pected to differ since $(\omega/\omega_0)^2$ is not negligible. The differences exhibited by the data in Table I are close to estimates which we have made on the basis of Dawes'[8] theory. We conclude that the experimental dc-induced optical second-harmonic coefficients are consistent with the data presented, to within 1.5 times the combined uncertainty.

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# Precision Infrared Zeeman Spectra of $CH_4$ Studied by Laser-Saturated Absorption*

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Zeeman splitting of the methane 2947.912-$cm^{-1}$ $F_1^{(2)}$ line was studied. The $g$ factor of the rotational magnetic moment of methane was measured to be $g_J = +0.311 \pm 0.006$ and it was found that $g_J(\nu_3 = 1)$ is equal to $g_J(\nu_3 = 0)$. A Doppler-generated "level-crossing" saturated absorption signal was observed and is described.

There have been a number of studies exploiting the very narrow linewidths obtained through a variety of nonlinear optical processes.[1-5] In this Letter we report an investigation of the Zeeman splitting of the $F_1^{(2)}$ component of the methane $P(7)$ $\nu_3$ line.[6] Some aspects of the Zeeman structure of this transition have already been investigated by Luntz and Brewer[4] using a level-crossing method. Exploiting the particularly good precision capability of methane saturated absorption and frequency-offset locking, we have been able to resolve the magnetic splitting of the methane infrared line despite the smallness of the methane rotational magnetic moment.

Nonlinear absorption of 3.39-$\mu$m He-Ne laser radiation by methane has been thoroughly described in Ref. 1. We departed in this experiment from the standard arrangement described there by using an absorption cell external to the laser cavity, thereby making it possible to change readily the type of light polarization and its orientation as well as the orientation of the magnetic field. Frequency-offset locking, an eminently satisfactory way of obtaining precision line shapes and line-center shifts, was again a central feature of the measurement technique. Because a particular laser frequency can be offset from a reference laser with almost the same precision as contained in the reference, we are able, with either digital or analog procedures, to program a frequency scan of excellent reproducibility. The fluctuations of the absolute frequency of the controlled laser are about 150 Hz for a 1-sec averaging time.

We briefly consider the experimental details. (1) A reference laser is locked to the center of the methane absorption signal. (2) The beat-frequency signal between this reference and a second laser, the local oscillator, is obtained and is frequency-locked to a convenient value, 4500 kHz. (3) Finally, a beat is obtained between the local oscillator and the powerful laser oscillator that illuminates the external absorption-cell system. The external cell system is either locked to its absorption-line center (the beat frequency being monitored as the magnetic field is changed) or to a programmed frequency offset from the local oscillator source. The desired line shapes of the external cell absorption are then extracted by multiple-pass signal-averaging methods.

A 1-m-long $He^3$-$Ne^{22}$ laser with about 7mW of output power is the radiation source for the external cell. After passing through a Rochon polarizing prism, about 8% of this output is steered into one detector. This signal is used to dynamically control the laser excitation, thus producing

a flat baseline under the saturated-absorption resonance peak. The remaining light passes a $\lambda/4$ quartz plate, a mode-matching lens, and the absorption cell, and finally reaches a 100% retroreflecting mirror. The beam, after retracing its path back through the $\lambda/4$ plate, is orthogonally polarized to the input beam. Thus the polarizing prism steers the return beam away from the laser source and into the signal detector. Isolation of the laser output is adequate to present severe interference effects, although these effects do cause moderate drifts and baseline changes.[7] Modulation of the retroreflecting mirror position diminishes further the net effect of the interference. The performance of the system for these measurements was very adequate: Zeeman shifts as small as $5\times10^{-12}$ of line-center frequency were measurable.

Experiments were also performed using linear polarization, obtained by replacing the $\lambda/4$ plate with a Faraday rotator, an yttrium-iron-garnet (YIG) crystal. For fields beyond about 300 G this 5.85-mm-long crystal rotates the plane of polarization of the 3.39-$\mu$m radiation by almost exactly 45°.[8] Thus the return beam passes the Rochon prism and takes essentially the same path to the signal diode detector as in the circular polarization arrangement. Comparably good isolation is achieved both ways. The plane of linear polarization can be rotated by 90° simply by reversing the magnetic field in the YIG crystal. Thus with a transverse magnetic field, it is a simple matter to obtain either $\Delta M=0$ alone, or $\Delta M=\pm1$ together. Similarly with a longitudinal magnetic field one may obtain pure $\Delta M=+1$ or $\Delta M=-1$ transitions using the wave plate, and $\Delta M=\pm1$ transitions with the YIG isolator.

The sense of circular polarization may be deduced from the orientation of the optic axis of the $\lambda/4$ plate relative to the vertical output polarization of the laser. The small birefringence of crystal quartz (0.007 at 3.39 $\mu$m) yields a first-order quarter-wave plate near our plate thickness of 0.13 mm. The sense of rotation is thus toward the "slow" axis, which is the optic axis in crystal quartz. The use of a fused-silica Fresnel rhomb was helpful in confirming this assignment.[9]

In Fig. 1 we show typical saturated absorption lines obtained with circularly-polarized light and longitudinal magnetic field. From observation of the sense of circular polarization relative to the direction of the magnetic field, the results shown in Fig. 1 allow us to determine that the $g$ value of the observed transition is positive.[10]

The value of $g_J$ was extracted with high preci-

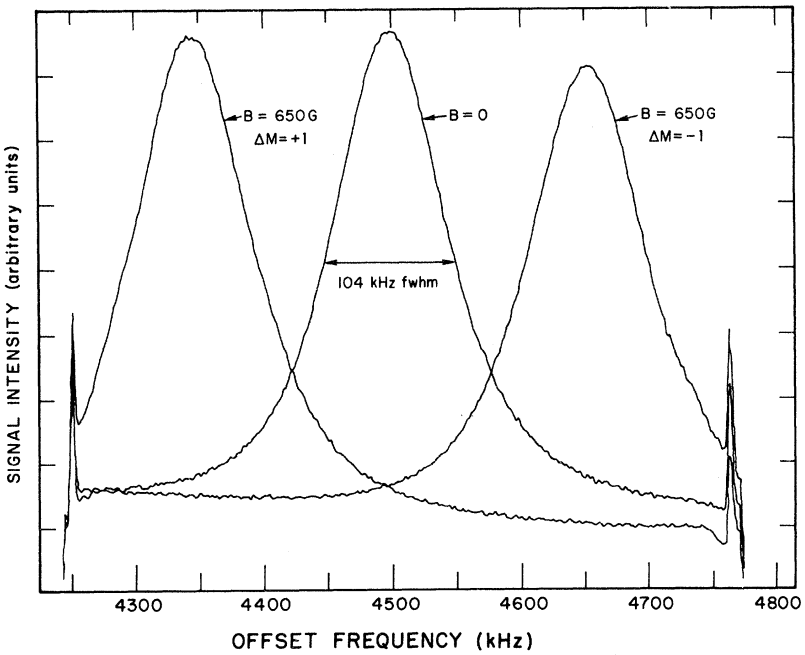


FIG. 1. Saturated-absorption resonance lines obtained for one sense of circular polarization at zero field and for opposite orientations of the solenoid field. The frequency scale indicates the real sense of line-center shift. The $\Delta M$ assignments were made by noting the orientation of the quarter-wave-plate optic axis relative to the plane of linear polarization of the laser-output radiation. See text. The sharp spikes at the ends of the sweep arise from electrical transients associated with the baseline-sampling circuit. The $CH_4$ pressure is about 1 mTorr.

sion from the locked mode of measurement: The external-cell laser frequency was locked to the zero of the first frequency derivative of the external-cell absorption signal. The change in the detected beat frequency between the external-cell laser and the local-oscillator laser was recorded for a variety of magnetic field strengths. No departure from a linear shift with field was observed.[11] With an uncertainty fixed primarily by the calibration accuracy of the gaussmeter used to measure the field, the result is $g_J = +0.311 \pm 0.006$.[12] Because, as described below, we did not detect a difference of $g_J$ between the upper and lower vibrational states, we can make a direct comparison with the molecular beam measurement of $|g_J|$ by Anderson and Ramsey.[13] Our result is in excellent agreement with their value, $|g_J| = 0.3133 \pm 0.0002$, and their conjecture about the probable sign of $g_J$ is evidently correct.[10]

With the magnetic field still parallel to the light beam but with linearly polarized light we obtain a triplet line structure shown in Fig. 2. The allowed $\Delta M = \pm 1$ transitions in this configuration are split apart in the expected way, but now we obtain a third central peak exactly midway between the two side peaks. From the nonlinear-physics viewpoint this is a novel feature, which to our knowledge has not been unambiguously exhibited before although this and other such effects were considered theoretically by Schlossberg and Javan.[14] In the usual situation, a signal is observed from the contribution of those absorbers having nearly zero axial velocity. In that case both running waves of the standing-wave field are able to interact with these absorbers. In the nonlinear regime of high light intensity, a narrow Lorentzian peak in the transmitted intensity is observed. On the other hand, the central peak of the triplet structure here arises from the contribution of molecules with a fixed nonzero axial velocity, the value of which is determined by the separation of the $\Delta M = \pm 1$ transitions. For this resonance, oppositely running waves of opposite circular polarization are absorbed, resulting in a saturation peak that is similar to the zero axial-velocity case. In the present experiment the splitting of the final-state energy, $2M$, arises from magnetic interaction with the external field, so $M = -g_J \beta_N H_0$. The velocity required for resonance, $v_z = cM/\nu_0$, is thus about 55 cm/sec. The increment of internal energy just corresponds to the Doppler-induced change of frequency, both transitions depleting the same initial state.

The earlier theoretical considerations resulted in a prediction that such an absorption would occur with an intensity that is the geometrical mean of the two side intensities. This prediction is not borne out by our observation. The central intensity is smaller than the side intensities by about a factor of 2 in Fig. 2 and also for a variety of

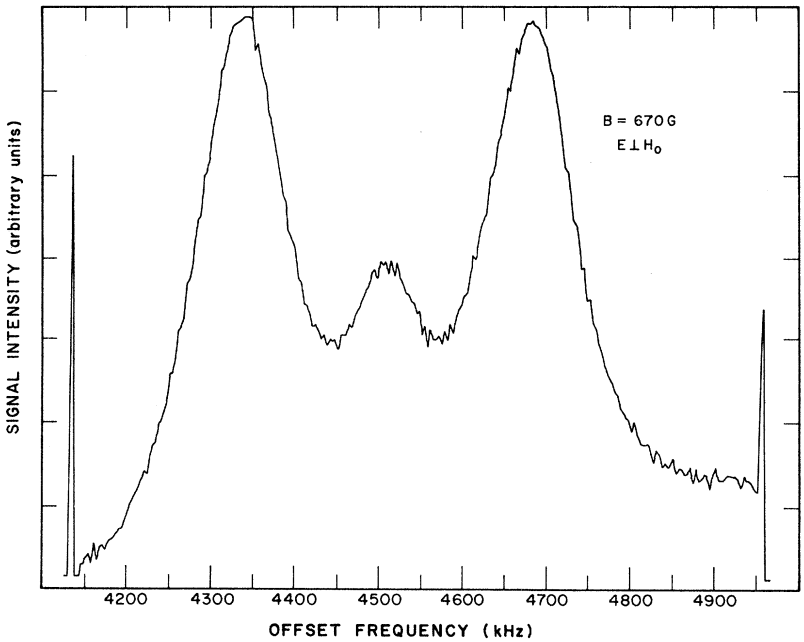


FIG. 2. Line splitting obtained with a solenoid field parallel to the light beam and with linear light polarization. The side peaks are the $\Delta M = \pm 1$ transitions allowed for this configuration. The center peak is the Doppler-generated "level-crossing" signal. The magnetic field is uniform to about ±1% over the 1-m absorption-cell length.

other conditions and splittings and configurations.[15] We note that in this Doppler-generated saturation signal, the absorbing molecules are required to travel axially along the light beam. They thus sample several oscillations of the standing-wave pattern during their lifetimes. No such sampling occurs for the absorbers traveling transverse to the light beam, the condition found in the standard case.[16]

The $\Delta M = 0$ transition was measured with linearly polarized light and a transverse magnet configuration. This study is of interest because it bears directly on the question of $g_J$-factor equality between the ground and first vibrational state of $\nu_3$. In the $\Delta M = 0$ situation, field inhomogenities do not significantly influence the line shape. Thus any observed broadening may be interpreted as arising from the difference of the magnetic energies (for a given $m_J$) in the upper and lower vibrational states.

Data were taken for two cases, $H_\perp = 0$ and $H_\perp = 1.3$ kG. The resonances were computer-fitted by Lorentz functions by an iterative least-squares program. The increase of linewidth due to the transverse magnetic field turned out to be $-1.6 \pm 2.6$ kHz, with no increase of residuals. Considering that the initial state has $J = 7$, we may conclude that the $g_J$ factors of the $\nu_3 = 0$ and $\nu_3 = 1$ vibrational states are equal to within something like 2 or 3 parts in $10^3$. Although it is known that the rotational constants of the two states differ by 5%,[17] this fact evidently does not ensure that the two $g_J$ factors differ by a comparable amount.[18]

It is almost superfluous to emphasize the remarkable spectroscopic precision and resolution now available with suitable laser methods and apparatus. The shift by 1 part in $10^{12}$ of the absolute position of a spectral line can be detected in about 2 seconds' integration. The resonance curves of Figs. 1 and 2 show a working spectral resolution of just about $10^9$ and a signal-to-noise ratio exceeding 50:1 for 5 minutes' integration. This high potential resolution is not deteriorated by drifts, acoustic or thermal disturbances, etc. —even for periods of hours—when appropriate use is made of frequency-offset-locking methods.

The authors are happy to acknowledge discussions between one of us (J.L.H.) and R. G. Brewer on this subject, and we thank him for communicating his results to us prior to publication.

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[9]Dark "channel spectra" may be observed in transmitted broadband light with the quartz crystals (and optionally the Fresnel rhomb) between crossed (parallel) infrared polarizers, corresponding to even (odd) multiples of $\pi/2$ differential phase delay. This set of wavelengths, along with the known birefringence in the near infrared, allows "bootstrapping" the birefringence into the 3-$\mu$m region where a modest ($\pm 5\%$) sample dependence is observed. We are grateful to D. A. Jennings and to W. H. Tanttila for making available to us their selections of $X$-cut quartz crystals.

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[11]For state-selected absorbers, the hyperfine interaction of 10 kHz, given by C. H. Anderson and N. F. Ramsey, Phys. Rev. 149, 14 (1966), would result in observable curvature of the frequency-field relationship near 50 G. However, our thermal distribution

of absorbers is not significantly aligned, so with fifteen different $m_J$ levels, one may only observe a modest broadening in the intermediate-field region.

[12]See for example C. H. Townes and A. L. Schawlow, *Microwave Spectroscopy* (McGraw-Hill, New York, 1955). An expression for rotational magnetic energy is given on page 292.

[13]Anderson and Ramsey, Ref. 11.

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[15]Any detector bias can be estimated by the dissimilarity of signal intensity of the two side peaks of Fig. 2. The Rochon prism may steer one orientation of linear polarization more effectively into the signal detector; a departure from this orientation causes a decrease in detected intensity. It is evident that the bias is small in Fig. 2.

[16]The Doppler-induced resolution of the two cavity waves into two distinct frequencies changes the details of the time averages appropriate to a calculation of the saturation signal. In the low-saturation limit, using the methods of Ref. 2 or Ref. 14, one easily derives a predicted factor-of-two intensity decrease of the saturation signal. This effect operates when the type of resonance being studied requires the absorber to interact with two resolvably different rest-frame frequencies, i.e., where the molecular internal-energy change has two distinct values.

[17]In Ref. 4; and G. Herzberg, *Molecular Spectra and Molecular Structure: II. Infrared and Raman Spectra of Polyatomic Molecules* (Van Nostrand, Princeton, N. J., 1945), p. 455.

[18]See Ref. 12, Sec. 11-6, p. 290 ff.

# Pulse Propagation in a High-Gain Medium

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It is found that ultrashort pulses in a high-gain 3.51-$\mu$m xenon laser propagate through the amplifying medium at a velocity less than the vacuum speed of light by as much as a factor of 2.5. The pulse velocity is a function of the gain and agrees with the group velocity.

It has been argued theoretically[1,2] that the velocity $v$ of pulse propagation in amplifying or absorbing media is equal to the group velocity $d\omega/dk$. Resonance dispersion may cause the group velocity to be greater (in an absorbing medium) or smaller (in a gain medium) than the phase velocity. These dispersion effects have been observed experimentally[3-5] using the weak 6328-Å transition in neon, but the changes in group velocity were less than 1 part in 1000.

In the following we report on the pulse velocity in a xenon discharge near its amplifying 3.51-$\mu$m transition. In this case the combination of high optical gain (>40 dB/m in our experiment) and narrow linewidth result in extremely large dispersion. The observed pulse velocity is less than $\frac{1}{2}c$. Furthermore, using an analytic expression for the gain dependence of the index of refraction of the Doppler-broadened transition, we show that the pulse propagation velocity agrees with the group velocity.

The classical group velocity is

$$v_g = \frac{c}{n + \nu dn/d\nu}. \tag{1}$$

The frequency-dependent index of refraction of an inhomogeneous Doppler-broadened medium has been given by Close.[16] If saturation is unimportant and if the homogeneous linewidth (~4 MHz) is negligible compared to the Doppler width $\Delta\nu_D$, this expression may be written as

$$n(\nu) = 1 + cgF(x)/2\pi^{3/2}\nu, \tag{2}$$

where $F(x)$ is Dawson's integral,

$$F(x) = e^{-x^2}\int_0^x e^{t^2}dt. \tag{3}$$

The frequency is measured by $x = [2(\nu-\nu_0)/\Delta\nu_D] \times (\ln 2)^{1/2}$, and $g$ is the small-signal incremental intensity-gain constant at line center.

Equations (1) and (2) can in principle be combined to obtain the frequency-dependent group velocity. We are most interested in the behavior near line center where $F(x) \approx x$. Then Eq. (2) may be written

$$n(\nu) = 1 + cg(\nu-\nu_0)(\ln 2)^{1/2}/\pi^{3/2}\nu\Delta\nu_D. \tag{4}$$

From Eq. (1) the group velocity is then given by

$$v_g = c(1+\beta)^{-1}, \tag{5}$$