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High-sensitivity measurements of the Kerr constant in gases using a Fabry–Pérot-based ellipsometer

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Abstract. The dc-Kerr constant was measured with high sensitivity in several gases, using a novel ellipsometer, which is based on a high-finesse Fabry–Pérot interferometer. An effective interaction length of 5.26 km was achieved by using a Fabry–Pérot interferometer with intracavity electrodes, yielding a single-pass sensitivity of ≈ 10 nanoradian. The dc-Kerr constants of CO_2, N_2, and O_2 were determined with a high accuracy and with good agreement with previous measurements.

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Electric-field-induced birefringence (dc-Kerr effect) can be used to measure molecular properties, such as the third-harmonic susceptibility and the hyperpolarizability. Kerr-constant measurements may provide information about the energetic bonds of the molecule. However, the induced birefringence in gases is typically very small. In conventional methods, the Kerr-induced phase retardation in the gas sample is in the range of micro-radians or less, therefore highly sensitive and highly accurate ellipsometers are used.

Recent progress in realization of very high-finesse Fabry–Pérot interferometers, which utilize low-loss high-reflectivity mirrors, opens new possibilities for extremely sensitive measurements. These cavities enable extremely long interaction length (up to tens of km) in a table-top setup. Up until now, they were mainly used for detecting weak absorption lines [1–3]. Other measurements of weak effects using Fabry–Pérot cavities include highly sensitive accelerometers [4] and displacement sensors [5]. The long interaction length is also attractive for measurements of small polarization changes. This technique was recently used for measurement of the Cotton–Mouton magneto-optic rotation in gases [6–9].

The Fabry–Pérot cavity is also advantageous for measuring the weak Kerr effect in gases, but to the best of our knowledge, such measurements have not been reported up until now. Measurements of Kerr effect inside a laser cavity were previously reported by Mak et al. [14]. Although demonstrating high sensitivity, the intra-laser-cavity technique cannot be used with compact low-noise monolithic lasers, for example the non-planar ring oscillator [10]. Furthermore, the polarization rotation induced by the Kerr effect must be compensated by another polarization rotator inside the laser cavity. In this paper we present a novel experimental method for external Fabry–Pérot-enhanced Kerr measurements. The effective interaction length was 5.26 km, four orders of magnitude longer than in a typical laboratory setup [11]. This method does not require additional polarization compensation and may be used with any single-longitudinal mode laser. Furthermore, it enables the study of gases with weak Kerr constant in a table-top setup, even with modest electric fields and at atmospheric pressure. The experimental setup is used to measure the Kerr effect of three gases – CO_2, N_2, and O_2.

The theoretical background is given in Sect. 1. The experimental setup, which combines a null-ellipsometer and high-finesse Fabry–Pérot cavity is described in Sect. 2. Measurements of the Kerr constants in several gases are presented in Sect. 3 and finally, Sect. 4 includes a short discussion and summary.

1 Theoretical background

An electric field applied to a centro-symmetric medium, such as gas or liquid, induces a birefringence in the refractive index [12]:

$$n_\perp - n_\parallel = \lambda_p B_0 \langle E \rangle^2 , \tag{1}$$

where $n_\perp$ and $n_\parallel$ are the refractive indices perpendicular and parallel to the electric field E, respectively, λ_p is the probe wavelength and B_0 is the (wavelength-dependent) Kerr constant [13], which is related to the wavelength-independent Kerr constant $K = B_0\lambda_p/n$ [14], and n is the refractive index. The refractive index and birefringence depend on the gas pressure, and B_0 is usually defined at a standard pressure of 1 atm.

A light field travelling through a birefringent medium, with the length L, will accumulate a differential phase shift:

$$\Delta\varphi(t) = 2\pi B_0 L E^2 \quad (2)$$

between the components of the light field which are perpendicular and parallel to the electric field. The wavelength-independent Kerr constant of gases is in the range of 10^{-17}–10^{-15} cm^2/statvolt^2, hence in a typical experimental setup ($L = 10$ cm, $E = 1$ kV/mm, $P = 1$ atm), the phase shift is fairly small, in the range of 5–500 nanoradian.

1.1 Null-ellipsometer

The rotation of the polarization of the optical field, induced by the Kerr effect, requires a sensitive polarimeter. A commonly used experimental polarimeter is the null-ellipsometer [15]: The sample is placed between two crossed linear polarizers. The polarizers are set for a zero-background measurement, i.e. when there is no rotation of polarization in the sample, light passing through the polarizer and sample is completely blocked by the analyzer. When the effect applied on the sample changes the polarization, the output field is no longer zero. In most cases a compensator (usually a quarter-wave plate) is added before or after the sample and is used to correct the impurities and the parasitic polarization effects of all the optical components and the investigated sample (due to the medium, the optical windows of the cell containing the sample, etc.). In our experiment a quarter-wave plate was used after the sample. This polarimeter is often called polarizer-sample-compensator-analyzer [15] null-ellipsometer.

1.2 High-finesse Fabry–Pérot interferometer

A photon entering a Fabry–Pérot cavity, is reflected back and forth between the cavity mirrors, attaining an effective interaction length of [16]:

$$L_{\mathrm{eff}} = \frac{2FL}{\pi}, \quad (3)$$

where L is the cavity length and F is the finesse – given by:

$$F = \frac{\pi\sqrt{R}}{1-R}, \quad (4)$$

where R is the intensity reflectivity of the input and output mirrors. Therefore, for example, using a cavity mirrors with intensity-reflection $R > 99.99\%$, one can achieve a finesse $F > 30\,000$ and the enhancement factor in the interaction length can be $2F/\pi > 20\,000$. Therefore using a high-finesse Fabry–Pérot cavity, the investigation of a phenomena that is linearly dependent in the interaction length can be increased by four orders of magnitude, by placing the sample in the cavity. This method is attractive for measuring very weak effects in a compact table-top setup. The high sensitivity attained by high-finesse Fabry–Pérot cavities is advantageous for measuring the (typically small) phase shift induced by the Kerr effect in gases. A null-ellipsometer can be combined with a Fabry–Pérot cavity by adding crossed polarizers before and after passing through the cavity. Assuming that the linearly polarized input light enters at $\pi/4$ radians with respect to the direction of the dc electric field and that $\Delta\varphi(\ll \pi)$ is the single-pass phase shift induced by the Kerr effect, the ratio between the power transmitted through the output polarizer, P_{out}, and the input power, P_{in}, is

$$\frac{P_{\mathrm{out}}}{P_{\mathrm{in}}} \approx \left(\frac{2F}{\pi}\right)\frac{\Delta\varphi^2}{4}. \quad (5)$$

2 Experimental setup

We have built a polarizer-sample-compensator-analyzer null-ellipsometer [15], see Fig. 1. We have constructed a 0.55-m-long Fabry–Pérot cavity, having two concave high-reflectivity mirrors with a 1-m radius. The cavity finesse was determined by a ring-down measurement [17] to be 17 250. In the cavity we placed two stainless-steel electrodes. The electrodes length was ≈ 47.9 cm and the spacing between them was ≈ 4.55 mm (the spacing is large enough compared to the laser diameter at the end of the electrode, which is 1.77 mm). From (3) and the length of the electrodes, the effective interaction length was $L_{\mathrm{eff}} = 5260$ m. The voltage applied to the electrodes was up to 5 kV. The Fabry–Pérot cavity was placed inside a vacuum chamber, which could be filled with various gases and the pressure was determined with an absolute pressure gauge.

The light source in our experiments was a monolithic single-longitudinal mode cw Nd:YAG laser (Lightwave Electronics model 122) emitting up to 300 mW at 1064 nm.

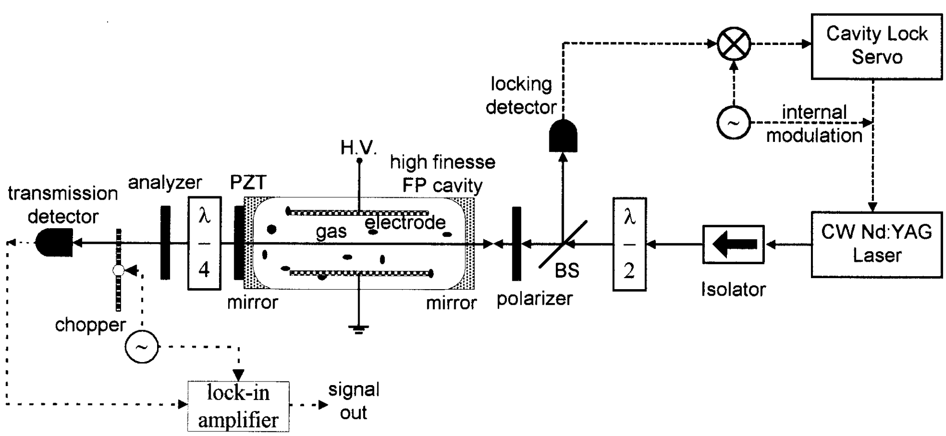

Fig. 1. Experimental setup for measurements of the Kerr effect in gases using a Fabry–Pérot-based ellipsometer. BS – beamsplitter, PZT – piezo electric transducer, H.V. – high voltage

The typical input power reaching the Fabry–Pérot cavity was 90 mW. To eliminate reflections from the cavity back to the laser we used a 40-dB optical isolator. The linear polarization was set by the half-wave plate to be at $\pi/4$ to the electric field induced by the electrodes. The highest Kerr-induced polarization rotation is achieved at this angle. The input and output linear-polarizers ("polarizer" and "analyzer") were of the Glan–Thompson type, with an extinction ratio $< 5 \times 10^{-5}$. The compensator was a zero-order quarter-wave plate. The transmitted power on resonance was $\approx 500\,\mu$W. We chopped the output beam at 1.4 kHz and detected the signal with a lock-in amplifier in order to eliminate background noise and reduce the detection bandwidth to ≈ 1 Hz and therefore to improve the detector sensitivity.

The frequency of the continous-wave laser was locked to the cavity resonance frequency using the Drever–Hall technique [18]. FM sidebands were generated by direct modulation of the piezo transducer attached to the NPRO crystal [10] at 928 kHz. This frequency was chosen owing to its low residual AM noise [19]. The servo was a proportional + integral servo whose parameters were chosen for optimum locking to the cavity [20].

3 Experimental results

We measured the Kerr coefficient for three gases: CO_2, N_2, and O_2 ($>$ 99% purity) at a pressure of 1 atm. The cavity finesse was measured before the beginning of each set of measurements. Several different voltage levels in the range of 0–5 kV were applied to the intracavity electrodes. When no voltage was applied, the leakage power from the ellipsometer was ≈ 2 nW, and this power level was comparable to, or higher than, the variation in transmitted power induced by the Kerr effect. Hence, the leakage power level was measured each time for a period of 60 s, followed by another measurement at high voltage for a period of 60 s. Figure 2 shows a time-trace of measurement with CO_2 at 3 kV.

To calibrate the system we measured the maximum output power, by a $\pi/2$ radians rotation of the analyzer. This was then used to convert the measured power change induced by the Kerr effect to rotation of the polarization direction. For example, the rotation of CO_2 at a voltage of 3 kV (electric field ≈ 660 V/mm) was 28.12 ± 1.4 mrad. Note that this angle is higher by several orders of magnitude with respect to the angles attained by conventional techniques [21]. Furthermore, the corresponding single-pass rotation in this case is only 2.6 microradian. Using (2), the Kerr constant can be calculated to be: $K(\mathrm{CO_2@1\,atm, 300\,K}) = 18.7 \times 10^{-16} \pm 5.92 \times 10^{-16}\,\mathrm{cm^2/statvolt^2} = 2.08 \times 10^{-24} \pm 0.66 \times 10^{-24}\,\mathrm{m^2/V^2}$ – in reasonable agreement with previous results [14]. Despite the high sensitivity of our method, the error range of the results is quite high due to the inaccuracies in the measurements of the electrodes gap, the electrode length, and the high voltage. Those measurements can be highly improved (for example: [21]).

Our setup can be used for making relative measurements between different gases with very high accuracy. In this case, the systematic uncertainties in determining the electrodes gap and length do not affect the measurement. We measured under the same conditions the output signal versus the electrode high voltage for CO_2, N_2, and O_2. This enabled us to determine the ratio between their Kerr coefficients. Figure 3 shows the Kerr-induced phase shift versus the square of the electric field. As expected, a linear dependence is obtained, and the Kerr coefficient can be derived from the slope of the curve. Furthermore, the ratio between the Kerr coefficient of the gases can be calculated with a high accuracy and is given in Table 1. A similar set of measurements were made to derive the Kerr-coefficients ratio of CO_2 and O_2, and is also given in Table 1. The error limits are based on the statistics of our set of measurements.

The dominant noise in our system was the intensity noise of the laser signal transmitted through the Fabry–Pérot cavity. This noise is higher than the laser intrinsic noise, since slight deviations of the laser frequency from the cavity resonance frequency, which are not corrected by the servo will

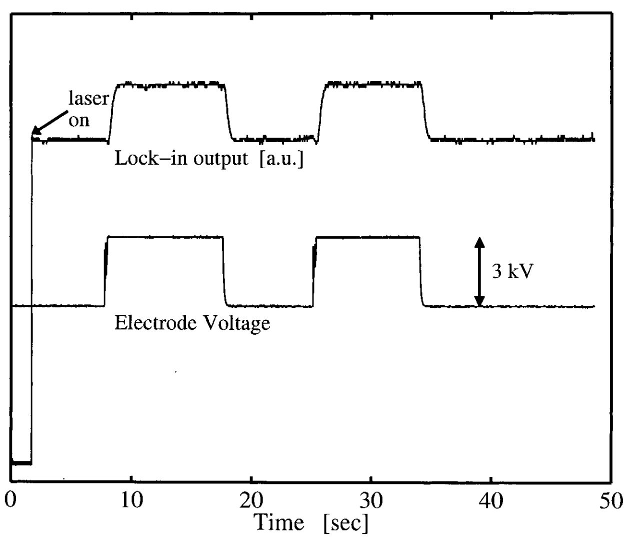

Fig. 2. The output signal of the ellipsometer with and without the high voltage, as a function of the time. The measured gas is CO_2 at a pressure of 1 atmosphere and a voltage of 3 kV applied to the electrodes

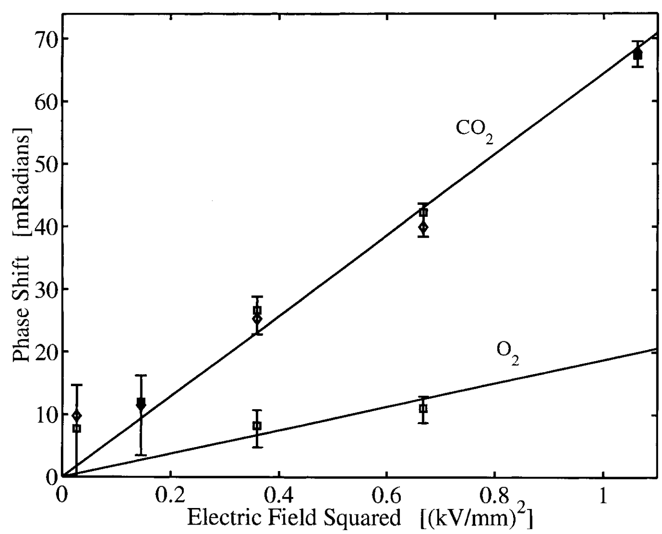

Fig. 3. The Kerr-indcued phase shift as a function of the square of the electric field. Both gases are at $P = 1$ atm

Table 1. The ratio between the Kerr coefficients of several gases

The ratio of the Kerr coefficients $P = 760$ Torr; $T = 300$ K	This work	Ref. [14]	Ref. [24]	Ref. [13]
$K(CO_2)/K(O_2)$	3.16(±1.41)	4(+2.92 − 1.73)	3.33	4
$K(CO_2)/K(N_2)$	7.85(±3.51)	9.33(+7.03 − 4.07)	10.71	5.83

be converted to intensity noise by the Fabry–Pérot cavity. The typical noise level was 0.5% of the average power, for a detection bandwidth of 1 Hz at the chopper modulation frequency of 1.4 kHz.

4 Discussion and summary

Using a novel technique of integrating a null-ellipsometer with a high-finesse Fabry–Pérot cavity, we demonstrated high-sensitivity measurements of the Kerr effect in a series of gases. By inserting the investigated sample in a high-finesse Fabry–Pérot interferometer, an extremely long interaction length (≈ 5 km) was achieved in a table-top setup. The Kerr constants of CO_2, N_2, and O_2 were determined with a high accuracy and with good agreement with previous measurements.

The high sensitivity of the cavity-based ellipsometer is attractive for measurements of gases with low Kerr constant. Furthermore, the experimental method presented here can be used to determine the dependence of the Kerr constant on gas pressure and temperature, and should be attractive for measurements at low pressures, where the sensitivity of the standard experimental methods may not be sufficient.

In our setup the single-pass sensitivity was in range of ≈ 10 nanoradian. A similar sensitivity in a conventional setup can only be achieved near the shot-noise limit [22]. Moreover, with several improvements outlined below, which are possible with current state-of-the-art technology, much higher sensitivity can be attained using the Fabry–Pérot-based ellipsometer.

Further improvements can be achieved using a cavity with a higher finesse. We have previously achieved with the same mirrors a finesse $> 86\,000$ and interaction length > 30 km [1], however owing to increase of the mirrors' loss level, the finesse has degraded to a level of 17 250. Whereas the highest electric field we have used in this work was ≈ 1 kV/mm and the gas pressure was 1 atmosphere, higher fields and gas pressure will increase the Kerr-induced phase shift and thus the measurement sensitivity.

Advanced cavity-based modulation and demodulation techniques can further improve the system sensitivity. For example, the laser frequency can be modulated at a multiple of the cavity free spectral range (≈ 270 MHz in our case) and the transmitted signal will be demodulated at the same frequency [3]. This method may enable near-shot-noise-limited detection, which, for 1 nW power on the detector and a cavity finesse of 100 000, would result in sensitivity of ≈ 10 picoradians for a 1 Hz detection bandwidth.

Our system uses a continuous-wave source, hence a servo-controller must be used for locking the laser frequency to the cavity resonant frequency. The system can be simplified (but at the expense of sensitivity) if a pulsed laser, whose pulse length is shorter than the cavity round-trip time in the cavity, is used as the light source. A similar approach is commonly used now in cavity- based absorption spectroscopy [7, 23].

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