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CONFIDENTIAL

SUBMITTED TO Southwest Conference on Optics, Albuquerque, NM, March 1985

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SENSITIVE DETECTION OF TUNABLE DIODE LASER ABSORPTION
BY POLARIZATION ROTATION*

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Abstract

Results are presented demonstrating that diode laser absorption sensitivity can be increased by using the Faraday or Kerr effect to rotate the polarization of the source so that crossed polarizers can be used to reduce source noise. The Faraday effect is demonstrated using nitrous oxide in a magnetic field, and the Kerr effect is demonstrated using methyl fluoride in an electric field. Some description of how absorption in an external field causes polarization rotation is presented as well as some conclusions on the types of molecules for which this technique is best suited.

*Work performed under the auspices of the U.S. DOE.

Introduction

In many applications of infrared spectroscopy the best source is a Pb-salt tunable diode laser because of its tunability (typically 100 cm^{-1} in the $3\text{-}30\text{ }\mu\text{m}$ region) and narrow bandwidth (0.0005 cm^{-1}). However, absorption sensitivity using tunable diode lasers is limited by intensity fluctuations of the diode laser output. This source noise of a tunable diode laser has been shown to have an excess noise 24 dB above the quantum noise limit when operated at threshold current and increases when operated at higher currents.¹ Thus, to improve the sensitivity of diode laser absorption spectroscopy so that an absorption signal becomes larger than a noise-equivalent signal, the source noise must be reduced.

The limiting sensitivity in an absorption measurement is due to statistical quantum noise in detecting the source intensity.² A technique that can ideally reduce source noise below the quantum noise limit for any absorption measurement is polarization rotation of the source using an applied external magnetic (Faraday effect) or electric field (Kerr effect). This technique was first used to enhance absorption sensitivity in infrared absorption by nitrous oxide at its first vibrational overtone, $2.7\text{ }\mu\text{m}$, using a color center laser as the source and a magnetic field to rotate the polarization.³ The work presented here shows that polarization rotation using either a magnetic⁴ or electric⁵ field can be used to reduce the source noise of a diode laser.

Polarization Rotation Spectroscopy

The basic principle of polarization rotation spectroscopy is to make the polarization of a linearly polarized source be rotated when the source is tuned to an absorption line of the gas to be studied. A polarization analyzer is

placed in front of a quantum-noise limited detector. The polarization analyzer is crossed relative to the source polarization so that signal reaches the detector only when absorption, and thus polarization rotation, occurs, consequently source noise reaching the detector is reduced.

Several methods exist to create an absorption that causes a polarization component perpendicular to the source polarization to occur. One method used with visible wavelength dye lasers is a saturation technique using two counter-propagating lasers to obtain Doppler-free spectra. Polarization saturation is achieved using a high-power circularly polarized saturating laser with a linearly polarized probe laser.⁶ Linear polarization is composed of equal amounts of left-handed circular (LHC) and right-handed circular (RHC) polarization. The effect of the circularly polarized saturating beam is to cause absorption of the probe beam to be restricted to circular polarization in the opposite sense of the saturating beam, which causes the linear polarization to become elliptically polarized. Then a polarization analyzer in front of the detector is crossed relative to the initial linear polarization of the probe laser so that the amount of elliptical polarization is measured with reduced source noise and increased sensitivity.

This circular polarization saturation technique is very similar in theory to the polarization rotation technique described here, which uses an external magnetic field directed along the source beam propagation direction, i.e., the Faraday effect. The magnetic field causes a rotational line to split into its M_J Zeeman components, causing a difference in absorption for LHC polarization, which undergoes $\Delta M = +1$ transitions, and RHC polarization, which undergoes $\Delta M = -1$ transitions. Thus, this technique causes a polarization rotation due to a

difference in absorption for LHC and RHC polarization, just as with the absorption saturation technique.

The difference in absorption for LHC and RHC polarization causes the initial linear polarization to be rotated in two ways. The difference in absorption causes the amplitudes of the electric vector \mathbf{E} of the LHC and RHC polarization components to be different, but with no relative phase change. The sum of these two components gives elliptical polarization, which has its minor axis perpendicular to the initial linear polarization. The difference in absorption for the LHC and RHC polarization components also causes a difference in index of refraction for the two components due to the Kramers-Konig relationship. This birefringence causes a relative phase shift between the LHC and RHC polarization components. A relative phase shift between the two components with no change in relative amplitudes would cause the resulting polarization to still be linear, but rotated relative to the initial linear polarization. When both effects are combined the resulting polarization is elliptical with the major axis tilted from the initial linear polarization direction due to the birefringence.

A polarization rotation technique analogous to the Faraday effect is the Kerr effect, which uses an electric field rather than a magnetic field. In theory, however, these two techniques are somewhat different because an external electric field must be directed transverse to the source beam propagation direction. This means the initial linear polarization of the source cannot be deconvoluted in LHC and RHC polarization components propagating along the external field direction. However, when the electric field direction is 45° relative to the source polarization, it can be deconvoluted into linear polarization components parallel and perpendicular to the external field. In a

manner analogous to the Faraday effect, the electric field splits an absorption line into its Zeeman components, which causes a difference in absorption for parallel polarization with $\Delta M = 0$ transitions, and perpendicular polarization with $\Delta M = \pm 1$ transitions. A difference in absorption causes the resulting polarization to be rotated but remain linear. The related birefringence causes the resulting polarization to become elliptical. The sum of the two effects again gives elliptical polarization with the major axis tilted relative to the initial linear polarization.

Using the Faraday effect and Kerr effect, polarization rotation is demonstrated for diode laser absorption in NO at $5.5 \mu\text{m}^4$ and for the ν_6 band of methyl fluoride at $8.4 \mu\text{m}^5$. The results are compared to other modulation techniques, which shows that this technique gives the best noise reduction and best sensitivity.

Experimental Apparatus

In both the Faraday-effect and Kerr-effect experiments, a Pb-salt tunable diode laser system designed to operate at regulated cryogenic temperatures was used, along with a one-meter monochromator to isolate one of the several modes emitted by the diode. The diodes were operated in the usual manner, using temperature for coarse wavelength tuning and current ramping to scan a range on the order of one wavenumber. Lenses or mirrors were used to focus the diode output onto the monochromator entrance slit, where the beam was mechanically chopped when obtaining direct absorption spectra. The beam exiting the monochromator was collimated and then passed through a cell and focused onto either an indium antimonide detector for $5.5 \mu\text{m}$ used in Faraday rotation in NO or a HgCdTe detector for $8.4 \mu\text{m}$ used in Kerr rotation in CH_3F . In the polarization

rotation experiments a set of crossed polarizers was used at both ends of the cell. The polarizers used consisted of a stack of ZnSe Brewster plates with an extinction coefficient of 1:500 at both wavelengths used. Birefringence in the cell windows in an empty cell was checked using the crossed polarizers and was found to be less than could be detected.

To obtain direct absorption spectra the beam was mechanically chopped at 400 Hz, with the signal fed into a lock-in amplifier. Wavelength scans were recorded on an X-Y recorder. Frequency modulation, which is the technique typically used for sensitive diode laser spectroscopy, was accomplished by adding a sinusoidal current to the DC or slowly ramped current into the diode. A comparable technique as far as sensitivity is Stark or Zeeman modulation of the absorption line, which was accomplished using either a Zeeman or Stark cell. The Zeeman cell consisting of a one meter Pyrex tube wrapped with copper wire and modulated using an LC circuit with a power supply giving a 880 Hz sinusoidal magnetic field directed along the cell with a strength up to 200 gauss. The Stark cell consisted of a one meter cell with internally mounted parallel plates with a one centimeter gap, two inch width, and one meter length, and was modulated using a high-speed switchable, high-voltage power supply giving up to 5000 V/cm and 200 Hz. For Kerr effect studies the electric field was at 45° to the laser polarization giving equal polarization parallel and perpendicular to the electric field. Absorption lines in CH_3F were identified in the usual manner using standard wavelength calibration gases⁷ along with known spectral assignments of CH_3F .⁸

Results and Discussion

Polarization rotation was observed in NO using the Faraday effect. The signal-to-noise ratio was compared to that obtained for direct absorption measurements and Zeeman modulation as shown in Fig. 1. In addition, a comparison with frequency modulation is shown in Fig. 2. Frequency modulation should give the same signal-to-noise as Zeeman modulation when using a modulated field strong enough to split the line into its Zeeman components greater than the Doppler linewidth (or pressure broadened linewidth). As can be seen, the polarization rotation signal has a reduced noise level compared to the frequency modulation technique, which is the standard technique to improve sensitivity in diode laser absorption.

In an experiment analogous to the NO experiment, polarization rotation was observed in the spectrum of the ν_6 band of methyl fluoride using the Kerr effect. Figure 3 shows a comparison of this technique to amplitude modulation and Stark modulation for the $P_{Q_0}(J)$ branch. As can be seen, the polarization rotation technique has an increased signal-to-noise ratio over the other techniques, especially for the lower J lines. However, for a symmetric top such as methyl fluoride several types of rotational transitions within one band are possible, some of which show a weaker polarization rotation effect than others. Other transitions in which polarization rotation could not be observed were the $R_2(J)$ line and lines of the $Q_1(J)$ branch. A weak polarization rotation signal was observed for low J lines of the $Q_0(J)$ branch, but with less sensitivity than with Stark modulation. This is a result of the difference in absorption for the two components of the initial polarization and the splitting of the Zeeman components of a rotational line being dependent upon the type of transition. This is illustrated in Fig. 4, which shows calculated line

strengths and line splitting for three different rotational lines. Line splitting must be larger than the Doppler width (or pressure broadened width) in order for a difference in absorption for the two polarization components to be observed.

Conclusions

Polarization rotation using the Faraday or Kerr effect has been demonstrated to be a useful technique in improving the sensitivity of diode laser absorption. However, as was demonstrated for methyl fluoride, not all rotational lines of a symmetric molecule show the same amount of polarization rotation. The reason for this is similar to the reason why only linear molecules with a non-degenerate ground state ($J \neq 0$) can show polarization rotation.

In practice, polarization rotation using the Faraday effect is easier to implement than that using the Kerr effect because of the problem of electrical breakdown of the gas using a Stark cell. However, most molecules do not have unpaired spin or electronic angular momentum, which is needed for the Faraday effect. For these types of molecules, the Kerr effect can be used as long as the molecule has a permanent electric dipole moment. So, in general, the technique of polarization rotation can be used to increase sensitivity in diode laser absorption, but it is more useful for certain types of molecules than for others.

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Figure Captions

Figure 1. Comparison of three different modulation schemes used to detect nitric oxide absorption. They are: (a) amplitude modulation using a 400 Hz chopper; (b) Zeeman modulation at 880 Hz; (c) Faraday polarization rotation. The NO pressure was 0.3 torr.

Figure 2. Comparison of (a) frequency modulation at 5 kHz to (b) Faraday polarization rotation at 880 Hz. The NO pressure was 0.1 torr.

Figure 3. Comparison of three different modulation schemes used to detect absorption at the P_{Q_1} branch of the ν_6 band of methyl fluoride, with increasing wavenumber to the right. They are (a) amplitude modulation; (b) Stark modulation; (c) Kerr polarization rotation. The modulation frequency was 200 Hz in all three methods. The Stark cell voltage was 400 V, and the CH_3F pressure was 0.1 torr.

Figure 4. Calculated relative intensities and line splitting in an electric field are shown for the (a) $P_{Q_1}(1)$, (b) $R_{Q_2}(1)$, and (c) $R_3(3)$ lines with the source polarized parallel and perpendicular to the electric field. The relative intensities are normalized so that the strongest component in each rotational line has the same strength. The relative frequency scale is the same for all three lines, so the splitting is the same for (a) and (b), but smaller for (c).

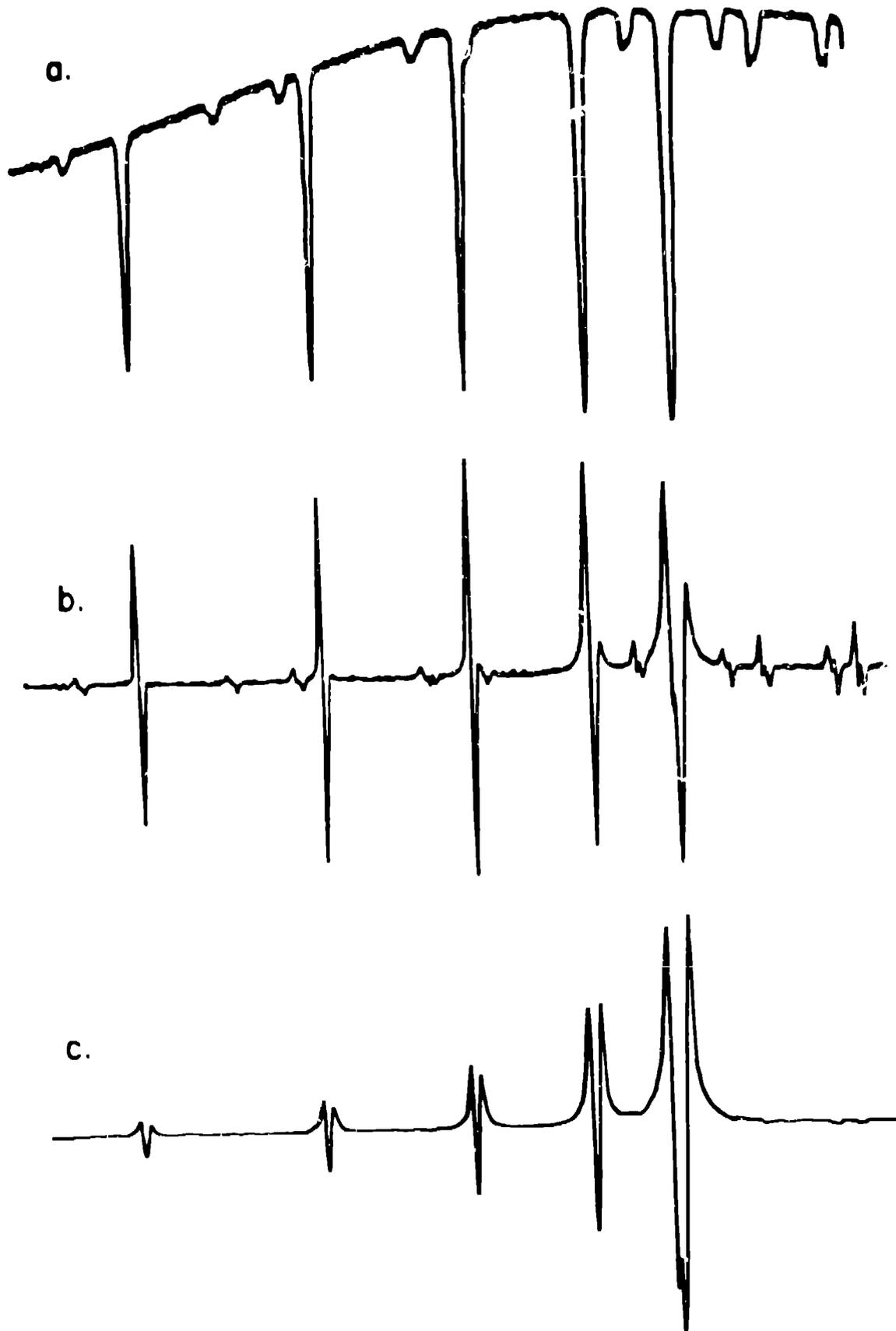


Figure 1

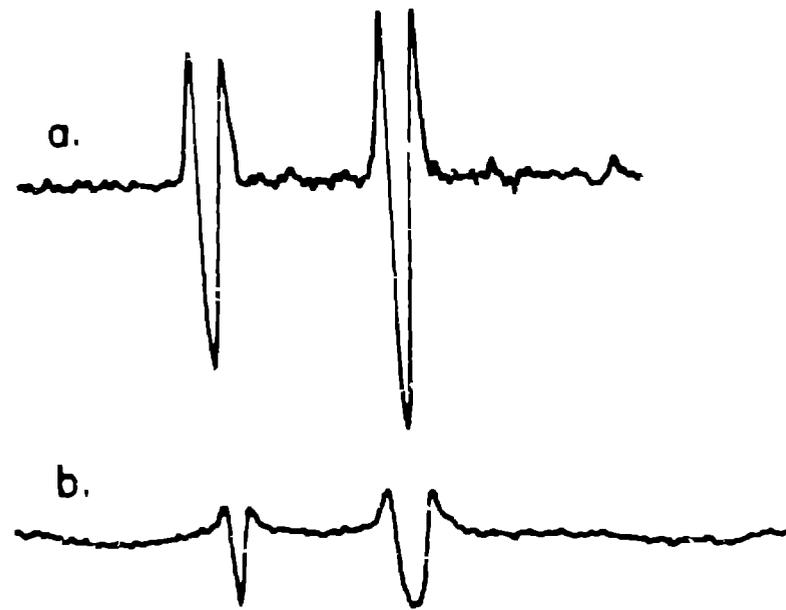


Figure 2



Figure 3

CHM-VG-6975

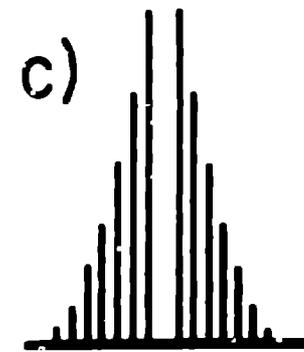
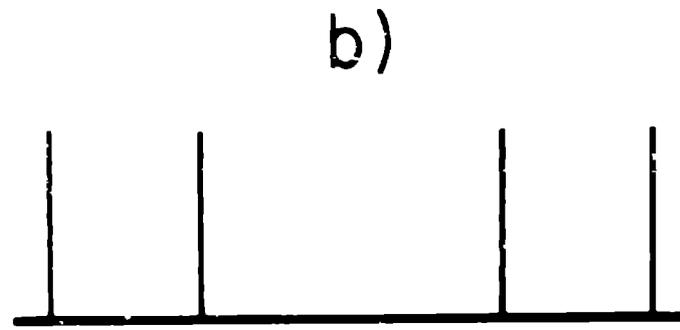
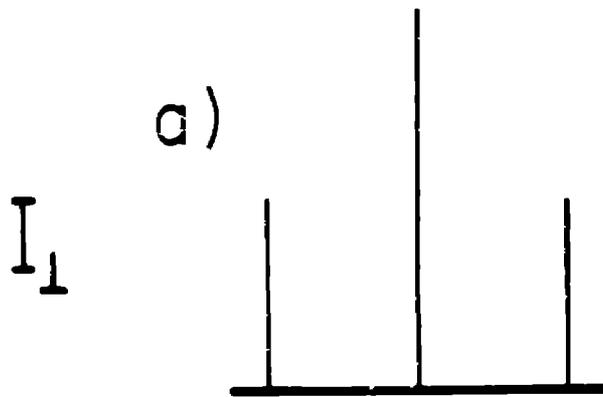
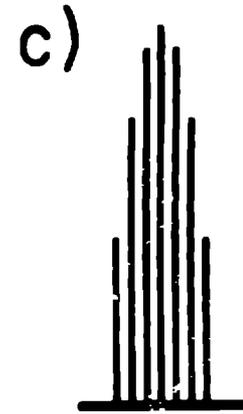
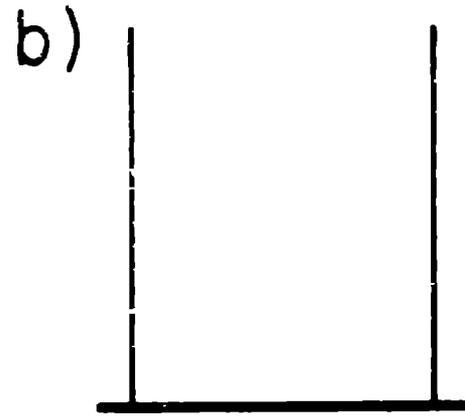
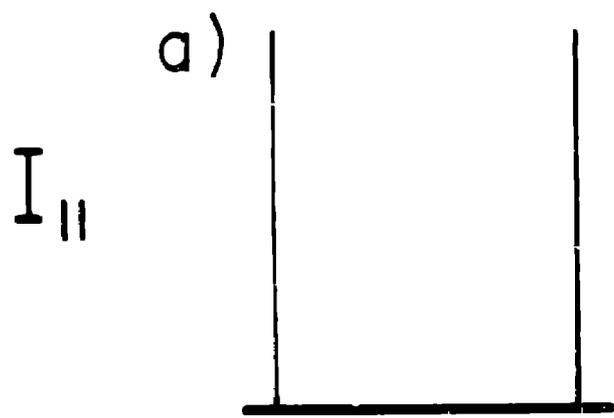


Figure 4