Nanoradian sensitivity Kerr effect apparatus

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The shot-noise limit to the sensitivity of ellipsometric phase shift measurements with a laser beam power of a few mW is of the order of 10^{-9} rad. An apparatus is described that realizes shot-noise-limited performance for dc Kerr effect birefringence measurements.

INTRODUCTION

Molecular properties such as polarizability anisotropy,[1–3] magnetizability anisotropy,[4,5] quadrupole moment,[6,7] and hyperpolarizability[8,9] may be measured by means of the field-induced birefringence of a sample. In the case of a gas phase sample the induced birefringence is often very small, of the order of a microradian. While previous workers[2–6] have reported the sensitivity of their apparatus as no better than $\pm 0.1\ \mu$rad for a single measurement, the ultimate limit to the sensitivity of a birefringence measurement set by photon counting statistics is much lower than this. The shot-noise limit to the sensitivity for a 1-min measurement employing a 5-mW laser beam is only 1 nrad. In the following we describe an apparatus for making electric-field-induced birefringence (dc Kerr effect) measurements at the shot-noise limit.

I. SHOT-NOISE LIMIT

In principle, the simplest method for determining the retardation ϕ of a birefringent element is to place it between perfect, crossed polarizers with its birefringence axis oriented at 45° to the polarization direction, and to measure the transmitted intensity of a laser beam directed through this sequence of three elements. The transmitted photon flux will be[7,10,11]

$$F = F_0(\phi/2)^2, \tag{1}$$

where

$$F_0 = P_0 \lambda / hc \tag{2}$$

is the photon flux of the incident laser beam of power P_0. The mean number of photons detected during a measurement time interval Δt will be

$$N = FQ\Delta t, \tag{3}$$

where Q is the quantum efficiency of detection. The number of photons actually detected in the interval Δt will be randomly distributed about the mean with standard deviation $\delta N = N^{1/2}$. These statistical fluctuations are called shot noise. The shot-noise limit (SNL) to the sensitivity of a measurement has been reached when the shot noise is equal to the signal: $\delta N = N$. For the simple apparatus just considered, the shot-noise limit to the sensitivity is

$$\phi_{\mathrm{SNL}} = 2(F_0 Q\Delta t)^{-1/2}. \tag{4}$$

For a 5-mW He–Ne laser emitting $F_0 = 1.6\times 10^{16}$ photons per second ($\lambda = 632.8$ nm), assuming $Q = 1$ and $\Delta t = 100$ s, one finds $\phi_{\mathrm{SNL}} = 1.6\times 10^{-9}$ rad.

In practice, stray birefringence of the order of 0.5 mrad is unavoidable, and crossed, imperfect polarizers transmit 10^{-7} or more of the incident light, so that a heterodyne method must be used to measure small retardations.[2,7] Both a large static retardation $\phi^{(0)}$ and a small modulated retardation $\sqrt{2}\phi^{(\omega)} \sin \omega t$ are placed between the crossed polarizers ($\phi^{(\omega)}$ is the root-mean-square amplitude of the modulation). The transmitted flux is now

$$F = F_0(\phi^{(0)2} + 2\sqrt{2}\phi^{(0)}\phi^{(\omega)} \sin \omega t + 2\phi^{(\omega)2} \sin^2 \omega t)/4. \tag{5}$$

Synchronously rectifying F using a lock-in detector tuned to the modulation frequency ω, the time-averaged signal flux will be

$$\overline{F^{(\omega)}} = (\sqrt{2}/\pi)F_0\phi^{(0)}\phi^{(\omega)}. \tag{6}$$

Since $\phi^{(0)} \gg \phi^{(\omega)}$, the noise will be dominated by the fluctuations in the unmodulated flux $F^{(0)} = F_0(\phi^{(0)}/2)^2$. As before, the shot-noise limit has been reached when the noise amplitude is equal to the signal amplitude:

$$(2\sqrt{2}/\pi)(F^{(0)}Q\Delta t)^{1/2} = \overline{F^{(\omega)}}Q\Delta t,$$

where the extra factor $2\sqrt{2}/\pi$ accounts for the effect of lock-in detection on the noise. The shot-noise limit for the heterodyne technique is just

$$\phi^{(\omega)}_{\mathrm{SNL}} = (F_0 Q\Delta t)^{-1/2}, \tag{7}$$

independent of the static birefringence and $2\times$ smaller than the result for the simple dc detection scheme.

II. CONSTRUCTION

Figure 1 shows the apparatus used to make measurements of the dc Kerr effect in gases. The light source is a 5-mW unpolarized He–Ne laser (Spectra-Physics 105-2). The polarizer and analyzer are Glan-laser prism polarizers with extinction ratio lower than 10^{-7} when tested with the 0.8-mm-diam laser beam. Placed between the two polarizers are a liquid CS_2 Kerr cell with 0.1-m-long electrodes and a gas Kerr cell with 0.5-m-long electrodes. The interelectrode gap is 3 mm in both cells (much wider than the laser beam) and the cells are sealed with fused silica windows. Because of the imperfect transmission through the optical elements, P_0 appearing in Eq. (2) is no longer just the incident laser power but is instead the power transmitted through the apparatus

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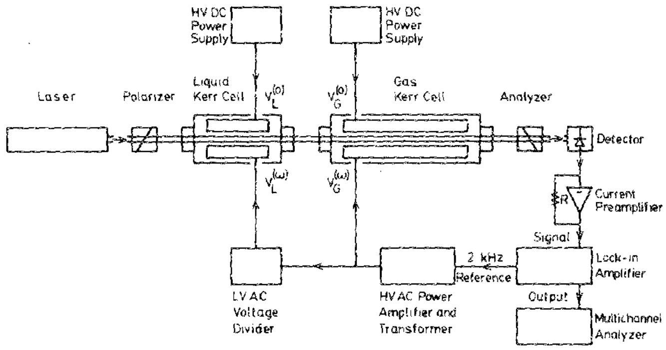

FIG. 1. Schematic diagram of apparatus for measuring the dc Kerr effect in gases. Voltages are measured with respect to the metal case of the gas Kerr cell as ground.

when the analyzer has been reoriented for maximum transmission. For this apparatus $P_0 = 2$ mW.

The static retardation $\phi^{(0)} = K_L V_L^{(0)^2} = 18$ mrad is generated by applying a high dc voltage $V_L^{(0)} = 3$ kV to one electrode of the liquid CS_2 cell (and applying zero dc voltage to the other electrode; electrode voltages are measured with respect to the metal case of the cell as ground). If at the same time a small ac voltage $V_L^{(\omega)} = 50$ mV is applied to the other electrode of the same cell, then a calibration signal $\phi^{(\omega)} = 2K_L V_L^{(0)} \; V_L^{(\omega)} = 600$ nrad is also generated. The Kerr constant $K_L = 2$ mrad/kV^2 of the liquid Kerr cell is itself calibrated by means of a Soleil–Babinet compensator temporarily inserted into the optical path immediately after the liquid cell. The voltages applied to the electrodes of the gas Kerr cell are typically $V_G^{(0)} = 3$ kV and $V_G^{(\omega)} = 500$ V, which generates a signal $\phi^{(\omega)} = 2\,K_G V_G^{(0)} V_G^{(\omega)} = 300$ nrad when the sample is atmospheric air. The modulation frequency is 2 kHz.

The light transmitted by the analyzing polarizer is detected by a small (5.6 mm^2 active area), low-noise silicon photodiode (Ealing 28-4505) operating in photovoltaic mode. Silicon photodiodes have an intrinsically high response over a wide spectral range.[12] This diode has $Q > 0.3$ for $\lambda = 200$–1100 nm, with $Q = 0.8$ at the He–Ne laser wavelength. For comparison, the quantum efficiency of a photomultiplier tube is only 5%–10% at best for this wavelength. This photodiode, operated at zero bias in conjunction with a current-sensitive preamplifier, contributes a negligible noise current spectral density of 3 fA Hz$^{-1/2}$. Finally, the preamplifier output is fed to a lock-in amplifier whose output is sampled and sent to a multichannel analyzer. The MCA accumulates a histogram of the lock-in output voltage, from which one may extract both the mean output and the noise amplitude. Noise measurements are calibrated by the thermal noise of a resistor substituted for the silicon photodiode. The thermal noise current density of a resistor R is

$$\delta i_R = (4kT/R)^{1/2}, \tag{8}$$

independent of frequency.

III. OPERATION

The important noise terms in the apparatus in addition to shot noise are (1) detector–preamplifier noise, (2) liquid Kerr cell birefringence noise, and (3) laser amplitude noise. The noise from all the additional noise sources must be made smaller than the shot noise. For the purpose of comparison, the shot noise is expressed as a noise current density at the detector output

$$\delta i_{SN} = e(QF_0)^{1/2}(\phi^{(0)}/2). \tag{9}$$

The first two noise sources pose no problem. The noise of the preamplifier (EG&G 5002) is just the thermal noise of the feedback resistor R which determines the gain. Up to the first range overload current of $i = 100$ nA the noise current density is $\delta i_R = 13$ fA Hz$^{-1/2}$ ($R = 100$ MΩ), and then up to 10 μA the noise is 130 fA Hz$^{-1/2}$ ($R = 1$ MΩ). The dc input current to the preamplifier is

$$i = eQF_0(\phi^{(0)}/2)^2. \tag{10}$$

Fluctuations of the Kerr birefringence of the liquid CS_2 are strongly dependent on the conductivity of the liquid due to water as an impurity. Dehydrating the CS_2 with 3A molecular sieve increases the resistivity to 1×10^{14} Ω cm and makes the birefringence fluctuations negligible.

Laser amplitude noise is due to electrical, thermal, and mechanical noise in the laser. Denoting the relative amplitude noise spectral density by $\delta P/P_0$, the noise current spectral density at the detector output due to laser amplitude noise is given by

$$\delta i_{LN} = (\delta P/P_0)i, \tag{11}$$

where i is given by Eq. (10). The measured amplitude noise spectrum of the He–Ne laser has a broad minimum between 2 and 10 kHz at the value $\delta P/P_0 = 4\times 10^{-7}$ Hz$^{-1/2}$ (the noise floor is higher than the manufacturer's specification of 1×10^{-7} Hz$^{-1/2}$, probably because of polarization noise in the randomly polarized output). Since $\delta i_{SN} \propto \phi^{(0)}$ while $\delta i_{LN} \propto \phi^{(0)2}$, one can, in principle, satisfy $\delta i_{LN} < \delta i_{SN}$ by reducing $\phi^{(0)}$.

The choice of an operating point is illustrated by Fig. 2(a) where the preamplifier-, laser-, and shot-noise currents have been plotted as functions of the bias retardation $\phi^{(0)}$ (assuming the parameters that describe our apparatus). One sees that there is a wide range, $\phi^{(0)} = 3$–60 mrad, where shot noise dominates. Operating with $\phi^{(0)} = 10$–20 mrad, the measured noise (the width of the lock-in output voltage histogram) agrees closely with the calculated shot noise of 14 nrad Hz$^{-1/2}$. Averaging the output for 1 min gives $\pm$ 3-nrad sensitivity, as determined from the scatter of successive measurements.

IV. LIMITATIONS

Since cw lasers with much higher power are readily available, the question naturally arises as to whether a much lower shot-noise limit could be realized. Figure 2(b) shows the situation for $P_0 = 500$ mW. If the laser-noise amplitude is $\delta P/P_0 = 10^{-7}$ Hz$^{-1/2}$, then the shot-noise limit of 1 nrad Hz$^{-1/2}$ is easily attained using $\phi^{(0)} = 5$–10 mrad. However, shot-noise-limited performance would be difficult or impossible to achieve if $\delta P/P_0 > 10^{-6}$ Hz$^{-1/2}$ because the required $\phi^{(0)} < 1$ mrad would be difficult or impossible to work with. An output power of 0.5 W or more in the visible

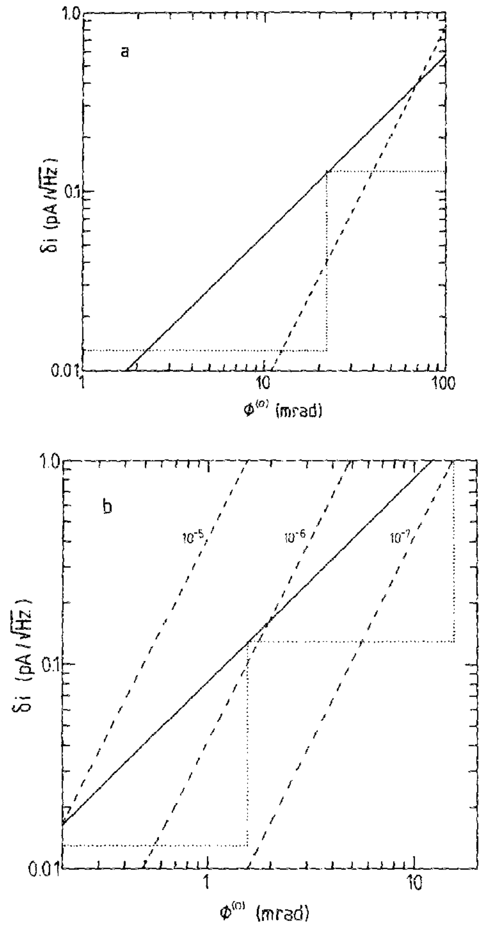

FIG. 2. Variation of the detector output noise current spectral density δi as a function of the bias retardation $\phi^{(0)}$, for shot noise [solid line, see Eq. (9)], laser amplitude noise [dashed line, see Eq. (11)], and preamplifier noise (dotted line). The preamplifier noise curve has the form of a staircase because the gain of the preamplifier must be switched to prevent overload as $\phi^{(0)}$ is increased. Shot-noise-limited operation is obtained for those values of $\phi^{(0)}$ at which the shot-noise curve lies well above the other curves. The parameters describing the apparatus are (a) $P_0 = 2$ mW, $Q = 0.8$, $\lambda = 632.8$ nm, $\delta P/P_0 = 4\times 10^{-7}$ $\mathrm{Hz}^{-1/2}$, and $\phi_{\mathrm{SNL}}^{(\omega)} = 14.0(\Delta t)^{-1/2}$ nrad, and (b) $P_0 = 500$ mW, $Q = 0.8$, $\lambda = 514.5$ nm, $\delta P/P_0 = 10^{-5}$, 10^{-6}, or 10^{-7} $\mathrm{Hz}^{-1/2}$, and $\phi_{\mathrm{SNL}}^{(\omega)} = 0.98(\Delta t)^{-1/2}$ nrad.

may be obtained from an argon-ion laser or a broadband dye laser pumped by an argon-ion laser. The measured amplitude noise for an argon-ion laser (Coherent Innova 90-5, light regulated, 0.5 W at $\lambda = 514.5$ nm) is $\delta P/P_0 = 5\times 10^{-6}$ $\mathrm{Hz}^{-1/2}$ in the range 200 Hz–100 kHz. For a dye laser (Coherent 599, R6G dye, 0.8 W at $\lambda = 590$ nm) the noise is 5×10^{-4} $\mathrm{Hz}^{-1/2}$ at 2 kHz and 5×10^{-5} at 20 kHz. Noise reduction by a factor of 50 would give satisfactory results with the argon-ion laser, but the dye laser would need a noise reduction factor of 1000 or more to reach the shot-noise limit. The required laser amplitude noise reduction might be obtained by means of an electro-optic light regulator. Alternatively, a more sophisticated detector arrangement could be used in which the laser beam rejected by the analyzing polarizer is sampled and used to correct for the amplitude fluctuations of the laser.

For measurements of nanoradian sensitivity to be useful, spurious background signals at the nanoradian level must be controlled or eliminated. There are three main sources of spurious signals: (1) electrical pick up, (2) occultation effects, and (3) window Kerr and stress birefringence effects. Electrical pick up is eliminated by breaking ground loops associated with the ac high voltage (radiative coupling is insignificant at 2 kHz). Floating the power amplifier driven by the lock-in reference output and using a ferrite-core transformer to generate the ac high voltage is effective in eliminating the important ground loops. Occultation refers to the modulation of the laser beam intensity that may arise when dust, lint, or the edge of an aperture moves in response to electrical, acoustical, or mechanical forces originating from the high electrical fields between the electrodes of the gas Kerr cell. A modulation amplitude of 10^{-6} will give a spurious signal of the order of 10^{-9} rad. Such spurious signals are eliminated by attention to cleanliness and by careful collimation of the laser beam.

The final sources of spurious signals are the windows of the gas Kerr cell. The fringing fields from the Kerr cell electrodes will produce a birefringent retardation of 1–10 nrad in the windows even when they are ten gap widths away.[13] This effect is easily eliminated by placing electrical shields around the windows. More difficult to eliminate is the window stress modulation that arises because there are substantial oscillating forces (of order 10^{-2} N) on the electrodes of the gas Kerr cell. The retardation induced in a window of thickness l subjected to a transverse stress σ is $\phi = (2\pi/\lambda)Cl\sigma$, where $C = -3.5\times 10^{-12}$ m^2/N is the stress-optic coefficient of fused silica at $\lambda = 632.8$ nm.[14,15] The mechanical coupling of the electrodes to the gas cell case and the gas cell case to the windows must be very weak to make window stress modulation insignificant since a stress of only 10^{-4} Pa will give a spurious signal of 1 nrad. As an alternative to reducing the stress, one may instead reduce the stress-optic coefficient C of the windows. Two possible materials are yttrium aluminum garnet or YAG($C = -0.52\times 10^{-12}$ m^2/N at $\lambda = 632.8$ nm)[15] and zero-stress-birefringence or Pockel's glass ($C = -0.028\times 10^{-12}$ m^2/N at $\lambda = 632.8$ nm),[7] but neither is readily available. Since spurious signals may only be present when both ac and dc high voltages are applied to the gas Kerr cell, their identification may require an empty cell test. Evacuating the gas Kerr cell to a pressure less than 10 mTorr allows one to apply high voltage without breakdown and reduces the Kerr signal of the gas to the prad level. In this circumstance any observed signal must be spurious.

In summary, apparatus for measuring the dc Kerr effect birefringence of gases has been constructed and a retardation sensitivity of 3 nrad has been demonstrated for a 1-min measurement time. An improvement in the sensitivity to better than 0.1 nrad seems to be feasible.

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