RECORDING OPTO-ACOUSTIC SPECTRA WITH AN ACOUSTO-OPTIC DETECTION SYSTEM

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A new device, called an "optophone", is described for the detection of weak transitions in gas samples, based on the deflection of a He–Ne laser beam from a thin pellicle in contact with the gas. A comparison of an optophone to a microphone shows comparable sensitivities for the 5–0 O–H overtone of CH_3OH.

1. Introduction

An essential feature of opto-acoustic spectroscopy has been the use of a microphone detection system [1]. This responds to pressure waves generated when a gas sample is irradiated intermittently by a frequency scanned (laser) light source. The chief virtue of this method is its high sensitivity, which allows weak optical transitions to be detected without the need for extremely long path lengths [2–7]. However, in certain cases, the contact of the listening device (microphone, piezoelectric transducer) with the sample may have undesirable consequences. One example is our recent study on the unimolecular decomposition of *t*-butylhydroperoxide by O–H overtone excitation in which metal-catalyzed surface decomposition was a serious interference [8,9]. Another example is our so-far-unsuccessful study of the overtone spectrum of HT in which the radioactive decay of the gas sample destroyed the microphone [10]. To overcome these problems, we have developed an alternative detection system based on the deflection of a light beam from a thin inert membrane in contact with the gas sample. We call this device an optophone.

Instruments for transforming an acoustic signal into an optical signal have a long history, beginning perhaps with experiments by Lord Rayleigh [11] who suspended a mirror inside a brass tube by means of a silk fiber. The first modern application of an optophone appears to be the study of Choi and Diebold [12] who examined with an iris–photodiode combination the deflection of a He–Ne laser beam by a reflecting diaphragm mounted as part of the wall of a Helmholtz resonator. In this way they observed the absorption of a 1 W CO_2 laser beam incident upon SF_6 gas diluted in N_2 (1 atm total pressure) inside the resonator. As pointed out by Choi and Diebold, the advantages of this acousto-optic detector are· (1) fast time response; (2) high sensitivity inherent in the use of a long lever arm as part of the optical detection system; and (3) resistance to chemical attack. However, because of fluctuations in the He–Ne laser beam intensity and because the reflecting diaphragm was exposed to the ambient surroundings, Choi and Diebold resorted to a complicated double-modulation technique.

Our acousto-optic detector differs from that of Choi and Diebold in two major respects. First, a thin nitrocellulose membrane having a suitable dielectric coating is placed inside the opto-acoustic cell between two chambers at equal pressure, one of which is exposed to the modelated output of a tunable dye laser. Second, we use a position sensing detector to measure the deflection of a low-power He–Ne laser beam from the pellicle. A direct comparison between the vibrational overtone spectra recorded with an electret microphone and with our optophone is made. We find that both detectors give nearly the same signal-to-noise ratio. This suggests that an optophone may have widespread applications in those situations where the use of a microphone is not practical.

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2. Experimental

Fig. 1 presents a schematic diagram of the experimental apparatus. An ion laser beam, which is amplitude modulated at 500–600 Hz by a mechanical chopper (PAR 192), pumps a home-built cw standing wave dye laser. The dye laser wavelength is tuned by a two-plate birefringent filter with a resolution of approximately 2 cm^{-1}. An opto-acoustic cell equipped with both a pellicle beam splitter (Oriel 3743, R/T = 50/50) and an electret microphone (Knowles BT-1759) is placed inside the cavity of the dye laser. The output of a 0.5 mW He–Ne laser (Spectra-Physics 155) is reflected from the pellicle beam splitter, and mildly focused (f = 50 cm) to the center of a position sensing detector (Silicon Detector Corporation SD-113-24-21-021). The distance of the light path between the beam splitter and the detector is about 1.6 m. In order to use this "lever arm" to advantage, the laser beam must not strike the pellicle exactly on center.

The position sensing detector is essentially composed of two identical photodiodes separated by 0.1 mm. The outputs of the two segments are fed individually into a common mode rejection circuit, also shown in fig. 1. The output of the rejection circuit serves as the input to a lock-in amplifier (PAR 186A). The position sensing detector is capable of sensing position shifts in the light beam at the detector as small as 6 × 10^{-3} cm under our operating conditions according to the manufacturer.

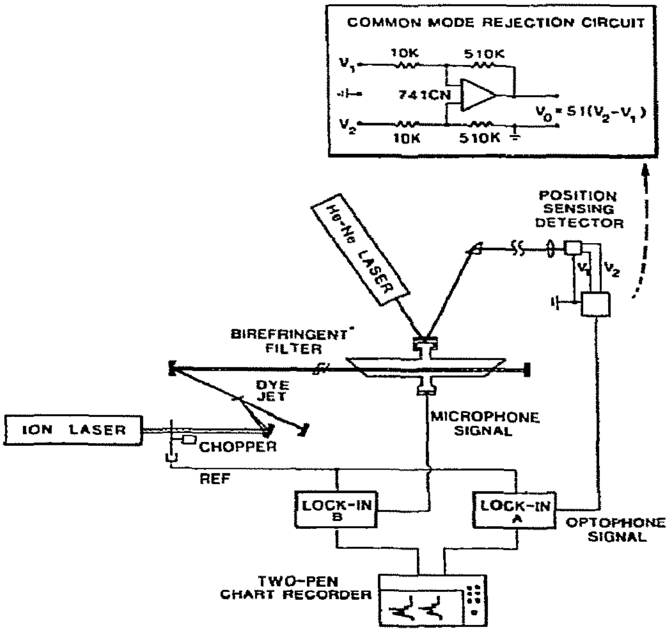

Fig. 1. Schematic diagram of the experimental apparatus showing the simultaneous use of a microphone and an optophone to record overtone spectra.

The microphone on the other side of the cell is powered by a 1.5 V battery and its output is fed into the other lock-in amplifier (PAR HR-8). Signals from the two lock-in amplifiers (with the chopping frequency as the reference input for both) are displayed by a two-pen stripchart recorder (Linear).

If normalized signals are desired, then the outputs from both lock-in amplifiers must be ratioed to the intracavity laser power. A quantity proportional to the intracavity laser power can be obtained by measuring the intensity of a reflection from one of the Brewster's angles with a power meter. For the purpose of comparing the sensitivities of the two detection systems, however, normalization of the signals were not carried out.

To make a fair comparison of the two methods, the cell is engineered in such a manner that the pellicle beam splitter and the electret microphone are placed in virtually identical environments (see fig. 2). The pellicle beam splitter is sealed inside the cell and the pressures on both sides of the beam splitter are equalized. This design minimizes ambient noise and also

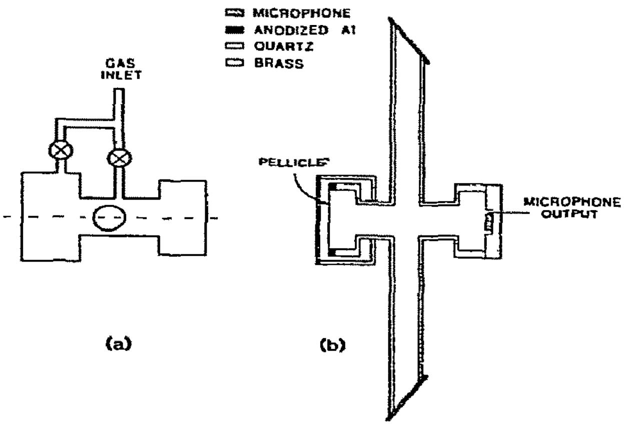

Fig. 2. Opto-acoustic cell having both a microphone and a pellicle beam splitter: (a) side view and (b) cross sectional view along the dot-dashed line in (a).

makes possible the study of gas pressures below or above atmospheric pressure. Since the pellicle beam splitter is only ≈7 μm thick, large pressure differentials across the beam splitter easily break the membrane, and must be avoided.

3. Results and discussion

Fig. 3 presents the power-normalized spectrum of the 5–0 O–H stretching overtone of methanol (20 Torr) taken with the optophone. Comparison of this spectrum with that published by Jasinski [13] using a microphone detection system shows it to be essentially identical. There are distinguishable features in this overtone region. Some of these features have been assigned by Jasinski [13] to P, Q, and R band contours.

The same overtone region as in fig. 3 was recorded simultaneously without power normalization by an optophone and a microphone using the apparatus shown in figs. 1 and 2. These spectra are compared in fig. 4 where the upper trace is from the microphone and the lower trace from the optophone. The spectra taken at 15 Torr (fig. 4a) with the two detection techniques appear identical. At 5 Torr (fig. 4b) the signal-to-noise ratio of the two spectra are comparable.

The major source of noise is believed to be ambient vibrations which can be substantially reduced by performing the experiment on a vibration isolation table. A cell with high acoustic resonance frequency can also be used to reduce the relative noise level due to mechanical vibrations. No effort has been made in the present design to match the response of either detector to the acoustic characteristics of the cell.

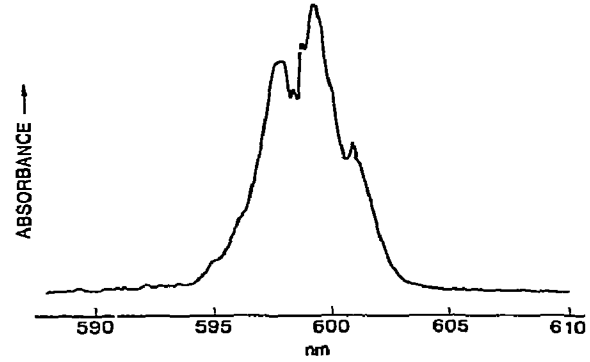

Fig. 3. The power-normalized 5–0 O–H stretching overtone in methanol (20 Torr) recorded using an optophone. The dye laser beam is modulated at 575 Hz.

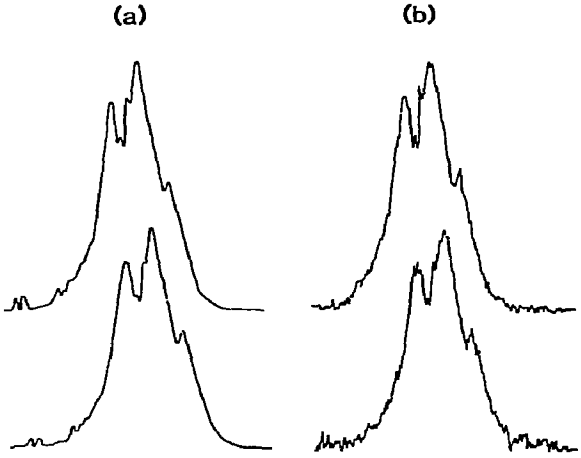

Fig. 4. Comparison of microphone (upper trace) and optophone (lower trace) outputs for the 5–0 O–H stretching overtone in CH_3OH at (a) 15 Torr and (b) 5 Torr. Both spectra have not been normalized for the variation of intracavity laser power with wavelength.

The present study demonstrates than an acousto-optic detector (optophone) employing a dielectric-coated pellicle (beam splitter), a low-power He–Ne laser, and a position sensing detector can provide approximately the same sensitivity as an electret microphone detector in opto-acoustic experiments. Further improvements in the sensitivity of the optophone are easy to imagine. For example, the pellicle might form one surface of an optical interferometer. Nevertheless, the present optophone detection system can already be used to advantage in situations involving highly corrosive or radioactive samples for which the use of a microphone is problematic. It is possible to avoid some of the difficulties associated with the exposure of metal surfaces of the microphone to highly corrosive or reactive gases by either coating the surface [8,9] or by replacing the microphone diaphragm by a quartz plate silvered on the side facing the back plate of the microphone [14]. Other detection techniques that avoid contact with the medium involve the deflection of a probe laser beam by the change in the refractive index caused by absorption of the pump laser beam (which may be the same as the probe beam) [15–17]. However, the simplicity, cost, ease of use, and availability of components of the optophone may make it the detection method of choice in many practical situations.

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