. **6**, 351 (1981)
4. R.W.P.Drever, J.L.Hall, F.v.Kowalski, J.Hough, G.M.Ford, A.Murley: Unpublished
R.G.Devoe, R.G.Brewer: Unpublished
G.A.Sanders, M.G.Prentiss, S.Ezekiel: Opt. Lett. **6**, 569 (1981)
5. T.F.Gallagher, R.Kachru, F.Gounand, G.C.Bjorklund, W.Lenth: Opt. Lett. **7**, 28 (1982)
6. P.F.Liao, G.C.Bjorklund: Phys. Rev. A **36**, 584 (1976)
7. D.Block, M.Ducloy, E.Giacobino: J. Phys. B **14**, L819 (1981)
8. M.D.Levenson, G.L.Eesley: Appl. Phys. **19**, 1–17 (1979)

Frequency Modulation Spectroscopy with a Pulsed Dye Laser

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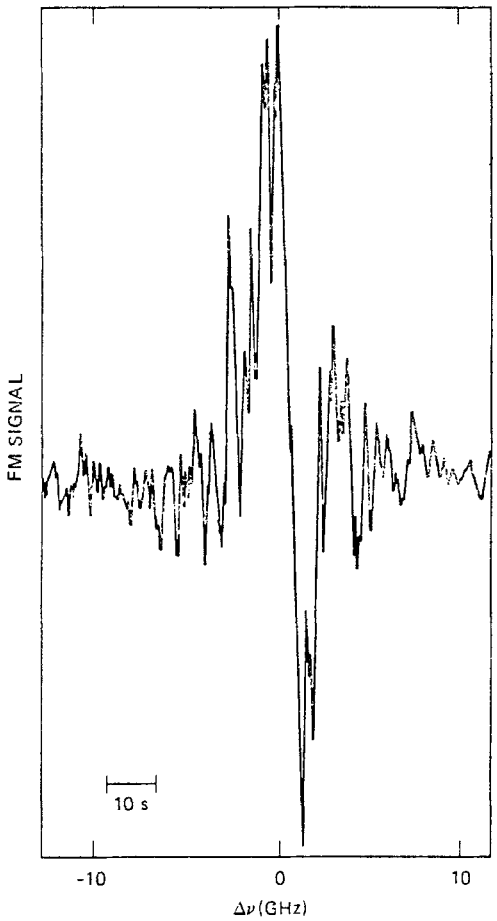
Recently a frequency modulation (FM) spectroscopy technique has been developed which offers significant improvement in the sensitivity and time resolution achievable by absorption spectroscopy [1, 2]. The initial demonstrations of this technique have been limited to single frequency continuous wave (cw) laser sources, although using acousto-optic modulation of the cw beam, μs time resolutions have been demonstrated [2].

In this paper, we report the first use of FM spectroscopy with nanosecond duration pulse pumped dye laser sources of relatively standard design. In addition to explicitly demonstrating time resolution on a nanosecond scale, these results show that it should be possible to perform FM absorption spectroscopy over the extremely broad spectral region covered by high power pulsed dye lasers and nonlinear crystals. In preliminary experiments with a non-optimized set-up, absorptions as small as 2% were easily detected with non transform limited 5 ns duration, 1 GHz bandwidth dye laser pulses having 30% pulse to pulse fluctuations in energy.

To appreciate fully the advantages of FM spectroscopy with a pulsed laser it is worth reviewing the principle behind FM spectroscopy as illustrated in its cw application. A phase modulator is used to put sidebands on a single frequency laser beam of frequency ν_0. This results in two sidebands at frequencies $\nu_0 \pm \nu_m$ where ν_m is the modulator frequency. The modulated beam is passed through a sample cell and impinges on a square law detector, which gives a beat frequency signal at ν_m only if there is a difference in the absorption of the two sidebands or if the index of refraction for the carrier changes relative to that of the sidebands. The absorption or dispersion signals arising from these two effects occur 90° out of phase, and it is thus straightforward to distinguish between them. Since with no absorption there is no signal this approach offers the possibility of observing very small absorption signals because there is zero background.

In our experiment, a transversely pumped Rh 6G dye laser provided pulsed radiation with 12 GHz bandwidth, 70 μJ pulse energy, and 30% pp fluctuation in energy. The bandwidth was narrowed to <1 GHz by passing the beam through an external scanning Fabry Perot etalon with 30 GHz free spectral range. Sidebands with intensity equal to 10% of the carrier were produced using a $LiTaO_3$ phase modulator driven with several watts of RF power with ν_m between 1 and 2 GHz. A 20 cm long I_2



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cell with a coolable sidearm provided the absorption lines. After passing through the I_2 cell, the beam was attenuated by a factor of 1000 and focused onto a PIN photodiode. The photodiode signal was passed through a 800 MHz high pass filter and homodyne detected using a double balanced mixer. The phase of the local oscillator provided to the mixer was adjusted using a variable length air line. The intermediate frequency output of the mixer was amplified by 40 dB, detected with a boxcar average with a 5 ns gate, and displayed on a chart recorder.

The laser was tuned to be coincident with an I_2 absorption line near 568 nm which provided a 2% deep absorption with the sidearm at 0 °C. The FM spectroscopy signals for $\nu_m = 2$ GHz were detected with a signal to noise of ~ 10. The FM signal lineshapes were recorded by monitoring the signal as the external etalon was scanned. Figure 1 shows typical results. These lineshapes were found to be in good agreement with cw results [1] and exhibited the expected dependence on the phase of the local oscillator.

The sensitivity was limited by noise caused by the beating of different frequency components within the bandwidths of the laser. Since ν_m was at most the bandwidth of the laser, this limitation is not surprising. Usable FM signals were obtained with ν_m as low as 1 GHz, although the noise was five times worse than at 1.5 or 2 GHz.

Experiments are currently in progress to enhance the sensitivity by increasing the ratio of ν_m to the laser bandwidth. Laser bandwidths on the order of 50 MHz can readily be produced using ganged, pressure tuned etalons external to the laser cavity. In addition, we plan to explicitly demonstrate the broad spectral coverage this technique offers by performing pulsed FM spectroscopy in the uv using frequency doubled dye lasers.

1. G.C.Bjorklund: Opt. Lett. **5**, 15 (1980)
2. W.Lenth, C.Ortiz, G.C.Bjorklund: Opt. Lett. **6**, 351 (1981)

Stark-Tuned Lamb-Dip and Sub-Doppler Diode Laser Spectroscopy of NH_3

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In a recent paper we described a Stark-tuned infrared double resonance experiment in NH_3 which used a CO laser pump in the 5 μm ν_4 band and a diode laser probe in the 10 μm ν_2 band [1]. Here we extend that work with the addition of a precision Stark cell having a field homogeneity $\Delta E/E \lesssim 3 \times 10^{-4}$. We will also give results of Stark-tuned Lamb-dip measurements on 16 new transitions in the ν_4 region, which were obtained using different CO isotopes and fields up to 20 kV/cm. The precision Stark cell allows us to identify various line broadening mechanisms, which were previously masked by field inhomogeneity, and to see new features in both our IRDR and Lamb-dip spectra.

The experimental setup is shown in Fig. 1, and a portion of one of the Lamb-dip spectra is shown in Fig. 2. This transition was studied by Johns et al. [2] who identified all of the strong resonances and many of the weaker ones. The laser **E** vector is $\perp$ to the Stark field, thus allowing only $\Delta M_J = \pm 1$ transitions, and most of the lines are 2- or 3-level resonances following this selection rule. Measured widths for these lines are $\lesssim 1$ MHz. There are 4-level collisional transferred resonances, one of which is indicated with the arrow in Fig. 2. These appear between the strong features and are significantly broader, having widths $\cong 2$ MHz. We have identified series of weak features not seen previously, which arise from 3-level resonances, one transition of which follows the usual $\Delta M_J = 1$ selection rule, while the other follows $\Delta M_J = 0, \pm 2$. Several such features are shown in Fig. 2. The breakdown of the ΔM_J selection rule arises from the ^{14}N nuclear quadrupole interaction, which mixes M_J states.

An example of the double resonance data is shown in Fig. 3, which shows the derivative of a diode laser scan of the $\nu_2 aQ(9,9)$ line at 921.255 cm^{-1} with a Stark field of 4278 V/cm and the CO laser locked via Lamb-dip stabilization to the $9 \leftarrow 8\nu_4 a^R R(9,9)$ line. The sub-Doppler features are associated with holes in the lower level M_J states. The strong central feature is the hole in the center ($v=0$) of the $M_J = 8$ state. This hole is collisionally transferred to the neighboring M_J states. Since the pump beam is a standing wave, transitions off resonance burn pairs of holes symmetrical

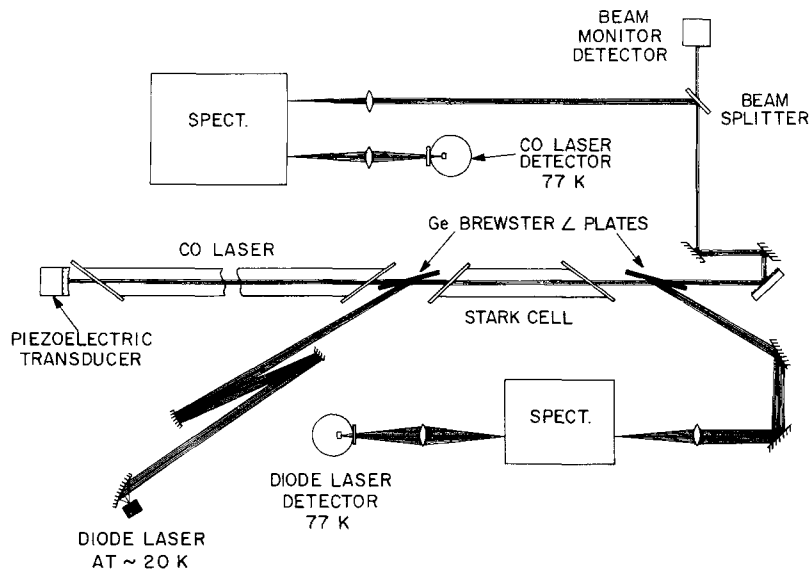

Fig. 1. Experimental setup for Stark-tuned Lamb-dip and IRDR spectroscopy. The 10 μm diode laser is polarized ⊥ to the CO laser and ∥ to the Stark field. The estimated intensities are 8 W/cm^2 for the CO laser and 2 mW/cm^2 for the diode

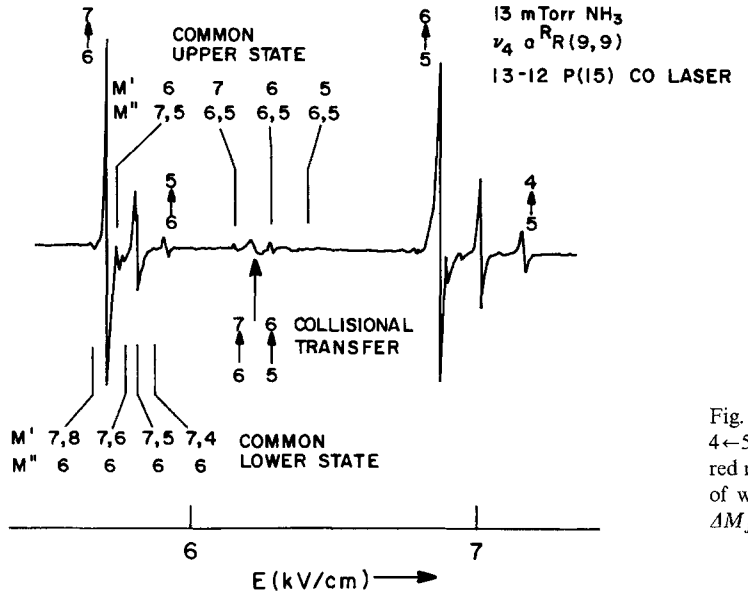

Fig. 2. Stark-tuned Lamb-dip spectrum of NH_3. The 7←6, 6←5, 5←6, and 4←5 lines are 2-level resonances. The broader 4-level collisional transferred resonance is in the center. The remainder are 3-level resonances, many of which involve transitions with the usually forbidden selection rules $\Delta M_J = 0, \pm 2$

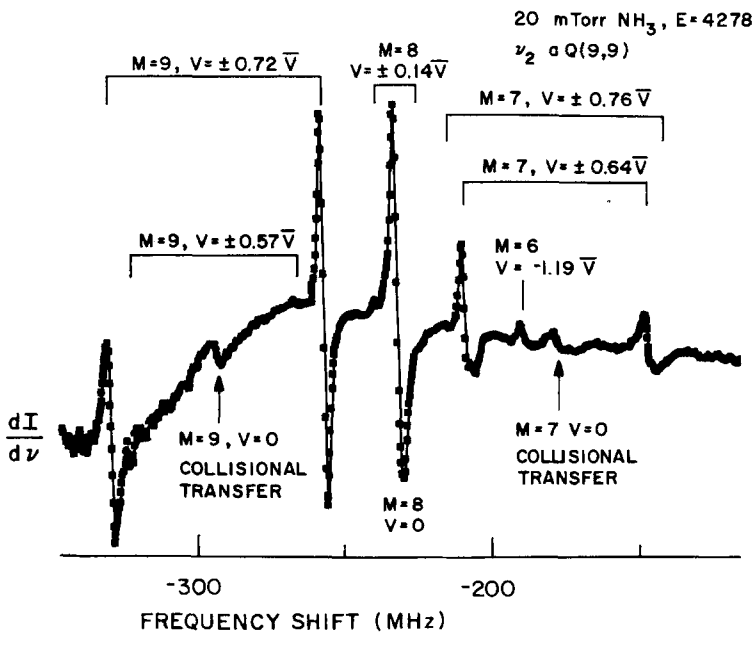

Fig. 3. A derivative diode laser scan showing sub-Doppler features in the ν_2 band induced by pumping with the CO laser in the ν_4 band. Features are labeled by the lower level M_J state and the velocity group in units of $\bar{v} = (2kT/M)^{1/2} = 5.33 \times 10^4$ cm/s. The frequency is relative to the zero field line

about the line center. For each lower level, there is one pair corresponding to the $\Delta M_J = +1$ and another to the much weaker $\Delta M_J = -1$ transitions. Within each pair the line with the two beams copropagating is $\cong 25\%$ narrower. The width (FWHM) of the narrowest transmission peaks is 3.2 MHz. The change in width with pressure was small and with laser power unobservable. Most likely the width is limited by the angular divergences of the laser beams ($f/D \cong 100$ and 300 for the diode and CO lasers, respectively) and the diode laser linewidth.

1. W.H.Weber, R.W.Terhune: Opt. Lett. **6**, 455 (1981)
2. J.W.C.Johns, A.R.W.McKellar, A.Trombetti: J. Mol. Spectrosc. **55**, 131 (1975)

Eigenvectors Flipping Spectroscopy

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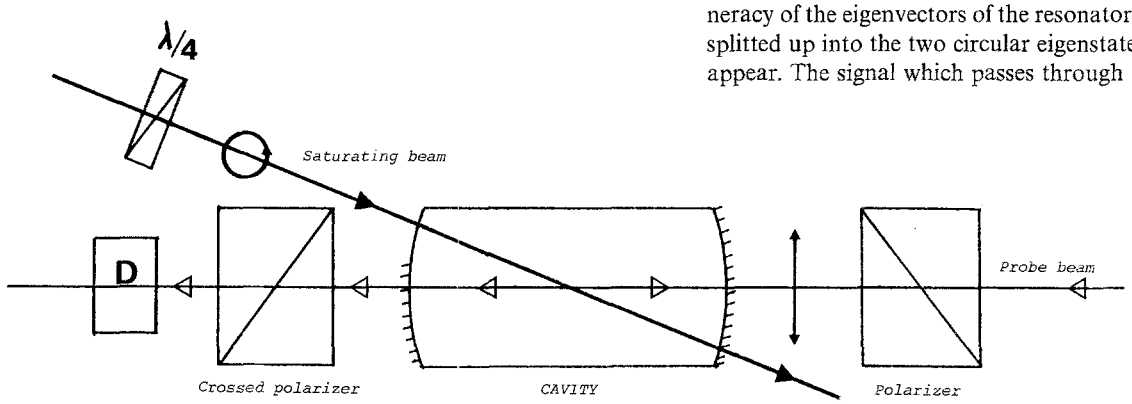
The polarization spectroscopy which has been developed these last years [1] leads to a signal/noise ratio 10^2 to 10^3 better than that given by saturated absorption for instance. However this enhancement of sensitivity is obtained with uncrossed polarizers which introduces then asymmetric resonances. We propose here to use the eigenvectors of an initially isotropic resonator for detection of small anisotropies induced in an atomic medium by a polarized saturating field. A probe field passing through the resonator is then splitted into the two eigenstates which appear in the medium leading to an eigenvector flipping spectroscopy. Recall attention on the fact that the physics of an isotropic resonator is very different than that of an anisotropic one where only deformation of an eigenstate is possible, as with an elliptical vibration for example [2, 3].

The experimental setup is represented on Fig. 1 together with the atomic levels scheme Ne^{20} which has been used here. A linearly polarized probe passes through an isotropic confocal resonator. The polarization is unchanged at the exit and the probe beam is switched off by the second crossed polarizer. Circular dichroïsm and birefringence are then induced in the atomic medium by a circularly polarized saturating field, which removes the degeneracy of the eigenvectors of the resonator. The probe field in then splitted up into the two circular eigenstates of polarization which appear. The signal which passes through the exit polarizer, when



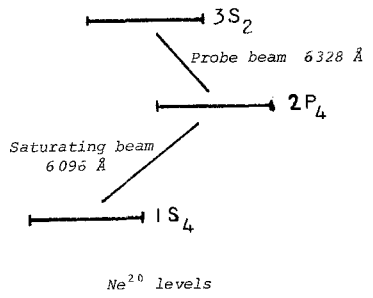


Fig. 1

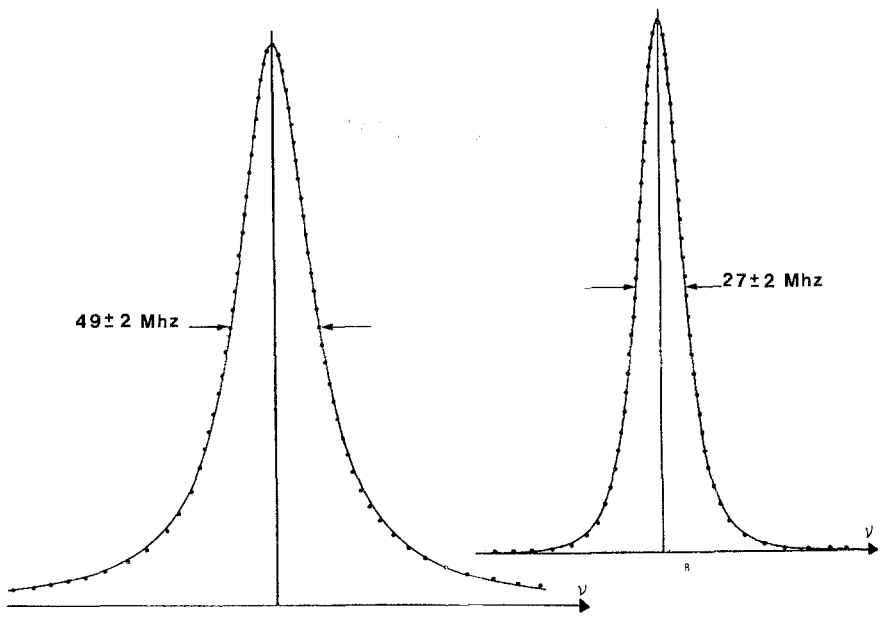


Fig. 2A and B. Experimental points and theoretical curves in polarization spectroscopy (A) and in eigenvector flipping spectroscopy (B)

the probe frequency is locked to the center of its Doppler profile, is given by:

$$I \sim \frac{T^2 E_0^2}{(1-R)^2} \cdot \left(\frac{1+R}{1-R}\right)^2 \cdot \Delta a^2 .$$

In this case indeed the birefringence is reciprocal and is compensated after a round trip, so only the circular dichroïsm Δa remains. The first term of the preceding expression represents the usual transmission of the cavity, the second one represents $4F^2/\pi^2$ where F is the finesse of the resonator. So in our case we expect an enhancement of sensitivity proportional to the square of the finesse, with also the square of a lorentzian as signal.

The classical polarization spectroscopy and eigenvector flipping spectroscopy have been performed simultaneously with the same atomic system. Experimental points and theoretical curves are represented on Fig. 2, leading to good fit in both experiments. The signal observed by the eigenvector flipping spectroscopy is obtained however with saturating field divided by 100. Taking account of the scale factor a gain of the order of several hundred is obtained. We recall that the half-height-width of the signal is smaller than that of the usual lorentzian ($\gamma \# 0.64\gamma_{ab}$). With optimized cavities gain of the order of 10^4 must be obtainable, still keeping symmetric resonances.

Hence, small atomic effects lead to large effects on the eigenstates of an isotropic resonator. The dynamics of the eigenstates of active [4] or passive [5] cavities reveal to be a powerful method for the measurement of small effects.

1. C. Wieman, T. W. Hänsch: Phys. Rev. Lett. **36**, 1170 (1976)
2. A. Sharmann, G. Hermann: Phys. Lett. **40** A, 293 (1972)
3. W. Radloff, H. H. Ritze: Opt. Commun. **35**, 203 (1980)
4. A. le Floch: Thèse, Rennes (1977) (unpublished)
5. A. le Floch, R. le Naour: C.R. Acad. Sci. (Paris) **288**, 225 (1979)

New Techniques for Sensitive Detection: Applications to Combustion Diagnostics

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The use of laser-based techniques for sensitive detection of atomic and molecular species has generated widespread interest. As part of a Sandia program in combustion diagnostics, we are currently developing crossed-laser-beam techniques to provide point measurements of minor species concentrations (<0.1%) in combustion environments. Spatially resolved versions of saturated absorption, optical Stark modulation, and optogalvanic spectroscopies have been demonstrated for the detection of atomic sodium in laboratory flames. We believe that the extension of these techniques to other atoms, molecules, and radicals will find many applications in combustion research.

Spatial resolution has been introduced into traditional line-of-sight absorption measurements by crossing a weak probe laser beam with an intense perturbing beam, and monitoring the change in absorption of the probe beam caused by the perturbing beam. The magnitude of the induced perturbation can be related to the absorption in the small volume common to the two beams. We have demonstrated this concept with two techniques. In saturated absorption spectroscopy [1], a resonant laser beam saturates the transition, decreasing the absorption of the probe beam. In optical Stark shift spectroscopy [2], an intense nonresonant beam induces a shift in the transition frequency, changing the absorption of the probe beam. Demonstrations of these techniques have been reported previously; further measurements will be presented at this conference.

Several spatially resolved versions of optogalvanic spectroscopy are also being explored. Intermodulated optogalvanic spectroscopy, a technique previously used for Doppler-free studies in gas discharges [3], has been adapted for making point measurements in flames (Fig. 1). In this method, two beams from a cw laser tuned to a transition in a flame species are "chopped" (amplitude modulated) at two different frequencies, and focused to a common point in the flame. Nonlinearities produced by optical saturation lead to modulations in the excited state population at harmonics of the chopping frequencies. A modulation at the sum of the two

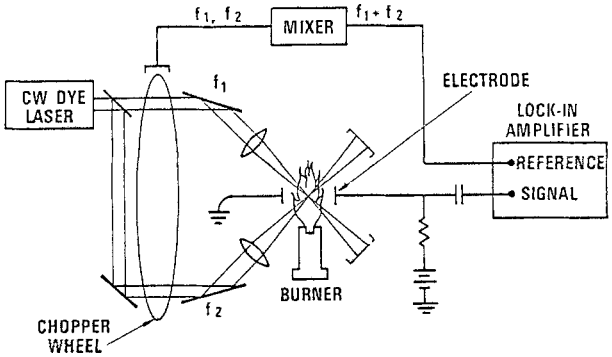

Fig. 1. Apparatus used to demonstrate spatially resolved intermodulated optogalvanic spectroscopy in flames

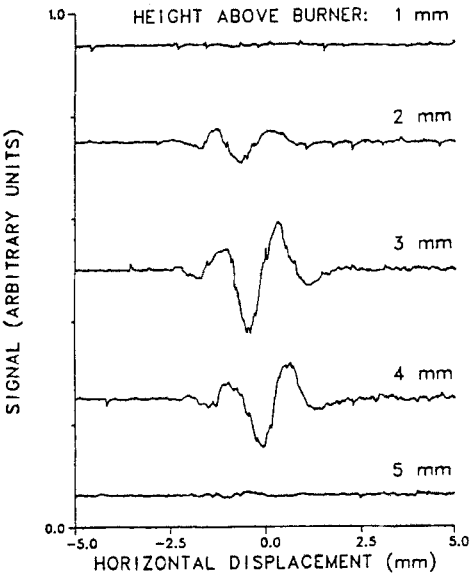

Fig. 2. Spatial scans showing the intermodulated optogalvanic signal at various heights above the burner

chopping frequencies is produced only in the small volume common to the two laser beams. This modulation is observed by using a lock-in amplifier tuned to the sum frequency to monitor the optogalvanic signal produced in the common interaction volume. This technique has been demonstrated in a study of atomic sodium aspirated into a hydrogen-air flame. The spatial scans shown in Fig. 2 were taken by tuning a cw dye laser to the

sodium $3S_{1/2}$–$3P_{3/2}$ transition, and translating the burner underneath the crossing point of the two beams from the laser. The "twin-peak" structure was also observed using spatially resolved saturated absorption and optical Stark modulation spectroscopies. The origin of the negative signal in the center of the flame is under investigation.

Finally, we have initiated efforts to explore applications of multiphoton optogalvanic and ionization detection schemes for combustion diagnostics. One of our major goals is the use of two-photon resonant excitation, three-photon ionization detection of atomic hydrogen in flames. Preliminary results from these studies will also be presented.

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1. J.E.M.Goldsmith: Opt. Lett. **6**, 525 (1981)
2. R.L.Farrow, L.A.Rahn: Opt Lett. **6**, 108 (1981)
   J.E.M.Goldsmith, R.L.Farrow: Submitted for publication
3. J.E.Lawler, A.I.Ferguson, J.E.M.Goldsmith, D.J.Jackson, A.L. Schawlow: Phys. Rev. Lett. **42**, 1046 (1979)