Reprinted from THE REVIEW OF SCIENTIFIC INSTRUMENTS, Vol. 41, No. 2, 226–227, February 1970
Printed in U. S. A.

A New LEPR Spectrometer*

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(Received 17 July 1969; and in final form, 26 August 1969)

A new spectrometer for conducting laser electron paramagnetic resonance experiments on gases is described. This spectrometer yields at least an order of magnitude better sensitivity than the first spectrometer used in this type of investigation.

THE first observation of laser electron paramagnetic resonance (LEPR) absorption in a gas at the 891 GHz HCN laser frequency was reported[1] recently. The spectrometer used in that experiment consisted of an HCN laser whose output was focused into a 10 cm long, 1.7 cm diam Fabry–Perot (F–P) interferometer which contained the gas. The F–P interferometer had a 0.75 mm diam coupling hole in each end to facilitate its use as a transmission cavity. Conventional field modulation and phase sensitive detection apparatus completed the spectrometer. Since the first experiment, the technique has been improved and the signal-to-noise (S/N) ratio increased over an order of magnitude.

The new technique consists of making the sample cavity an integral part of the HCN laser. A sketch of the experimental arrangement is shown in Fig. 1. The sample cavity portion of the laser consists of a 50 mm quartz tube. The diameter was dictated by the available gap between the magnet pole faces and quartz was chosen because of its low thermal expansion coefficient. One end is terminated by a gold coated 50 mm diam flat mirror with a 1 mm exit hole located at the center to monitor the laser power. The other end of the sample cavity consists of a polyethylene membrane mounted at the Brewster angle which serves not only to separate the gas under investigation, but also to establish the polarization. The mirror arrangement is completed by a 75 mm diam 1.65 m focal length mirror at the other end of the 2.3 m system which contains a 1.5 m discharge. The laser discharge cathode was placed 65 cm from the center of the magnet to minimize the magnetic field interaction with the laser plasma. The curved mirror is mounted on a piezoelectric driver which is positioned axially by a micrometer drive. The driver is modulated at 30 Hz to permit continuous monitoring of the output power. The spacing between the mirrors is held nearly constant by Invar rods.

The sample cavity attachment shown in Fig. 2 is attached by three equally spaced bolts to a Lucite block terminating the 7.6 cm diam section of the laser. Three 90° slots centered 120° apart facilitate rotation of the Brewster window and thus permit selection of the desired polarization relative to the external magnetic field. The elliptical O-ring channel in the Brewster angle block is easily made by cutting a brass block and two concentric close-fitting tubes at the Brewster angle, then sliding the intermediate tube back and soldering it in place at an appropriate distance to accommodate the O-ring. The polyethylene membrane is held in place during assembly by two-sided adhesive tape.

The new spectrometer has several distinct advantages compared to the earlier version: First, the relative drift between the resonant frequencies of the F–P interferometer and the laser and consequent reduction of the absorption signal have been eliminated. Second, it is possible to select the desired polarization of radiation inside the sample cavity. The relative intensities of $\Delta M=0$ and $\Delta M=\pm 1$

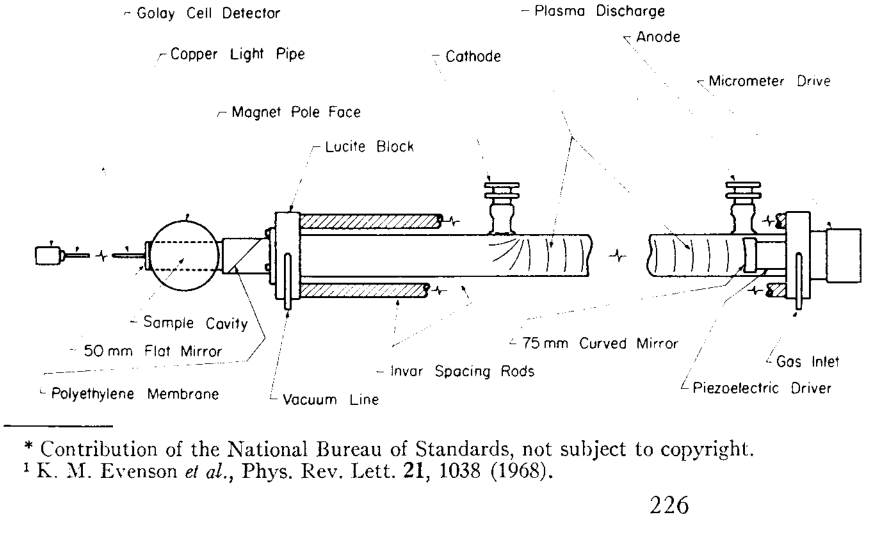

FIG. 1. Sketch of the improved experimental setup for a laser electron paramagnetic resonance experiment.

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[1] K. M. Evenson *et al.*, Phys. Rev. Lett. 21, 1038 (1968).

transitions can be measured by adjusting the beam splitter at 45° from the direction of H_0. Third, with a piezoelectric modulator driving one mirror, it is no longer necessary to use the mechanical chopper to maintain optimum laser output. Last, the S/N ratio is improved by more than an order of magnitude as is evident in Fig. 3. The effective path length (EPL) is given by [2]

$$\mathrm{EPL} = LF\eta/\pi,$$

where L is the length, F the finesse, and η the filling factor. For the old system, the EPL, based on a measured value of the finesse and unity filling factor, is 260 cm. For the new system a calculation of the finesse and a filling factor of 0.06 yield an EPL of 490 cm; thus the improved effective path length does not wholly account for the increase in S/N ratio. Since the ratio of fractional absorption to equivalent path length was the same for each spectrometer, we infer that the laser was probably operating in the linear portion of its gain curve so that the presence of a lasing medium within the combined sample–laser F–P cavity did not produce any severe nonlinear effects upon the oxygen absorption signal. The total laser power transmitted through the external F–P used in the previous experiment produced a signal of about 200 mV on the Golay detector, while the power from the internal cavity system corresponded to about 5 V. With the external F–P the maximum oxygen absorption was about 1/2000 and with the new spectrometer about 1/1000 of the power of the Golay detector. In addition to the elimination of the aforementioned relative drift problem, the greater over-all sensitivity in the new spectrometer results partially from the increased effective path length and also from a higher incident power on the Golay cell so that the Golay detector noise is less effective in degrading the signal.

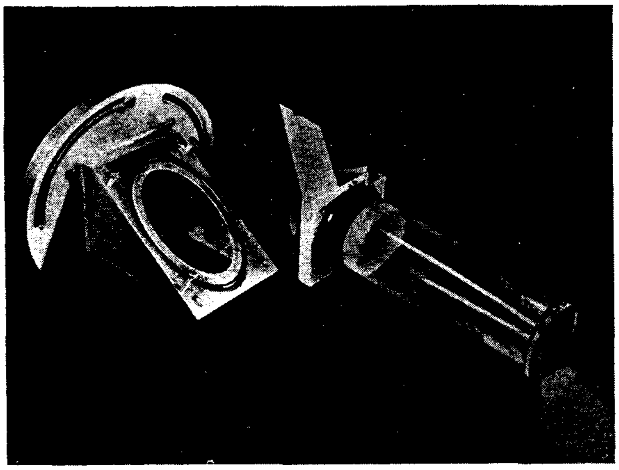

FIG. 2. Sample cavity attachment is shown disassembled for illustrative purposes. The quartz tube is sealed to the laser mirror and upper portion of Brewster angle block by black sealing wax. A single rubber O-ring in the lower left portion of the block presses the polyethylene membrane against the upper portion and provides an adequate vacuum seal in both chambers. The pipe fitting on top of the sample cavity is connected to the laser proper to avoid membrane rupture on evacuating the system.

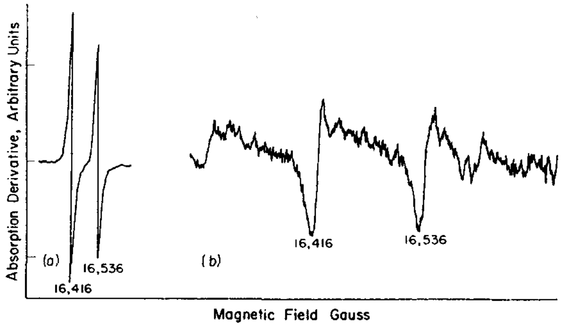

FIG. 3. (a) Trace of spectrometer output of the new system indicating S/N ratios of about 70 and 60 for transitions at 1.6416 and 1.6563 T. The larger oxygen absorption signal is 3 mV peak-to-peak and the sweep rate is 0.025 T/min with a 1 sec time constant on the output. (b) Same two transitions using old system, with much poorer S/N ratio. Peak-to-peak amplitude of the larger signal is about 175 μV and the sweep rate is 0.01 T/min with a 3 sec time constant. Also shown are other transitions due to mixed polarization of radiation inside sample cavity.

We wish to thank G. T. McNice for theoretical calculation of finesse for the 2.3 m F–P, R. C. Mockler for helpful discussions, and V. Derr for useful comments.

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[2] E. P. Valkenburg and V. E. Derr. Proc. IEEE 54, 493 (1960).