

# Intracavity polarimeter for measuring small optical anisotropies

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An intracavity polarimeter for measuring small optical anisotropies is described. It is based on an optically pumped sodium dimer ring laser that supports two counterpropagating, orthogonally polarized modes. The optical anisotropy of an intracavity sample is detected by its effect on the beat frequency between the two modes. An analysis of the noise shows that a phase-retardation sensitivity of  $6 \times 10^{-8}$  rad can be achieved. A preliminary measurement of the Kerr effect in  $\text{CO}_2$  is reported.

## INTRODUCTION

A medium is said to be optically anisotropic when its properties depend on the polarization of the incident light. Optical anisotropies, such as birefringence, optical activity, and Faraday rotation, can occur naturally in a material or be induced by external forces or fields. These effects are conventionally measured by observing changes in the polarization of radiation that has traversed a sample.<sup>1</sup> In the case of field-induced effects, such measurements are often made relative to a standard such as nitrobenzene or carbon disulfide.<sup>2</sup> Typical reported<sup>3-5</sup> phase retardations for electric-field-induced birefringence (the Kerr effect) are of the order of  $5 \times 10^{-7}$  rad. It is of interest to improve this sensitivity to study small effects such as the optical activity associated with parity violation in atoms,  $\chi^3$  anisotropies, and optical anisotropies in thin films. It is also desirable to make absolute measurements of various anisotropies in systems simple enough to permit a quantitative comparison with theory. In this paper we describe a new polarimeter that is both absolute and sensitive. It is based on a dual-polarization laser with an intracavity cell.

The technique is simple in principle. The pioneering work of Doyle and White<sup>6</sup> showed that an anisotropic cavity can support two mutually orthogonal polarization modes. The difference in frequency between the two modes is a direct measure of the anisotropy of the cavity and can be determined by beating the two together with a square-law detector. If an anisotropic sample is introduced into the cavity or if an additional anisotropy is induced in a sample already present in the cavity, the *change* in the beat frequency is a measure of this extra anisotropy. This is the feature that we exploit in our polarimeter. In practice one must take into account effects of the gain medium, the residual anisotropy of the optical components, and whether the extra anisotropy alters the polarization modes of the original cavity. We have found that these complicating effects can be minimized by using an optically pumped sodium dimer ring laser<sup>7-10</sup> and have used such a laser to construct and test a polarimeter with a sensitivity of  $6 \times 10^{-8}$  rad. The potential exists to extend this sensitivity further by several orders of magnitude.

The paper begins with a discussion of design criteria and a description of the experimental apparatus. Next the sensitivity of the technique is estimated by analyzing fluctuations in the beat frequency that are due to noise. We conclude with a preliminary measurement of the dc Kerr effect in  $\text{CO}_2$  and a summary.

## APPARATUS

Figure 1 shows a schematic layout of the apparatus. Since the setup is rather complex, we preface the description of the apparatus with a discussion of the properties of an anisotropic cavity and optically pumped sodium that led us to the design shown in the figure.

Consider a sodium dimer ring laser that is longitudinally pumped by a single-frequency argon-ion laser. It is well known<sup>11</sup> that for many dimer lasers the gain results from a resonant Raman- or  $\Lambda$ -type transition, as shown in Fig. 2. In this figure levels 1 and 2 are occupied and unoccupied rovibrational levels of the ground electronic state, respectively. In our system, in which sodium is pumped by blue-green light, level 3 belongs to the excited  $B$  state. For such Raman processes, the amplification profile in the forward direction, i.e., in the direction of the pump beam, is sharply peaked, while that in the backward direction is broad and low.<sup>11</sup> Consequently, longitudinal optical pumping of a ring dimer laser can produce unidirectional laser operation in the same direction as the pump beam. For sodium, the effective Doppler width in the forward direction is comparable with the spectral free range of our 3.2-m ring cavity, and one can obtain a single longitudinal mode without additional frequency-selective intracavity elements. Furthermore, lasing can be restricted to the lowest-order transverse mode by properly matching the pump-beam profile to the  $\text{TEM}_{00}$  mode of the ring-laser cavity. In the following paragraph we examine the polarization of such a mode.

The residual optical anisotropy of the ring laser must be known if one is to predict the polarization modes of the bare laser, i.e., without the sample present. As is shown in Fig. 1, the windows of the sodium gain cell are perpendicular to the optic axis of the ring cavity, so that orthogonally polarized

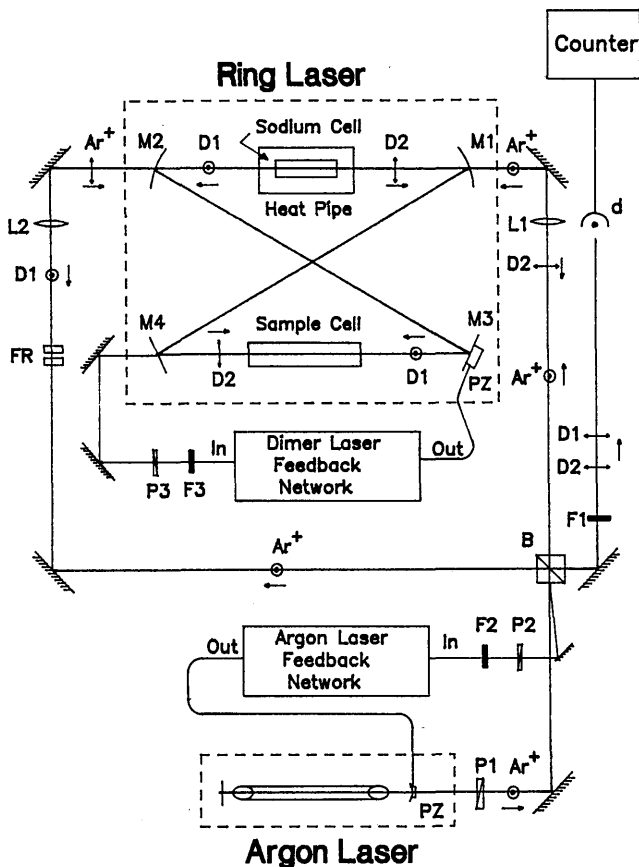


Fig. 1. Schematic diagram of the intracavity polarimeter. Abbreviations are defined in text.

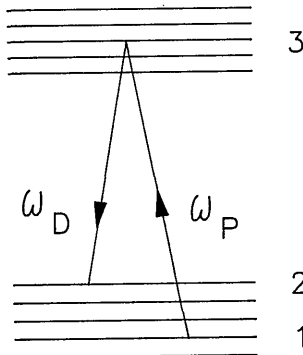


Fig. 2. Energy-level diagram of an optically pumped dimer laser. For our apparatus,  $\omega_P$  is the frequency of the  $Ar^+$  pump beam and  $\omega_D$  is the frequency of the  $Na_2$  laser.

modes will experience identical losses. The use of thin windows and the absence of étalons, optical diodes, etc. in the cavity ensures that residual strain birefringences play a negligible role in determining the polarization modes of the cavity. We find experimentally and have confirmed by calculations<sup>7</sup> that dominant source of anisotropy in the ring cavity is the phase shifts on reflection at the mirrors; *s*- and *p*-polarized beams experience different phase shifts for reflections at nonnormal incidence. Consequently, the modes of the bare laser are linearly polarized parallel and perpendicular to the plane of the ring. We have found that either of these polarization modes can be selectively excited by

pumping with light of that polarization. This implies that the gain for light polarized parallel to the pump polarization is greater than for the orthogonal polarization. In summary, the pump beam can be adjusted such that a single-longitudinal, single-transverse, single-polarization mode with a single direction of circulation is excited in the ring laser. We pump bidirectionally with orthogonally polarized beams whose polarizations are aligned with the modes of the ring cavity and thus obtain two dimer laser beams counterpropagating around the ring that themselves have orthogonal polarizations.

Now consider what happens when a birefringent sample is inserted into the cavity or when a birefringence is induced in a sample by applying an external field. We shall describe a simple plane-wave theory for a ring laser with a cavity of length  $L$  and index  $n$  containing an intracavity sample cell of length  $l$  with principal indices  $n_1$  and  $n_2$ . The cell is oriented in the cavity such that its principal axes are aligned with the polarization modes of the bare laser. This leaves the polarization modes the same as those of the bare laser. The cavity resonance condition for mode 1 is  $2\pi m_1 = (2\pi\nu_1/c)[n(L-l) + n_1l] + \phi_1$ , where  $m_1$  is the longitudinal mode number and  $\phi_1$  the total phase shift at the mirrors. Thus the frequency difference, or beat frequency, between the two polarization modes is given by

$$\nu_b = \nu_2 - \nu_1 = \frac{c(m_2 - \phi_2/2\pi)}{n(L-l) + n_2l} - \frac{c(m_1 - \phi_1/2\pi)}{n(L-l) + n_1l}. \quad (1)$$

We are interested in cases in which the birefringence,  $\Delta n = n_1 - n_2$ , is so small that  $\nu_b$  is much less than the spectral free range of the cavity. In that case the counterpropagating beams have the same longitudinal mode number, and Eq. (1) becomes

$$\nu_b - \nu_b^0 = f\nu\Delta n, \quad (2)$$

where  $\nu_b^0$  is the bare-laser beat frequency, i.e., the beat frequency for  $n_1 = n_2$ . The filling factor,  $f$ , is  $l$  divided by the total optical length of the cavity. Equation (2) is the basic equation relating the measured change in the beat frequency to the optical anisotropy.

If dispersion of the gain medium can be neglected, fluctuations in the cavity length of the ring laser affect the frequency of both counterpropagating modes equally. This common mode rejection leads to a beat frequency that is insensitive to changes in the mean frequency of the laser arising from mechanical and thermal variations in the cavity length. Assuming a beat-frequency resolution of 1 Hz and a filling factor of the order of 1, it follows from Eq. (2) that the polarimeter is sensitive to birefringences of the order of  $10^{-15}$ .

The above discussion was couched in terms of linearly polarized modes and birefringence. With the addition of quarter-wave plates to the cavity, one could obtain left- and/or right-circularly polarized light in the region of the sample cell, and one could then observe either the Faraday effect or optical activity.

With this background, we turn now to consider the details shown in Fig. 1. The ring-laser cavity, formed by mirrors M1-M4 in a bow-tie configuration, contains both the sodium and the sample cell. Heating of the dimer cell was provided by a double heat pipe,<sup>12</sup> a design that yields a homogeneous

temperature and density distribution of the sodium vapor. We find that optimum performance of the ring laser occurs with the argon buffer gas at a pressure of 3 to 5 Torr, which, for proper heat-pipe operation, requires a temperature in the range of 430 to 490°C. The bidirectional pumping was achieved by splitting the output beam, labeled Ar<sup>+</sup> in Fig. 1, of a single-frequency argon-ion laser with a 50/50 beam splitter, B. Lenses L1 and L2 were adjusted to match the Gaussian profiles of the pump beams to the profile of the TEM<sub>00</sub> modes of the ring-laser cavity. The output polarization of the argon laser was originally perpendicular to the plane of the ring laser. The polarization of one pump beam was rotated by 90° by two Fresnel rhombs, FR. As indicated above, this pumping scheme excites two orthogonally polarized, single-frequency, counterpropagating modes, D1 and D2, inside the ring-laser cavity.

In order to measure the beat frequency we must combine beams D1 and D2 on a detector, such that their polarizations are the same. The two outputs transmitted by the dichroic mirrors M1 and M2 retrace the path of the two argon pump-laser beams. Beam 1, which was vertically polarized in the ring cavity, becomes horizontally polarized and parallel to the polarization of beam 2 on passing through the Fresnel rhombs. The two beams are combined at the beam splitter and directed onto d, an RCA Model C30950E avalanche photodiode. This detector has a sensitivity of 0.2 V/μW and a bandwidth of 50 MHz. The frequency of the beat signal was measured with an HP5316A frequency counter interfaced to a computer.

The experimental apparatus just described would be complete if dispersion in the sodium gain medium could be neglected. However, fluctuations in the differential mode pulling by the gain medium limit the stability of the beat frequency and hence the sensitivity of the apparatus. Our estimates,<sup>8</sup> based on the data of Man and Brillat,<sup>13</sup> show that fluctuations in the differential pulling can cause variations in the beat frequency of some tens of kilohertz. These fluctuations are due to changes of the position of the modes within the gain profile and changes in the magnitude of the gain due to variations in the pump intensity. Several steps were taken to reduce the fluctuations in the beat frequency. Fluctuations caused by acoustic and thermal noise sources were reduced by enclosing both lasers in acoustically shielded boxes. The stability of the beat frequency was further enhanced by locking the frequency of the argon and the dimer lasers to external reference cavities, by stabilizing the intensity of the argon laser, and by matching the gains of the ring laser through the alignment of the pump-laser beams. Each reference cavity had a finesse of 30 and was housed in a pressure-tight, temperature-stabilized container. Alignment of the laser beams to the reference cavities was purposely off axis to reduce the coupling of the reflected beams to the rest of the system. Frequency fluctuations in both lasers were reduced to less than 1 MHz over a period of 10 sec by a feedback system and piezoelectric (PZ) crystals, which were used to adjust the lengths of the lasers. The polarizers P1-P3 and filters F1-F3 helped to isolate optically the different sections of the system and thus also contributed to the stability of the polarimeter.<sup>7</sup>

The pump source was a single-frequency Spectra-Physics Model 52 Ar<sup>+</sup> laser. Intensity fluctuations in the frequency range 0.01 to 10 Hz were less than 1%. In order to achieve

the required frequency stability with our simple feedback system, we modified the laser. The plasma tube, along with its magnet and water-cooling supply, was suspended by wires. The mirrors and wavelength-selective elements were mounted directly on a vibration isolation table. This partly isolates the acoustic noise associated with the cooling water from the Invar rods that define the length of the cavity.

There are approximately 45 laser transitions in sodium when it is pumped with an argon laser. We find experimentally that it is easiest to obtain stable single-line, single-frequency operation of the ring laser by pumping with the 457.9-nm line of the argon laser while lasing on the 560-nm line of sodium. For a pump-laser power in the 10-mW range, the output power of the dimer laser is of the order of 10 μW. Operating with the widest possible wavelength separation between the pump and dimer lasers results in better stability because it lowers the finesse of the ring cavity at the pump frequency. A lower finesse means there is less resonant enhancement of the pump intensity inside the ring cavity. Thus a variation in the length of the ring cavity produces a smaller change in the stored pump power and concomitantly a smaller change in the amplitude of the gain.

We mentioned above that the largest source of anisotropy is the phase shift at the mirrors. This leads to a beat frequency  $\nu_b^0$  of 2.7 MHz for the bare laser. It is advantageous to decrease this intrinsic beat frequency. We know from theory<sup>14</sup> that a smaller beat frequency should lead to a reduction in differential mode pulling. In addition, the frequency counter used in this experiment gave six digits of resolution. As a result, beat frequencies greater than 1 MHz would limit the resolution to 10 Hz or to a birefringence of 10<sup>-14</sup> for our polarimeter. The beat frequency was reduced below 1 MHz by introducing an antireflection-coated optical flat into the cavity. The flat was squeezed to produce a phase shift due to strain birefringence, which partially compensated for the phase shift at the mirrors.

For Eq. (2) to remain valid, the intracavity cell must be aligned with the polarization modes of the cavity, and thus it is necessary to maintain stable and well-defined polarization modes. For this reason, care was taken not to overcompensate for the phase shift due to the mirrors. Overcompensation occurs when other sources of birefringence in the cavity become important enough significantly to alter the polarization modes of the cavity. If the beat frequency were reduced to zero, the laser would be effectively isotropic. Stephan and Hugon<sup>15</sup> have shown that in such lasers the polarization modes are extremely sensitive to stray effects. It was found that the stability of the beat frequency improved as the frequency was decreased to approximately 10 kHz, where the laser became unstable. As a compromise, the laser was operated with the bare-laser beat frequency reduced to ~200 kHz, which resulted in slightly improved stability and permitted a resolution of 1 Hz on the frequency counter.

Finally, it is known that the homogeneous width of the gain curve is ~10 MHz,<sup>13</sup> and thus the observed stability at values of the mode splitting much smaller than 10 MHz requires an explanation. We interpret the lack of mode competition as being due to velocity selection. For pump frequencies off the center of the absorption line, the resonant Raman condition preferentially selects a specific class of sodium dimers with nonzero velocity along the direction of the pump. For bidirectional pumping, therefore, the gain

of each dimer laser mode is obtained from separate counter-moving groups of molecules, and one expects little mode competition. Coupling of the modes by means of scattering at the mirrors does not appear to be a significant. This concludes the description of the intracavity polarimeter. In the next section we determine the sensitivity of the apparatus under the conditions described above.

### BEAT-FREQUENCY STABILITY

In the experiment described below we wish to measure the change in the beat frequency on application of a static electric field to an intracavity sample. Thus it is the variation of the beat frequency that limits the precision with which the change in frequency may be measured. The optimum conditions under which to carry out a measurement will depend on the exact nature of the variations, and consequently an essential preliminary measurement is the characterization of the temporal behavior of the beat frequency.

Figure 3 shows the variation in the beat frequency,  $\nu_b^0$ , over a single 3-min time interval. We see that the variations are mostly deterministic,<sup>16</sup> i.e.,  $\nu_b^0$  is an irregular yet continuous function of time. Deterministic noise arises from variations in the length of the cavity, from fluctuations in the pump power, etc. Apart from drift, we see from the figure that  $\nu_b^0$  has a mean excursion of  $\sim 1$  kHz, which is much greater than the targeted 1-Hz precision. Because the noise is deterministic, for the Kerr effect one can employ rapid modulation of the field so that the variation in  $\nu_b^0$  is small over one cycle of modulation. When differences are taken between successive field-on and field-off frequency measurements there is a large cancellation of the noise. The optimum modulation frequency can be chosen by considering the Allan variance<sup>16</sup> of  $\nu_b^0$ .

The Allan variance,  $\sigma^2$ , is often used to characterize frequency stability. For  $N$  consecutive measurements, each of duration  $\tau$ ,  $\sigma^2$  is given by  $\sigma^2(\tau) = \sum\{y[(n+1)\tau] - y(n\tau)\}^2/2(N-1)$ , where  $y(n\tau)$  is the deviation of the frequency of the  $n$ th

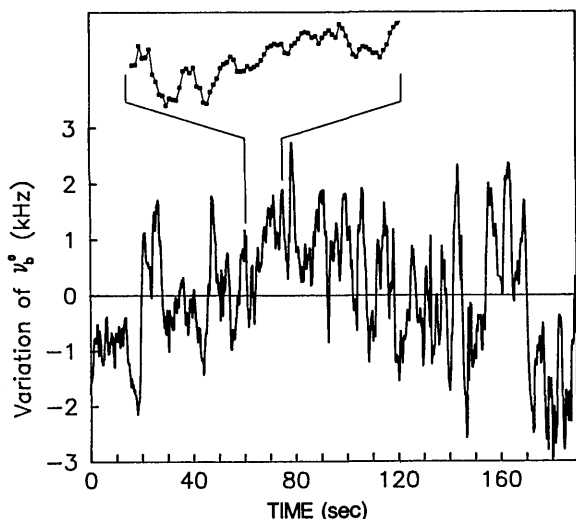


Fig. 3. Measured variation in the beat frequency of the bare laser, over a 3-min time interval. The mean beat frequency was approximately 400 kHz. Successive data points were measured for an integration time of 100 msec and separated by a 90-msec dead time. The expanded section shows the variation over a 15-sec period.

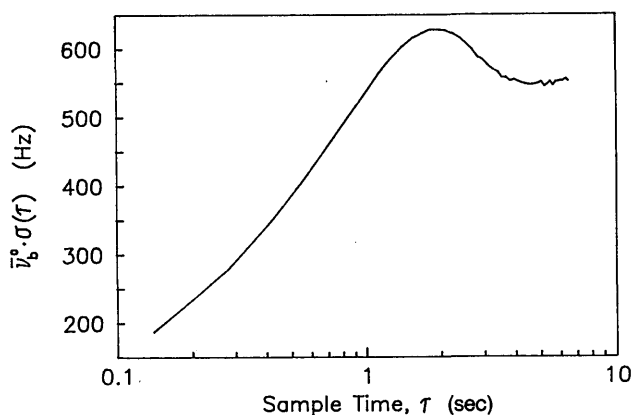


Fig. 4. Plot of the average beat frequency  $\nu_b^0$  times the square root  $\sigma(\tau)$  of the Allan variance as a function of sampling time. This is a measure of the stability or rate of change of the beat frequency at various sampling times.

measurement from the mean frequency, divided by the mean frequency. The Allan variance is a measure of the fractional average fluctuation from one reading to the next contiguous reading. The minimum value of  $\sigma^2(\tau)$ , as a function of  $\tau$ , gives the most suitable modulation frequency ( $1/\tau$ ) for phase-sensitive detection. The Allan variance, as defined above, assumes zero dead time between measurements. We will neglect the presence of a nonzero dead time in our measurements, as we do not require a precise characterization of the noise.

The Allan variance as a function of time was computed for the data of Fig. 3. For each point the integration time,  $\tau_c$ , was 100 msec, and the dead time was 90 msec. These data, consisting of 1000 consecutive measurements of the beat frequency, were lumped into sets of equal size,  $m$ . The average frequency of each of the  $2000/m$  sets was used as a new set of frequencies corresponding to a measurement time of length  $m\tau_c$ . The square root of the Allan variance,  $\sigma(m\tau_c)$ , was then calculated for this new data set. This procedure was one of convenience, since a single record of the frequency could be used to construct an Allan variance for a large range of  $\tau = m\tau_c$  simply by using a computer and altering the choice of  $m$ .

Figure 4 shows the results of these calculations, where we have plotted the mean frequency  $\bar{\nu}_b^0$  times  $\sigma(\tau)$ . The approximate asymptotic value of 600 Hz at long times reflects the mean frequency excursions apparent in Fig. 3. We are interested in the short-time behavior of  $\bar{\nu}_b^0\sigma(\tau)$ , where the minimum of approximately 190 Hz occurs at the shortest sampling time. Other data sets showed various minimum values of  $\bar{\nu}_b^0\sigma(\tau)$ , but the rapidly decreasing value at short times was characteristic of all Allan variance plots. This indicates that it would be desirable to have still higher modulation frequencies because the minimum in  $\sigma(\tau)$  has not been reached.

The potential sensitivity of the apparatus was calculated by analyzing data, like those of Fig. 3, without modulating the anisotropy of the intracavity cell. One simply pretends that consecutive readings correspond to field on, field off. As there is no real field, the change in the beat frequency will be zero within the noise or sensitivity of the apparatus. On a particularly good night, when the acoustic noise in the building was low, the average deviation was found to be 36 Hz for a

data run consisting of 2000 points sampled at the maximum rate. This translates into a sensitivity of 1.2 Hz, approaching the limit of 1 Hz set by the frequency counter. A resolution of 1.2 Hz corresponds to a phase shift of  $6 \times 10^{-8}$  rad or, for a large filling factor, to a birefringence of  $2 \times 10^{-15}$ . For the modest filling factor used in our preliminary experiment, the sensitivity was quite sufficient to permit measurement of the Kerr constant of  $\text{CO}_2$  at low pressures.

### TEST OF THE APPARATUS: THE KERR CONSTANT OF $\text{CO}_2$

The Kerr effect was discovered by the Reverend John Kerr in 1875. While Kerr expressed his results in terms of a phase shift, the length of the sample, and the electric-field strength, it is now common practice to define a Kerr constant on a molecular basis. Like Buckingham and Orr,<sup>17</sup> we define the low-density molar constant  $A_K$  by

$$A_K = \frac{2}{27} \frac{\Delta n}{\rho E^2}, \quad (3)$$

where  $\Delta n$  is the difference between the index of refraction of light polarized parallel and perpendicular to the applied field  $E$  and  $\rho$  is the molar density. For our polarimeter, Eq. (2) directly relates a measured change in beat frequency to  $\Delta n$ , and thus we can determine  $A_K$ , provided that we also know or measure  $E$ ,  $\rho$ , and the filling factor  $f$ .

The intracavity Kerr cell was a 26-cm stainless-steel cylinder closed on the ends by two antireflection-coated fused-silica windows 3 mm thick. It was designed for use up to 50 atm. A uniform electric field was supplied by two parallel metal plates separated by a distance of  $3.80 \pm 0.03$  mm. The voltage applied to the Kerr cell was square-wave modulated with a peak voltage of  $1.91 \pm 0.01$  kV, using a Kepco OPS2000 power supply under the control of the computer. This corresponds to an electric field  $E$  of  $503 \pm 4$  kV/m.

Matheson Aquarator grade  $\text{CO}_2$ , with impurities less than 21 parts in  $10^6$ , was used to fill the cell to pressures ranging from 1 to 50 atm. The cell was evacuated to 100 mTorr and filled twice to 50 atm to purge impurities. The temperature of the cell was  $25.0 \pm 0.5^\circ\text{C}$ , and pressures were measured using a calibrated Bourdon gauge. The pressures were converted to molar densities,  $\rho$ , using thermodynamic data.<sup>18</sup>

To calculate  $f$  we must know the length of the Kerr cell and the optical length of the laser cavity. The index of refraction of air,  $\text{CO}_2$ , and the sodium gain medium was taken to be unity. The five fused-silica windows ( $n = 1.46$ ) inside the cavity occupied a total length of 1.5 cm, while the physical length of the cavity was measured to be  $322 \pm 2$  cm. This gives an optical length of  $323 \pm 2$  cm for the cavity. The length of the electrodes in the Kerr cell was  $20.00 \pm 0.05$  cm. The effective length over which the electric field was applied is somewhat longer than the plates, owing to the fringing fields at both ends. We solved Laplace's equation numerically and calculated the effective length of the cell by using the equation

$$l = \frac{2 \int_0^\infty E^2(x) dx}{E^2(0)}. \quad (4)$$

Here  $E$  is the component of the electric field perpendicular

to the axis of the cell and the field plates. The result is  $l = 20.14 \pm 0.05$  cm, which yields a filling factor,  $f$ , equal to  $0.0625 \pm 0.0004$ .

The change in the beat frequency was measured at several pressures ranging from 1 to 50 atm. The modulation frequency was 3.5 Hz, i.e., field on for 1/7 sec, field off for 1/7 sec, a frequency determined by the maximum rate of data transmission to the computer. Figure 5 shows the change in the beat frequency,  $\nu_b(E) - \nu_b^0$ , due to the Kerr effect, as a function of molar density. An error-weighted least-squares procedure was used to fit the data to a straight line, giving a slope of  $0.274 \pm 0.002$  kHz  $\text{m}^3/\text{kg}\cdot\text{mol}$  and an intercept of  $1.6 \pm 3.4$  Hz. The line passes through the origin within experimental error, as expected. Combining Eq. (2) and Eq. (3) with the slope of the line in Fig. 5 and the values of  $\nu$ ,  $f$ , and  $E$  yields the value of  $(2.40 \pm 0.05) \times 10^{-27}$   $\text{C}^2 \text{m}^5/\text{J}^2$  for the molar Kerr constant of  $\text{CO}_2$ .

Previous measurements of the Kerr effect in  $\text{CO}_2$  include the work of Quarles<sup>19</sup> and Buckingham *et al.*<sup>20</sup> We deduce from the early work of Quarles that the dispersion in the molar Kerr constant is small over the visible spectrum. We can therefore compare our results obtained at 560 nm with the measurements of Buckingham obtained at 633 nm. Interpolating Buckingham's measurements of  $A_K$ , which were made over a range of temperatures, we find that  $A_K = 2.78 \times 10^{-27}$   $\text{C}^2 \text{m}^5/\text{J}^2$  at 298 K. This is 14% higher than our result. Buckingham claims an absolute accuracy of  $\pm 5\%$  arising from difficulties in calibration, while our estimated random error is  $\pm 2\%$ . There is a systematic difference between the two results, but we do not consider the discrepancy to be serious for an initial experiment.

The most probable source of systematic error is associated with matching the principal axes of the field-induced birefringence to the axes of the polarization modes of the cavity. Well after the experiment was performed we reexamined the polarization modes as the beat frequency was reduced with the optical flat. It was found that the polarization modes occasionally rotated as much as  $10^\circ$  when the beat frequency was reduced to values typical of those used in the experiment. We have estimated<sup>7</sup> the systematic error that such a rotation would have and find that it would decrease the change in the beat frequency or Kerr constant by some 15%. A misalignment of the Kerr cell would have a similar effect.

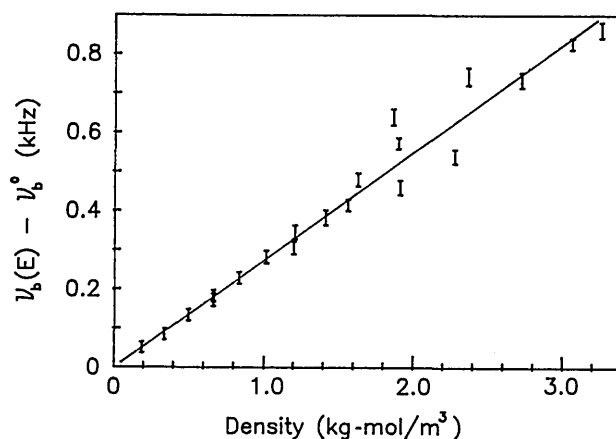


Fig. 5. Density dependence of the change in the beat frequency due to the Kerr effect in  $\text{CO}_2$ . The maximum density corresponds to a cell pressure of approximately 50 atm.

Simple experimental steps can be taken in future experiments to eliminate such systematic errors.

There are a number of possible ways to decrease the random error or to increase our signal-to-noise ratio. The small filling factor of 0.065 was a consequence of using a cell that was on hand from a previous experiment. Filling factors of close to 1/2 are possible and would increase the signal by an order of magnitude. The Allan variance measurements reported above indicated that higher modulation frequencies are needed to reduce the noise. We have made some preliminary noise measurements with a phase-locked loop to convert the beat frequency to a voltage. This permits modulation frequencies of up to 100 kHz, rather than the 3.5 Hz used here, and should increase the signal-to-noise ratio by another 2 orders of magnitude.

## SUMMARY AND CONCLUSIONS

In this paper we have described the development and testing of a sensitive intracavity polarimeter based on an optically pumped sodium dimer laser. The sensitivity of the polarimeter is of the order of  $6 \times 10^{-8}$  rad. An analysis of the noise of the beat frequency indicates that higher modulation frequencies should permit us to increase the sensitivity significantly. The absolute Kerr constant of CO<sub>2</sub> was measured with an estimated random error of  $\pm 2\%$ . Even at the present level of sensitivity, both the dc and the optical Kerr constants, including Raman resonances, should be measurable absolutely in dilute systems.

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## REFERENCES AND NOTES

1. C. G. Lefevre and R. J. W. Lefevre, "The Kerr effect," in *Techniques of Chemistry*, A. Weissberger, ed. (Wiley Interscience, New York, 1972), Vol. 1, Part 3C, pp. 399-427.
2. A. D. Buckingham and D. A. Dunmur, "Kerr effect in inert gases and sulphur hexafluoride," *Trans. Faraday Soc.*, **64**, 1776 (1968).
3. A. K. Burnham, L. W. Buxton, and W. H. Flygare, "Kerr constants, depolarization ratios, and hyperpolarizabilities of substituted methanes," *J. Chem. Phys.* **67**, 4990 (1977).
4. A. D. Buckingham and H. Sutter, "Gas phase measurements of the Kerr effect in some *n*-alkanes and cyclohexane," *J. Chem. Phys.* **64**, 364 (1976).
5. One of us has recently shown that it is possible to achieve the shot-noise limit of the conventional technique [D. P. Shelton and R. E. Cameron, *Rev. Sci. Instrum.* (to be published)]. The fundamental sensitivity of the intracavity technique is better and will be discussed in a future publication.
6. W. M. Doyle and M. B. White, "Generation of combination tones by the interaction of orthogonal oscillations in a gas laser," *Appl. Phys. Lett.* **10**, 224 (1967).
7. S. C. Read, "Measurement of the Kerr effect in carbon dioxide using intracavity polarimetry," M.Sc. thesis (University of Toronto, Toronto, Canada, 1987).
8. Lai Ming, "Frequency stabilization of an argon ion laser and a sodium dimer ring laser for use in optical anisotropy measurements," M.Sc. thesis (University of Toronto, Toronto, Canada, 1985).
9. T. B. Cave, "An investigation of the sodium dimer ring laser for use in optical anisotropy measurements," M.Sc. thesis (University of Toronto, Toronto, Canada, 1983).
10. A. D. Guest, "Dispersion measurements with a dual-frequency infrared helium-neon laser for use in optical anisotropy measurements," M.Sc. Thesis (University of Toronto, Toronto, Canada, 1981).
11. B. Welleghausen, "Optically pumped cw dimer lasers," *IEEE J. Quantum Electron.* **QE-15**, 1108 (1979).
12. H. Scheingraber and C. R. Vidal, "Heat pipe oven of well-defined column density," *Rev. Sci. Instrum.* **52**, 1010 (1981).
13. C. N. Man and Brillet, "Gain lineshapes in optically pumped dimer lasers," *Opt. Commun.* **45**, 95 (1983).
14. W. R. Bennett, "Gaseous optical masers," *Phys. Rev.* **126**, 580 (1962).
15. G. Stephan and D. Hugon, "Light polarization of a quasi-isotropic laser with optical feedback," *Phys. Rev. Lett.* **55**, 703 (1985).
16. J. A. Barnes, A. R. Chi, L. S. Cutler, D. J. Healy, D. B. Leeson, T. E. Mcgunigal, J. A. Mullen, Jr., W. L. Smith, R. L. Sydnor, R. F. C. Vessot, and G. M. R. Winkler, "Characterization of frequency stability," *IEEE Trans. Instrum. Meas.* **IM-20**, 105 (1971).
17. A. D. Buckingham and B. J. Orr, "Electric birefringence in molecular hydrogen," *Proc. R. Soc. London Ser. A* **305**, 259 (1968).
18. J. H. Dymond and E. B. Smith, *The Virial Coefficients of Gases: A Critical Compilation* (Clarendon, Oxford, 1980), p. 37.
19. G. G. Quarles, "The dispersion of the electro-optical Kerr effect in carbon dioxide," *Phys. Rev.* **46**, 692 (1934).
20. A. D. Buckingham, M. P. Bogaard, D. A. Dunmur, C. P. Hobbs, and B. J. Orr, "Kerr effect in some simple non-dipolar gases," *Trans. Faraday Soc.* **66**, 1548 (1970).