Measurement of the thermal contribution to the nonlinear refractive index of air at 1064 nm

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The thermal contribution to the nonlinear refractive index of air at 1.064 μm was measured with a high-finesse Fabry–Perot cavity and a 500-mW cw laser beam. At room temperature and pressure, the nonlinear refractive-index coefficient of air was found to be $n_2^{(\mathrm{th})} = (-1.9 \pm 0.2) \times 10^{-14}$ cm^2/W for a beam waist radius of 0.23 mm and was found to be independent of the relative humidity. The thermal nonlinearities of N_2, O_2, and CO_2 were also measured, and it was found that the dominant contribution to air is its O_2 content.

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Thermal nonlinearities[1–4] are commonly much larger than the electronic nonlinearities of the same material, often by several orders of magnitude. However, since thermal time constants are also orders of magnitude longer than those for their electronic counterparts, the use of cw or long-pulse lasers is generally necessary for the study of such thermal effects. The intensity that is achievable with such lasers thus becomes the limiting factor in exploring these nonlinear optical processes.

High-finesse Fabry–Perot cavities[5] can be used to study nonlinear optical processes with cw lasers of only modest power. A nonlinear medium placed inside the cavity experiences an intensity B times larger than that of the incident laser beam, where the cavity buildup factor can readily be found to be

$$B = F\sqrt{T}/\pi\,, \tag{1}$$

where F is the finesse of the cavity and T is its overall transmittance.

We have made use of this enhancement of the optical intensity to perform accurate measurements of the nonlinear refractive indices of several gases. Our procedure is to fill the cavity with the gas being studied, measure the change in the resonance frequency of the cavity as a function of optical input power, and from this deduce the power-dependent change in the refractive index of the gas. In particular, the frequency shift $\Delta\nu$ of the cavity resonance can be expressed as

$$\Delta\nu = -\nu\Delta n/n \approx -\nu\Delta n\,, \tag{2}$$

where ν is the frequency of the light, n is the nominal refractive index of the gas, and Δn is the induced change in this quantity. This frequency shift is measured by means of locking the laser frequency to that of the cavity resonance and then measuring the laser frequency through use of an additional, lower-finesse, scanning Fabry–Perot cavity.

The measured change in refractive index Δn can be interpreted as a thermally induced change through use of the relation

$$\Delta n = (\mathrm{d}n/\mathrm{d}T)\Delta T\,, \tag{3}$$

where for an ideal gas $(\mathrm{d}n/\mathrm{d}T) = -(n-1)/T$. Equation (3) and the expression for $(\mathrm{d}n/\mathrm{d}T)$ show that the thermal nonlinearity of a gas must be negative. The temperature increase induced by the presence of the laser can be predicted by solution in steady state of the equation of heat transport, leading to the result that[6]

$$\Delta T = (\alpha P/\pi\kappa)[\ln(R/\omega_0) + 0.635]\,, \tag{4}$$

where α is the linear absorption coefficient of the gas, κ is its thermal conductivity, ω_0 is the laser beam radius, and R is the radius of the thermal enclosure surrounding the laser beam. For the gases that we have studied, α is a very small quantity (of the order of 10^{-8} cm^{-1} or less) and is believed to be due to overtones and combination tones of molecular vibrations. Since the change in refractive index is seen to depend on the total power (rather than on the intensity) of the laser beam, thermal nonlinearities of this sort can best be described by the coefficient n_P defined by the relation $\Delta n = 2n_P P$ rather than by the usual relation $\Delta n = 2n_2 I$, where the factors of 2 arise since both the forward and the backward waves in the cavity contribute to the heating process. However, for a fixed geometry an effective value of n_2 can be introduced by means of the relation $n_2^{(\mathrm{th})} = n_P/\pi\omega_0{}^2$. Note also that through use of Eqs. (2)–(4) it is possible to determine the value of the absorption coefficient α on the basis of measured values of $\Delta\nu$.

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Our experimental setup is shown in Fig. 1. The high-finesse cavity (HFC, which contains the gas under investigation) is constructed from 1-m radius-of-curvature mirrors spaced by 10 cm. It has a finesse of 12,000 and a transmission of 12%, giving a buildup factor B of 1300. The cavity mirrors are rigidly mounted to a Zerodur spacer inside a metal cylinder of radius $R = 1.27$ cm, which we have suspended in a vacuum chamber to minimize noise-pickup effects. A 500-mW cw Nd:YAG nonplanar ring oscillator laser[7] (NPRO) is mode matched and frequency locked to the high-finesse cavity. A Faraday isolator (FI) is used to protect the laser from backreflections. A half-wave plate (HWP) is used in conjunction with a polarizing beam splitter (PBS) to control the power that is incident on the cavity. Frequency locking is achieved with a standard Pound–Drever–Hall feedback system.[8,9] Weak sidebands at 40 MHz are placed on the beam with an electro-optic modulator (EOM). When the fundamental frequency is on resonance with the cavity, the sidebands are reflected because of the narrow bandwidth of the cavity. The reflected light is directed by the polarizing beam splitter to detector D1, and the electrical output signal of the detector is combined with the 40-MHz modulation source by use of a standard rf mixer (X). The low-frequency components of the mixer output provide the error signal, which is fed back to the fast and slow frequency controls on the laser, which keeps the laser frequency locked to the cavity resonance. The light transmitted through the high-finesse cavity falls onto a low-finesse cavity (LFC; $F = 1000$), which is scanned continously by means of piezoelectric transducers to monitor the laser frequency.

Owing to practical constraints from the locking and detection systems, the input power could not be reduced completely to zero but instead was varied over the range 240 to 520 mW. This procedure produces a variation of 360 W in the power inside the cavity. Typical data are shown in Fig. 2, which illustrates the shift of the resonance peak when the input power is varied with the cavity filled with CO_2. The peak on the left-hand side corresponds to low incident power, and the peak on the right-hand side is the shifted resonance obtained at higher incident power.

From data such as those shown in Fig. 2, the nonlinear and absorption coefficients are found with the procedure described above. Since the absorption coefficient and dn/dT are each proportional to the air pressure, the measured frequency shift $\Delta\nu$ is expected to be proportional to the square of the pressure. To minimize experimental error we took the data at pressures of 0.25, 0.5, 0.75, and 1 atm and fitted them to a quadratic curve, as shown in Fig. 3. We attribute the small shift at zero pressure to effects that are due to heating of the mirrors.

From the data fit shown in Fig. 3, a shift of 2.3 MHz is found for 1 atm of air at room temperature. From this result we find a value of the thermal nonlinear coefficient of air of $n_P = -1.1 \times 10^{-11}\ \mathrm{W}^{-1}$ or of $n_2 = -1.9 \times 10^{-14}\ \mathrm{cm}^2/\mathrm{W}$ for our beam waist radius of 0.23 mm. This latter result should be compared with the value $n_2 = 5 \times 10^{-19}\ \mathrm{cm}^2/\mathrm{W}$ that was measured earlier[10] with a short-pulse laser, where thermal effects did not contribute. We thus conclude that the values measured in the present experiment are dominated by thermal effects. For these data, the change in refractive index is 8×10^{-9} and the change in temperature is 8×10^{-3} K. The absorption coefficient of air can also be extracted from these

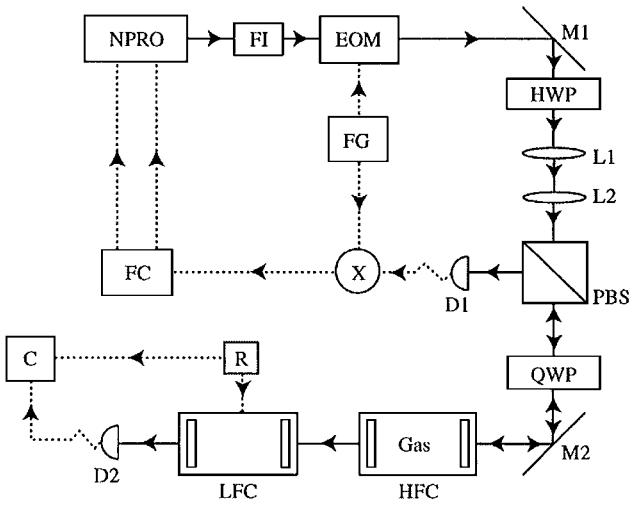

Fig. 1. Schematic of the experimental setup. Optical paths are shown as solid lines, and electrical paths are dotted lines. FC, feedback controls; FG, 40-MHz function generator; R, ramp generator driving the piezoelectric transducers; C, computer for data capture. M1, M2, mirrors; L1, L2, lenses; QWP, quarter-wave plate; D2, detector. See text for other definitions.

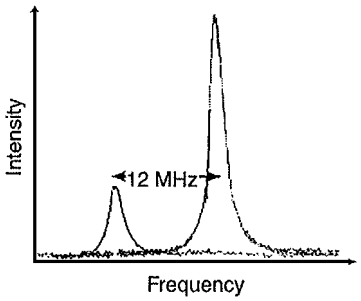

Fig. 2. Traces showing the shift of the cavity resonance frequency between (left) low power and (right) high power.

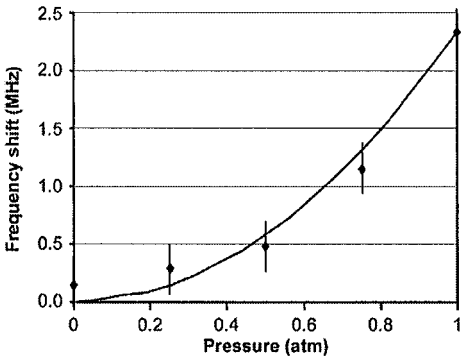

Fig. 3. Shift of the cavity resonance frequency as a function of the pressure of air within the cavity, for a difference in circulating laser power of 360 W.

data. Using the value[11] $\kappa = 0.25$ mW/cm K, we find $\alpha = (3.8 \pm 0.4) \times 10^{-9}$ cm^{-1}. This result is within the range of expected values.[12]

We have also measured the thermal nonlinear coefficients of N_2, O_2, and CO_2 and found, respectively, at 1-atm pressure,

$$n_P(N_2) = (-1.9 \pm 0.6) \times 10^{-12}\ \mathrm{W}^{-1},$$
$$n_2(N_2) = (-3.3 \pm 1.0) \times 10^{-15}\ \mathrm{cm}^2/\mathrm{W},$$

$$n_P(O_2) = (-9.6 \pm 0.9) \times 10^{-11}\ \mathrm{W}^{-1},$$
$$n_2(O_2) = (-1.7 \pm 0.2) \times 10^{-13}\ \mathrm{cm}^2/\mathrm{W},$$

$$n_P(CO_2) = (-3.8 \pm 0.4) \times 10^{-11}\ \mathrm{W}^{-1},$$
$$n_2(CO_2) = (-6.6 \pm 0.7) \times 10^{-14}\ \mathrm{cm}^2/\mathrm{W},$$

where as before the values of n_2 are referenced to a beam waist radius of 0.23 am. The corresponding absorption coefficients are found to be

$$\alpha(N_2) = (6.7 \pm 2.0) \times 10^{-10}\ \mathrm{cm}^{-1},$$

$$\alpha(O_2) = (4.2 \pm 0.4) \times 10^{-8}\ \mathrm{cm}^{-1},$$

$$\alpha(CO_2) = (5.7 \pm 0.6) \times 10^{-9}\ \mathrm{cm}^{-1}.$$

Based on these data and the relative proportions of the constituents in air, O_2 is seen to be the dominant contribution to the nonlinear refractive index of air.

In addition, we measured n_2 as a function of the relative humidity of the air. The results indicate that $\Delta\nu$ and consequently n_2 are independent of the relative humidity.[13] According to our model this implies that the absorption coefficient is also independent of relative humidity, contrary to our initial expectation that water vapor would absorb appreciably at this wavelength.

It is interesting to consider the accumulated phase shift experienced by a beam of light in interacting with a high-finesse cavity. Recall first that under single-pass propagation through a distance L of nonlinear material the nonlinear phase shift acquired by a beam of intensity I is given by $\phi_{\mathrm{NL}} = n_2(\omega/c)IL$. When such an interaction occurs within a high-finesse cavity, the intensity I and the effective interaction path L are each increased by an amount proportional to the finesse F of the cavity, leading to an F-squared enhancement of the nonlinear optical response.[14] For our cavity, this enhancement is of the order of 10^6. Thus, although the measurement of thermal nonlinearities of this magnitude would be impractical with common techniques such as a Z scan, because of limitations on laser power the cavity enhancement makes the measurements straightforward. This fact also suggests that gas-filled, high-finesse Fabry–Perot cavities provide a convenient configuration in which to study nonlinear dynamical and transverse[15] effects.

In conclusion, we have measured the thermal nonlinear optical response of air and other gases, using a high-finesse cavity. For air, we measured the value $n_2 = -1.9 \times 10^{-14}$ cm^2/W and found it to be independent of the relative humidity and dominated by the effects of O_2. More generally, we have shown that the use of high-finesse Fabry–Perot cavities allows the study of nonlinear effects with low-power lasers.

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