

Computer control of broadly tunable lasers: conversion of a color center laser into a high resolution laser spectrometer

J. V. V. Kasper, C. R. Pollock, R. F. Curl, Jr., and F. K. Tittel

The conversion of a commercial color center laser (Burleigh FCL-20) into a computer-controlled high resolution laser spectrometer is described. The techniques and methods developed are generally useful for the control of any broadly tunable cw laser. Superposition of several kinds of computer-controlled feedback upon a table look-up (feed-forward) procedure is used for each of the control elements. This feedback removes the coupling between the tuning elements and eliminates the effects of the nonideal properties of the control elements such as PZT hysteresis and creep. The resulting spectrometer is capable of acquiring high resolution (instrumentally limited by free running laser jitter to ~ 3 MHz) spectra which can be precisely measured (~ 0.0003 cm^{-1}). The spectrometer has routinely made continuous single-mode scans of 30,000 points separated by 10 MHz covering 10 cm^{-1} .

I. Introduction

Molecular and atomic spectroscopy applications of dye and color center lasers require accurate and convenient means of controlling and scanning the laser output wavelength. Wavelength tuning of these lasers is made difficult by the number of dispersive elements which must be adjusted simultaneously in order to obtain continuous scanning. A number of techniques have been developed which automatically track several of the tuning elements together to achieve a limited continuous tuning range. Examples of this approach are the Spectra-Physics model 380/580 and Coherent model CR-599/699 unstabilized and stabilized cw dye lasers which achieve continuous scans exceeding 30 GHz by locking the etalon to a cavity mode and then scanning the cavity length of the laser and reference cavity with tilting Brewster oriented optical flats. Disadvantages of this technique are that the tuning range is limited to relatively short distances due to the free spectral range (FSR) of the etalon and the limited angular tuning range of the Brewster scanning plates. Also, because frequency tuning relies totally on a feedback signal,

large frequency excursions can occur whenever the servo loop (even briefly) loses lock. The tuning range of these systems could be extended by resetting the etalon and cavity length in a manner which adequately simulates continuous frequency scans; however, this task is generally too complex with analog control.

An excellent way to overcome these problems is to employ a minicomputer to adjust all tuning elements simultaneously and to reset the etalon and cavity lengths in modulo steps of one FSR. In addition to extending the laser tuning range, computer control simultaneously provides access to computer data acquisition and makes possible applications previously considered unfeasible. Some such applications include on-line processing of spectral information for rapid trace analysis and spectroscopy of short-lived or weakly absorbing species where low signal levels require data averaging.¹ Several groups, including the authors, have applied some form of computer control to tunable lasers in recent years. Examples include cw and pulsed dye laser spectrometers,²⁻⁶ optical parametric oscillators,⁷ difference frequency generator,⁸ and diode lasers.⁹

In this paper, recently developed comprehensive software and hardware techniques for a computer-controlled tunable single frequency color center laser operating in the 2.3 – 3.3 - μm region are described. The present work is closely related to a previous paper¹⁰ on a computer-controlled color center laser and builds on that work. New to this work is the introduction of automatic and semiautomatic calibration methods and the various kinds of feedback necessary to provide long range continuous scans. The techniques used for control of long range continuous scanning, for automatic frequency calibration of each tuning element, and for

J. V. V. Kasper is with University of California, Chemistry Department, Los Angeles, California 90024; when this work was done C. R. Pollock was with Rice University and is now with U.S. National Bureau of Standards, Boulder, Colorado 80303; the other authors are with Rice University, Houston, Texas 77001.

Received 29 July 1981.

0003-6935/82/020236-12\$01.00/0.

© 1982 Optical Society of America.

automatic acquisition and storage of data have been detailed. This laser is capable of long range scans ($>10 \text{ cm}^{-1}$) with a resolution of 1 MHz, automatic frequency and wavelength calibration, long term drift stabilization, and computer-aided data acquisition. Although the present discussion is limited to the color center laser, many of the same considerations would also be involved in the computer control of other broadly tunable lasers with similar general characteristics. In fact, many of the concepts and algorithms herein described were developed for a computer-controlled tunable frequency-doubled dye laser.²

The computer hardware and interfacing system are described in Sec. II. Then, in Sec. III the nature of the problems encountered in frequency control and scanning are outlined and the methods of solution are described. In Sec. IV the calibration procedures required to obtain the operating properties of the tuning elements are described. In Sec. V some actual example spectra and operating procedures are described.

II. Hardware

The computer-controlled color-center laser spectrometer consists of a single-mode color center laser, a minicomputer with appropriate interfaces and software, and diagnostic instrumentation (Fig. 1).

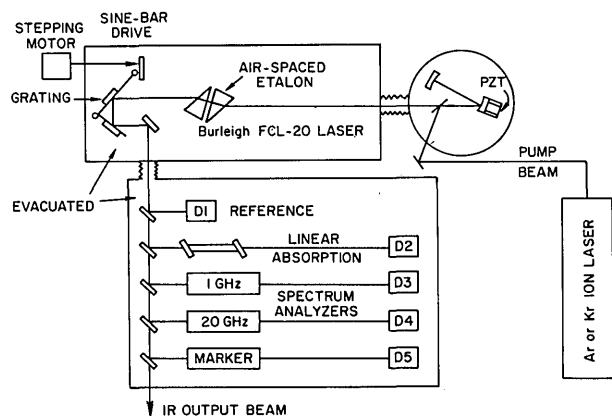


Fig. 1. Schematic of the color center laser spectrometer.

The operating principles of the color center laser have been thoroughly discussed in the literature¹¹⁻¹³; only general characteristics and those important to computer control and tuning are discussed here. The FCL-20 laser uses $F_A(II)$ color centers in Li-doped crystals of KCl and RbCl, and $F_B(II)$ color centers in Na-doped KCl. The single-mode tuning range of the laser extends from 2.3 to 3.3 μm , with output powers of the order of 20 mW. The crystals can be optically pumped either with an argon-ion (KCl:Li and KCl:Na crystals only) or with a krypton-ion laser (all three crystals).

In design and operation, the color center laser is similar to the cw dye laser. The laser is tuned by appropriate adjustment of three wavelength selective elements: a Littrow-mounted diffraction grating, an airspaced etalon, and a PZT mounted cavity mirror, as indicated in Fig. 1. The diffraction grating functions as both a tuning element and output coupler. The first-order reflection coarsely tunes the laser, while the zeroth-order reflection couples 10% of the power out of the cavity.¹⁴ The grating is mounted in such a way as to keep the direction of the output beam fixed and independent of the grating angle.

Under computer-controlled operation, the grating sine bar drive is rotated by a stepping motor (Superior Electric M061-FC02 driven by a STM103 control module with 400 steps/revolution) via an antibacklash 3-1 reduction gear to give a resolution of $\sim 0.75 \text{ GHz/step}$. The position of the grating is defined in terms of motor steps from a limit switch which is monitored continuously by the computer. Of key importance to computer operation of the laser is the reproducibility of the setting of the grating position. The stepping motor itself can be repositioned to within an accuracy of one step over a total range of 30,000 steps. Care must be taken to insure that the mechanical linkage of the sine bar drive of the FCL-20 does not exhibit excessive looseness or backlash in order that the actual grating position will be reproducible to reasonable levels (i.e., resettable to within $\sim 2 \text{ GHz}$).¹⁵

In the absence of an intracavity etalon, the color center laser operates on two longitudinal modes which

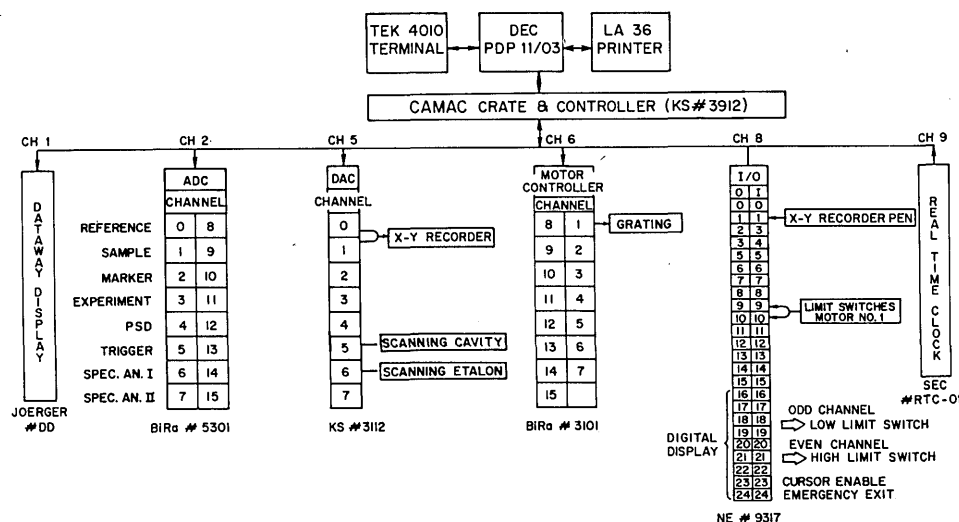


Fig. 2. CAMAC interface of color center laser to DEC PDP-11/V03 minicomputer.

are separated in frequency by 10 GHz as determined by the spatial hole burning effect in a standing wave cavity.¹⁶ Insertion of an airspaced etalon (the Burleigh FCL-500 etalon used for this work has a free spectral range of 22.1 GHz and 30% broadband reflective coating¹⁷) gives stable single-mode operation. The free running single-mode linewidth is <3 MHz.¹⁸ The frequency of the mode is determined by the cavity length, which is adjusted by the PZT driven cavity folding mirror. The airspaced etalon and PZT mounted mirror are driven by separate high voltage amplifiers (Burleigh RC-42 ramp generators) which are controlled by voltages from the computer.

The laser system is interfaced to a laboratory minicomputer using various CAMAC modules and a Kinetic Systems 3912 crate controller. Figure 2 shows the details of the interface. The DEC LSI-11/V03 minicomputer has a 28 K word memory and DEC RT-11 operating system. The operator communicates with the system via either a Tektronix 4010 graphics terminal or a DEC LA36 Decwriter. The computer in turn communicates with the laser system through the CAMAC interface. A BiRa 3101A driver controls the stepping motor used for positioning the grating. Resonant speeds of the motor, which can cause a loss of steps, are avoided by software comparison of, and if necessary, adjustment of the motor stepping rate, with a real-time clock (Standard Engineering RTC-01). Signals to control the high voltage amplifiers for the etalon and cavity PZTs are generated in two channels of a 12-bit digital-to-analog converter (Kinetic Systems 3112). Other DAC channels are used for data output to an x-y recorder. A 24-bit digital input/output module (NEC 9017U) is used to read the status of the limit switches. The I/O module also controls two alphanumeric displays for read-out of current data from various inputs and from the system program (e.g., motor position, wavelength, frequency).

Data from various signals, such as the laser power, spectrum analyzers, transmission through an absorption cell, etc., are acquired by a 12-bit differential analog-to-digital convertor (BiRa 4301). All signals must be buffered by operational amplifiers in order to lower the signal input impedances enough so that cross talk in the ADC is negligible upon switching channels. A 60-Hz line time clock is used for timing of most data acquisition and scanning operations. For such applications as noise measurements at higher frequencies, data are acquired at an 8-kHz rate derived from the RTC-01 clock.

The use of CAMAC modules makes the system versatile and easy to expand, but CAMAC is not the only or necessarily the best approach for interfacing a laser to a computer. All input/output functions could be handled by employing a four-channel D/A converter, sixteen-channel A/D converter, and a 16-bit input/output register, all of which are standard peripherals for most minicomputer systems.

In some modes of scanning it is necessary to lock the etalon onto the oscillating cavity mode.¹⁹ Figure 3 shows the schematic of the feedback loop used for etalon

stabilization. A 1-kHz dither from the local oscillator of a phase sensitive detector (PSD) is added to the computer-derived etalon control voltage in the high voltage amplifier (Burleigh RC-42). By monitoring the laser output with a PSD, whether the etalon position is leading or lagging the cavity mode can be determined and the appropriate correction made. The feedback loop is closed through the computer, which monitors this PSD output signal and adjusts the etalon to keep it in step with the cavity. The details of this feedback loop are discussed in Sec. III.

The diagnostic instrumentation used to characterize the laser output consists of a reference detector, a sample cell, a marker cavity, and two spectrum analyzers (Fig. 1). Laser power is monitored by the reference detector, and the resulting signal is used both in the calibration of the laser and in normalization of data. The sample cell consists of a 20-cm long tube enclosed with CaF₂ Brewster windows and with a 2-cm i.d. to allow for the possibility of multipass absorption measurements. The sample cell is normally filled with a gas whose spectrum is accurately known in the desired operating region to provide wavelength calibration. For example, NO is an excellent reference species for the 2.4–2.7- μ m region.²⁰ Alternatively, a digital wavemeter could be employed for direct wavelength calibration.²¹ A temperature compensated confocal cavity similar to that of Hercher,²² with 15-cm radius of curvature CaF₂ concave mirrors, is used to provide reference markers approximately every 500 MHz. The marker cavity mirrors are broadband coated for 95% reflectivity giving a finesse of ~ 60 . Two spectrum analyzers monitor the laser operation. One spectrum analyzer is a plane-parallel Fabry-Perot cavity with a FSR of 16.3 GHz, and the other is a confocal Fabry-Perot cavity with a FSR of 1.0 GHz. The coarse analyzer is useful for detecting and correcting two-mode operation, which occurs, for example, when the grating and etalon are not tracking properly. The fine analyzer is useful for confirming details of single-mode scanning. Both use CaF₂ optics broadband coated for 95% reflectivity. All detectors are room temperature lead sulfide covered with germanium windows. The beam splitters are 5-mm thick CaF₂ uncoated wedged flats. The wedge is oriented so

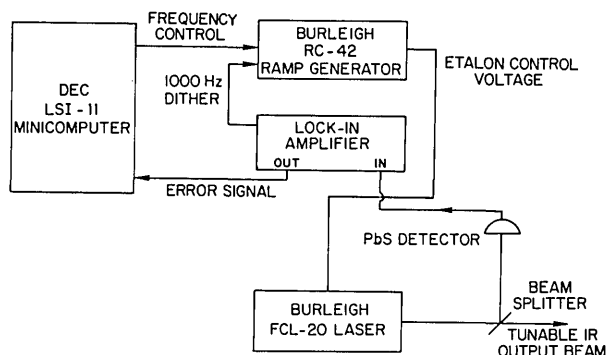


Fig. 3. Feedback locking of etalon spacing to the cavity length of the color center laser.

that the two reflections (one from each surface) diverge, thereby reducing interference effects on the detector.

Throughout the 2.5–2.8- μm region there exist strong atmospheric absorption lines due to water and CO_2 . Figure 4 shows two power tuning curves for the Li-doped KCl color center laser. One curve is taken through 20 Torr m of air, while the other curve corresponds to an evacuated cell. The laser output is almost totally attenuated at some wavelengths. Because of the strong variations in power and in the finesse of the marker cavity and spectrum analyzers, it is necessary to operate the instrumentation described above in an evacuated enclosure. The use of the evacuated chamber has some advantages besides reducing atmospheric absorptions of the laser beam. Since the marker cavity is evacuated, pressure variations which can seriously affect the marker positions are negligible. Also, acoustic vibrations in the atmosphere which can introduce random low frequency noise on the signals are nearly completely eliminated.

One difficulty with using an evacuated box is the inability to easily adjust optics without elaborate mechanical feedthroughs or small servomotors. The following scheme was developed so that the system can be aligned at atmospheric pressure and remain aligned upon evacuation. The beam splitters, spectrum analyzers, marker cavity, detectors, and sample cell are clamped onto 1.59-cm (0.625-in.) stainless steel bars connected together into a rigid frame. The frame is kinematically supported at only three points located near stiff corners of the chamber as further protection against warp-induced misalignment. The optical alignment of the system has remained true with only minor adjustments over months of operation and is virtually unaffected by evacuation or opening of the chamber.

III. Scanning

Three tuning elements must be simultaneously adjusted in order to achieve continuous single-mode scanning: the grating, the etalon, and the cavity length. The grating determines on which etalon mode the laser operates. The etalon determines which cavity mode lases. The cavity length determines the exact output frequency of the laser. The cavity length can be tuned by applying a voltage (0 to ~ 1000 V) to a PZT on which the laser folding mirror is mounted. Similarly, the etalon can be tuned by applying a voltage (again ~ 0 –1000 V) to a PZT on which one of its mirrors is mounted. The grating angle is changed by turning a screw sine bar drive under computer control using a stepping motor as described in Sec. II. The concept of continuous frequency scanning is in first order one of tracking the three tuning elements together so that the laser output mode changes frequency smoothly and continuously. To carry out this task, it is necessary to have detailed information concerning the tuning characteristics of the three tuning elements. How such information is obtained is the subject of Sec. IV on calibration. In this section we shall simply assume the availability of all such information.

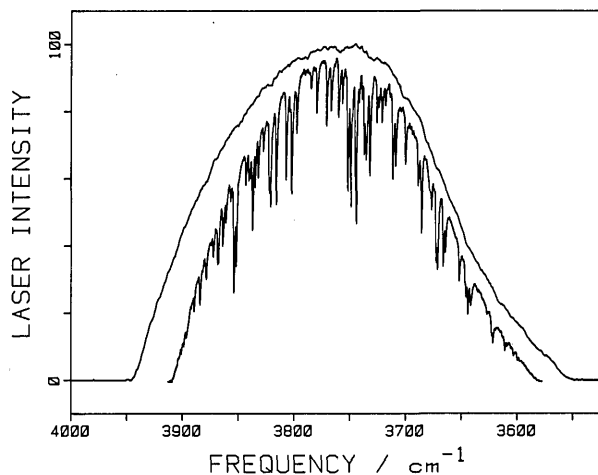


Fig. 4. Power tuning curve for the FCL-20 laser using the Li-doped KCl crystal in the absence and presence of atmospheric water absorptions.

The most obvious difficulty which arises upon tracking the tuning elements is that the tuning ranges of both the cavity and the etalon are limited by the finite extension that the PZT elements can attain before experiencing electrical breakdown. Thus it is necessary to ratchet or setback repeatedly both the cavity and etalon voltages once they have tuned beyond one (or more) FSR (a displacement of one-half wavelength). That is, in ramping up the PZT voltage, a point will be reached at which the voltage can be reduced to a much lower value which corresponds to the next lower-order mode of the cavity or etalon (increasing the PZT voltage increases the lengths of these cavities, therefore scanning the laser to lower frequency). The laser frequency will be the same as before the setback if the PZT is retracted exactly one-half wavelength. As a result, a scan actually consists of a number of short scans, and care must be taken that the segments match without gaps or overlaps.

The calibration procedure provides the necessary information for this scanning process by determining the FSRs and developing tables of the voltage tuning characteristics of both laser cavity and etalon PZTs and by developing a table relating the grating motor position to absolute frequency. At the simplest level, laser scanning is done by table look-up. The grating motor position, etalon voltage, and cavity voltage required for a particular output frequency are looked up in three different tables and set by the computer. This is a feed-forward open-loop concept. Its accuracy is surprisingly good but not high enough to achieve the desired goal of continuous single frequency scanning over wide regions.

Feed-forward fails for reasons other than insufficient accuracy. Even if the tables could be constructed with high accuracy, the laser cavity and etalon will occasionally get out of synchronization as a result of thermal drift or PZT hysteresis, and the laser will hop a mode.

A second problem is that the three tuning elements interact with each other so that instead of having three simple tables for look-up, a much more extensive 3-D table would be required. For example, the laser cavity length changes when the grating angle is changed or when the etalon PZT is expanded. Finally, all three elements are actual physical devices which possess nonideal properties. The PZTs exhibit both hysteresis and creep (creep is the slow changing of the PZT dimensions for some time after the PZT voltage is changed). The grating drive introduces mechanical vibrations which in the worst case may actually detune the laser.

As a result, open-loop feed-forward control must be supplemented by feedback corrections aimed at keeping the laser scanning continuous and smooth. Several types of feedback control are employed. First, as mentioned in Sec. I in connection with commercial cw dye lasers, the etalon is locked to the cavity mode which is lasing. The etalon PZT has an additional small sinusoidally oscillating voltage applied continuously at 1 kHz. The reference detector output is monitored by a phase sensitive detector (PSD) synchronized with the 1-kHz etalon dither (Fig. 3). If the etalon tracking is leading or lagging the cavity scan, the PSD output will be nonzero (either positive or negative). The PSD output is acquired by the computer and a correction to the etalon table is computed which adjusts the etalon voltage to track the cavity more accurately. Computer closing of the feedback loop is essential because very large noise spikes in the PSD output are introduced when either the cavity or the etalon voltage is ratcheted back or the grating motor is stepped. This computer closing is also desirable because a more sophisticated filtering of the noise inherent in the laser amplitude fluctuations can be made. The PSD can be sampled repeatedly for several PSD time constants to verify that a mismatch between the cavity and etalon is indeed developing.

Second, it has already been noted that the cavity length is affected by the grating angle. From Fig. 1 it can be seen that the axis of rotation of the grating is not on its front surface in the center of the laser beam. Thus, when a motor step changes the grating angle, the output frequency of the laser jumps discontinuously. The magnitude of the jump averages 17 MHz, with an unreproducibility however of about ± 5 MHz. This phenomenon is illustrated in Fig. 5 which shows the transmission through the marker cavity as the laser is scanned in a continuous single mode. The grating steps have been deliberately made when the laser frequency was on the side of the marker cavity features. Clearly illustrated is the coupling between grating angle and cavity length. To prevent these frequency jumps, grating steps are only made immediately after scanning through a marker cavity peak. At that point, data acquisition is halted, the frequency is set back ~ 100 MHz, and the laser is rescanned over the marker peak. The laser cavity length is then adjusted so that the centroid of the new marker peak matches the one obtained before the grating step, and scanning is resumed. The

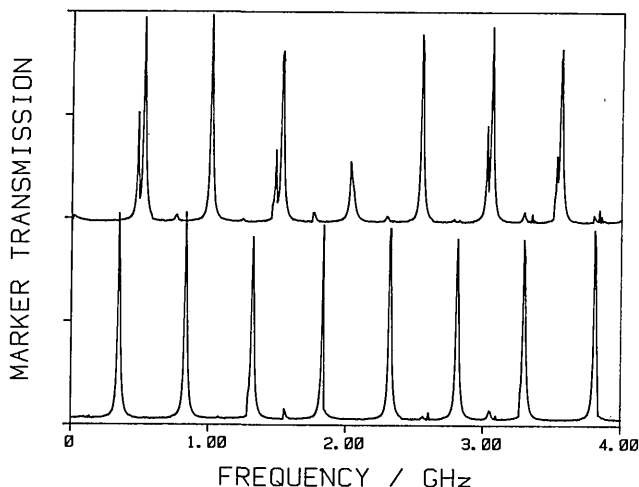


Fig. 5. Frequency scans of marker cavity without (upper trace) and with (lower trace) cavity length compensation when the grating is moved. The grating steps were deliberately made on the marker features.

marker cavity scan resulting from this procedure is also shown in Fig. 5. When the need arises to move the grating (about every 1.5 GHz) a reference marker is found, and a check is made to see if the cavity or etalon PZT is about to be reset. If so, the step is simply deferred to the next marker peak.

A very serious perturbation to the laser operation takes place when the etalon PZT is ratcheted back. Ideally the etalon would be set back exactly one FSR to the same cavity mode as prior to the flyback. Unfortunately, due to cumulative effects such as creep and hysteresis, it is impossible to predict exactly which cavity mode will be running after the system has settled following an etalon flyback. This is minimized by allowing a settling time of ~ 1 sec after resetting. Nevertheless, it is never certain by this procedure alone that the etalon is picking the correct cavity mode rather than an adjacent one. Moreover, the laser cavity length is affected by the etalon spacing. Thus, retuning the laser to the proper frequency after an etalon voltage reset must be done with some feedback through a matching procedure. In actual operation, the coarse and fine spectrum analyzer traces are acquired both just before an etalon voltage flyback and again after settling. Then the etalon and cavity length are adjusted by the amount required to match the spectrum analyzer traces acquired before and after etalon voltage flyback.

After the laser elements have been properly calibrated (as described in Sec. IV) it is possible to set the laser simply and rapidly to any desired frequency within ~ 0.05 cm^{-1} and make a continuous scan of any desired length within the range of operation of the particular color center crystal in use. In the absence of nearby strong water lines, the scan is linear between markers to ± 5 MHz. Therefore, to measure accurately the absolute frequencies of spectral lines, the spectrum of a known reference substance is obtained simultaneously, and the unknown line positions are found by interpolation using the reference marker cavity peaks.

The scanning procedure is summarized in Fig. 6 which shows the position of each tuning element during a 5.0-GHz scan. The grating makes discrete steps as necessary when the laser frequency coincides with a marker peak. The etalon and cavity length smoothly track, flying back after exactly one or two FSR. Clearly evident are the small corrections to the cavity length necessitated by either the grating steps or the etalon flybacks.

The most significant remaining problem associated with scanning is caused by tenacious water absorption features which affect the laser cavity operating mode and the marker cavity frequency position when the laser is operating in the vicinity of such a feature. The magnitudes of these shifts depend on the previous history of the laser cavity and diagnostics box, since water desorption occurs slowly. Near a waterline, the marker cavity FSR is altered slightly so that the marker cavity peaks are not in the right place. Therefore, caution must be used when measuring spectral lines in the neighborhood of a waterline.

There are a number of additional minor points regarding the scanning procedure which should be made. The first of these is that the periodic nature of the etalon and cavity scans is exploited to reduce the size of their cross-reference tables to slightly more than one FSR for the etalon and two FSRs for the laser cavity. The frequency look-up in the table to determine the voltage required is the frequency modulo times the element FSR for the etalon and modulo twice the FSR for the laser cavity.

As the laser is scanned to longer wavelengths, the etalon and cavity are moving to lower-order modes. Since the rate of change of frequency with spacing of a cavity is proportional to the mode order, table look-up for the voltage setting of these elements requires a computation to take into account the mode order. Since the necessary spacing will be proportional to wavelength, the etalon and cavity tables are simply scaled with wavelength.

Feedback is not introduced in ratcheting back the laser cavity. When the laser cavity is set back, the setback is ~ 15 MHz more than one FSR; a settling time of ~ 150 msec is allowed, and then the laser is scanned 15 MHz before restarting data acquisition. This procedure allows devices with external time constants to equilibrate prior to the continuation of the scan. With this procedure, the matching of cavity scan segments is accurate to ~ 10 MHz.

In summary, linear frequency scanning requires that the laser cavity frequency dependence on both the grating angle and etalon voltage be taken into account. The use of the marker cavity peaks in making a grating step removes the dependence of the laser cavity length on grating angle. The matching procedure using the acquisition of the fine spectrum analyzer compensates for the dependence of the laser cavity frequency on etalon voltage. To insure linear scanning, an additional etalon voltage dependent correction to the laser cavity scanning is made throughout the ramping of the etalon.

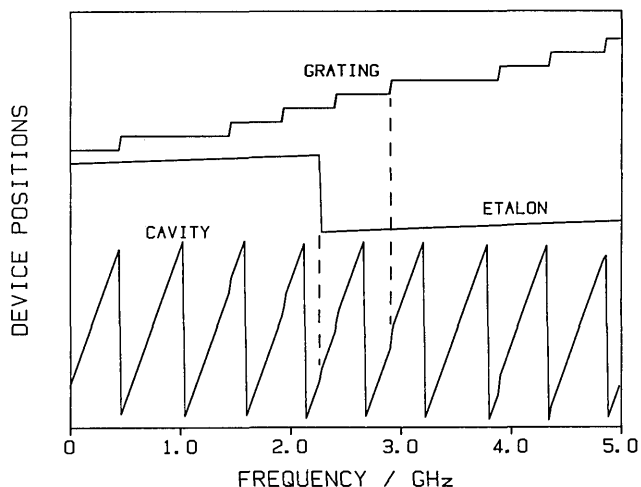


Fig. 6. Tuning element positions during a 5.0-GHz scan.

We have been describing a series of solutions to a series of problems which arise naturally from the physical properties of the control devices. It is worth noting that these solutions can themselves cause problems. For example, it is necessary to scan the etalon well past (~ 150 MHz) the point where a FSR setback would return it to the beginning of the etalon table. Otherwise, the resulting correction after resetting could provide a table look-up for the etalon voltage which corresponds to a point before the beginning of the etalon table. If this were allowed to happen, the etalon would do a set-forward back to the high voltages at the other end of the etalon table.

IV. Calibration

The key to reliable scanning lies largely in the development and subsequent use of accurate cross-reference tables. Each tuning element must be calibrated in position as a function of frequency. For the case of the etalon and laser cavity, it is also necessary to determine their respective free spectral ranges. The algorithms described herein perform these calibrations accurately, rapidly, and routinely.

The accuracy required for each table varies with the respective tuning element. For single-mode operation the grating must be centered on the etalon within ± 0.3 etalon FSR or ± 7 GHz.²³ For the grating this accuracy can be achieved without difficulty. For the etalon to stay centered on a cavity mode within ± 0.3 cavity FSR, it must remain within a 200-MHz range. This is not easily achieved because of temperature drifts and hysteresis effects. The overall accuracy of the scanning depends on the magnitude of these effects for the cavity PZT, and such drifts and variations limit the accuracy to ± 10 MHz.

Calibration of the grating is straightforward. The grating position is controlled by a stepping motor via a sine bar drive. Ideally such a sine bar drive should change the wavelength in a linear fashion as the motor is turned. However, in practice, periodic imperfections in the mechanical linkage (e.g., an eccentric screw) cause

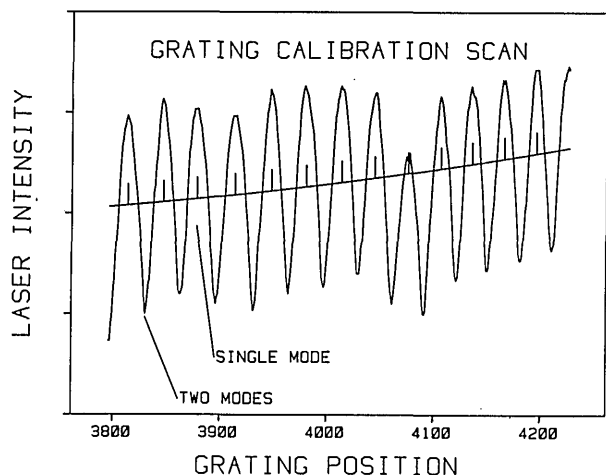


Fig. 7. Laser output power as a function of grating position with fixed etalon spacing. The peaks represent successive FSRs of the etalon.

the wavelength to change at varying rates during a scan. Since the grating must be centered on a particular order of the etalon to insure single frequency operation, the exact relation between motor position and grating position must be carefully mapped out and built into the cross-reference table.²⁴ Techniques for calibration by known dense spectra of calibrating gases have been developed. As a general calibration technique, they suffer from the lack of suitable spectra in many regions. A more general method of calibration is to use the etalon as an intracavity marker. With the etalon in the cavity, the laser operates in a single mode except when the grating is centered between two consecutive transmission orders of the etalon. At this point the laser operates in two modes with reduced overall power. If the grating is scanned while both the etalon and laser cavity length remain fixed, the output power of the laser is modulated in amplitude. Maxima occur when the grating is centered on an etalon mode and minima when it is between consecutive orders of the etalon. Figure 7 shows the output power over a short segment of such a scan. The software first performs a quadratic fit to segments of the data. Centroids are then calculated for each peak lying above the quadratic to find the motor position at the maxima. The smooth line in Fig. 7 represents the quadratic fit to the data; the vertical ticks show the centroid positions for the peaks lying above the fit. The grating cross-reference table is constructed from these positions.

This calibration technique has several advantages over other methods that use either a monochromator or a dense reference spectrum. There are approximately thirty-five FSRs (one FSR = 22 GHz) of the etalon per turn of the sine bar drive screw. This large number of data points yields a detailed map of the screw nonlinearity as seen in Fig. 8. Here the deviations from a quadratic fit of the data in the table are plotted. Since the deviation from linearity is as large as 35 GHz,¹⁵ correction is essential to the proper tracking of the

grating and etalon. With this procedure perfect synchronization of the grating with the etalon can be achieved because each peak is separated by exactly one FSR of the etalon. Furthermore, the procedure is fast. The grating table can be built over a 300-cm^{-1} range within 10 min. To complete the table, one point of the table must be assigned an absolute wave number, and the frequency interval between peaks, equivalent to the etalon FSR, must be entered. Reasonable estimates can be made initially with the exact values being determined subsequently from the spectrum of a well-known species.

Calibration of the etalon and cavity PZTs is complicated by three characteristics of these devices. First, a change in temperature can cause a small shift in frequency. Temperature drift is a slow process and, although not usually significant in a single scan, does become apparent over several hours by the apparent shift of spectral features. Second, the PZTs voltage response function is nonlinear.¹⁰ As with the nonlinear grating drive, it is necessary to carefully map out this function in order to have linear scans in frequency and to assure proper tracking of the wavelength tuning elements. Finally, the most serious problem exhibited by the PZTs is a slow relaxation effect occurring after a change in the applied voltage. There is a slow creep in the laser frequency even when the voltage is held constant. If left uncompensated, this effect can seriously limit the reproducibility of all scans. Not only would the laser often operate on the wrong cavity mode, but the magnitude of voltage necessary to scan the etalon one FSR would depend on the scan speed. The operation of a feedback loop to control these effects is described in Sec. III.

Two different approaches have been used to construct cross-reference tables for the etalon. One procedure, similar to that used for the grating calibration, uses the laser cavity modes as a frequency scale. Although the power is not noticeably modulated as the etalon scans

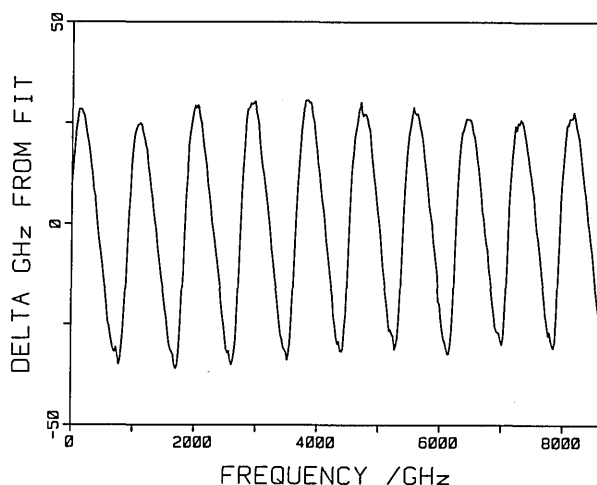


Fig. 8. Deviations from a quadratic fit of the positions of the etalon maxima vs grating motor position. The oscillations are thought to be due to screw eccentricity.

over adjacent cavity modes, the output of the phase sensitive detector observing the etalon dither indicates how the etalon is positioned with respect to the cavity mode. Figure 9 shows a typical output of the PSD as the etalon is scanned over several adjacent cavity modes. The PSD signal sweeps upward through zero volts each time the etalon centers on a cavity mode. The first derivative of these features is determined using a Savitsky-Golay smoothing function²⁵ to obtain maxima at the centered position. Since the maxima are separated by one laser cavity FSR, the frequency voltage characteristics of the etalon can be mapped out in detail. The accuracy of the table does depend on the accuracy of the value used for the cavity FSR. Because of mode pushing effects due to intracavity water adsorption, it is necessary to perform this calibration in a region with no such features. Such a water absorption and its effects are evident in Fig. 9.

There are some important points to this method of etalon calibration which, although not obvious at first, must be considered. The etalon cross-reference table should extend beyond one FSR of the etalon as was discussed above. To scan the etalon more than one-half of an FSR, it is necessary to move the grating an appropriate amount during the scan. However, as discussed above, the cavity length is changed slightly every time the grating moves. The resulting frequency shift almost always causes a perturbation in the pattern of markers each time the grating is moved. A second problem associated with the grating, a slight but significant mode pulling effect, prevents linear scanning over even short segments of the etalon range. When the transmission peak of the etalon is not centered on the grating's reflection peak, the cavity mode appears to be pulled toward the center frequency of the grating when monitored by the PSD. Thus as the etalon scans a region over the peak of the fixed grating, the cavity modes will appear red-shifted on the low side and blue-shifted on the high side of the grating, introducing systematic curvature into the subsequent cross-reference table. Thus it is necessary to simultaneously scan the grating when the etalon is being calibrated in this manner. To compensate for the resultant cavity length shift, the PZT mirror is moved an appropriate amount each time the grating is stepped. Because the actual shift is not reproducible, a small random error is introduced. The data points (~100) are thus fitted to a quadratic polynomial to smooth out this error and generate the final table.

The second approach to calibration of the etalon is less automatic but can be performed without moving the grating. Furthermore, the FSR of the etalon itself, rather than that of the cavity, provides the frequency information. Figure 10 shows two etalon scans made with both the cavity PZT and the grating held fixed. The frequency decreases with increased voltage to the etalon as the laser hops to lower cavity modes. The fixed grating dispersive loss causes the frequency to shift back to the next order of the etalon when it gets too far from the center wavelength of the grating. The same frequency region is again retraced. Features thus ap-

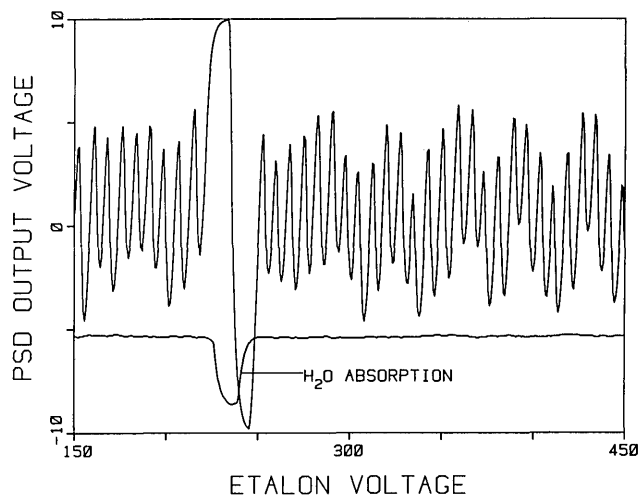


Fig. 9. Phase sensitive detector output as etalon is scanned over cavity modes. Large signal feature is due to cavity mode pulling by intracavity water absorption.

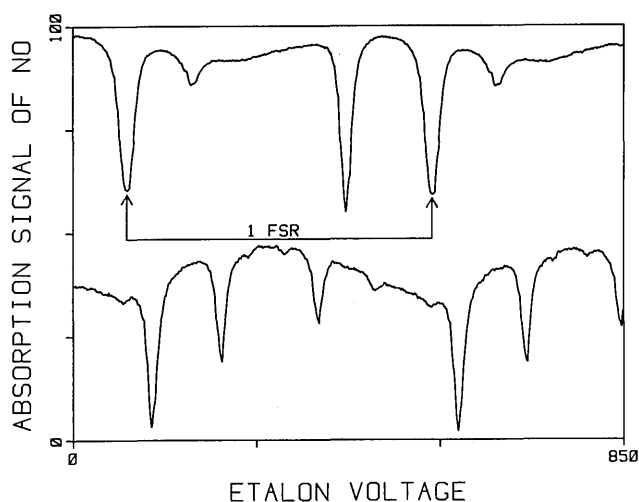


Fig. 10. Nitric oxide overtone absorption spectrum observed by scanning only the etalon. The laser frequency repeats every FSR of the etalon producing the repeated features. The spacing of these repeated features in etalon voltage is used in etalon calibration.

pear repetitively during a scan at an apparent shift in frequency of exactly one FSR. The ratio of the FSR to the voltage difference is the average etalon response over the region. Repeating spectral features can be found which appear at different etalon voltages thus permitting the response to be determined over the desired voltage range. Figure 11 shows the variation of the etalon response with the average voltage. A linear function is fit to this response curve and is integrated to give the etalon cross-reference table. This procedure is not totally automatic but is facilitated by software routines which perform the fitting and integration for pairs of features identified by the operator. The accuracy of the cross-reference table is limited by the drifts discussed above to ± 1 cavity mode or 300 MHz.

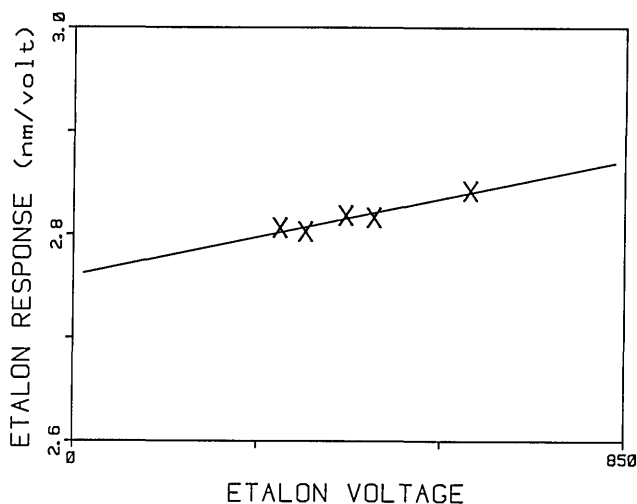


Fig. 11. Dependence of etalon response (rate of change of frequency with voltage) on applied voltage.

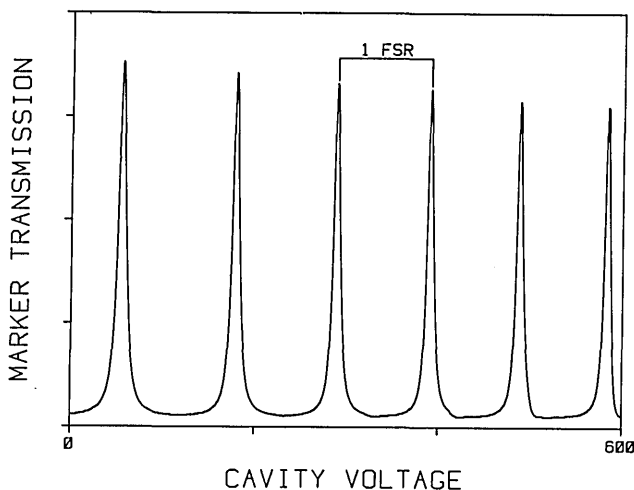


Fig. 12. Marker cavity signal as laser cavity is scanned. This is the same marker peak reappearing on different order cavity modes. The cavity PZT is calibrated by using the PZT voltages corresponding to the marker centroids.

The PZT driven cavity mirror has properties similar to those of the etalon. Since there is no convenient marker with a large number of calibration points within one cavity FSR, calibration by the first of the two methods described above is not possible. The 500-MHz marker does provide convenient features to which the second method can be applied. The cavity PZT is scanned several FSR while the grating and etalon are held fixed.²⁶ The same marker feature is repeated each time the cavity scans with the mode in the next lower order as seen in Fig. 12. The same software routine used for the etalon determines the cavity PZT frequency response, finds a polynomial fit thereto, and builds the cross-reference table for the PZT. For this procedure the cavity FSR is required; this can be determined iteratively (see below). Figure 13 shows a typical PZT

reponse curve. Drifts due to relaxation effects result in uncertainties of approximately ± 6 MHz. Drifts due to temperature changes during a scan of 10 min were found to be comparable. The relative accuracy is thus ± 10 MHz for such a scan.

Unfortunately, such a table is accurate only for the conditions used for generating it. Due to the physical limitations of the PZT, scans at different rates exhibit deviations of as much as 20 MHz. A final calibration of the PZT is performed using the positions of the marker features obtained during a long single-mode scan. Such scans can be obtained with good markers over a 10–15-GHz region without stepping the grating motor. The FSR of the marker is accurately known so that the true frequencies of the markers can be compared with the expected frequencies. The laser cavity FSR and the cavity PZT table are then appropriately adjusted. Software has been developed which allows this process to be carried out rapidly and conveniently.

The FSRs of the etalon and cavity need only be accurately determined once following alignment. Since the dimensions which set the FSRs remain stable under normal operation of the laser, it is reasonable to assume that these numbers remain constant over a substantial period of time. In both cases it is necessary to look at a well-known spectrum. The FSR of the etalon is determined by observing two spectral features which are very far apart (100 cm^{-1} or more). The position of each feature is determined to better than 1% of an FSR by scanning the etalon a short range. This fractional difference is added to the total number of etalon free spectral ranges between the features, which is known from the grating calibration. Depending on the accuracy of the known frequency separation between the two features, the FSR can be determined to as well as two parts in 10^4 , i.e., 5 MHz.

The cavity FSR is similarly found, except in this case the two spectral features must be within one etalon FSR

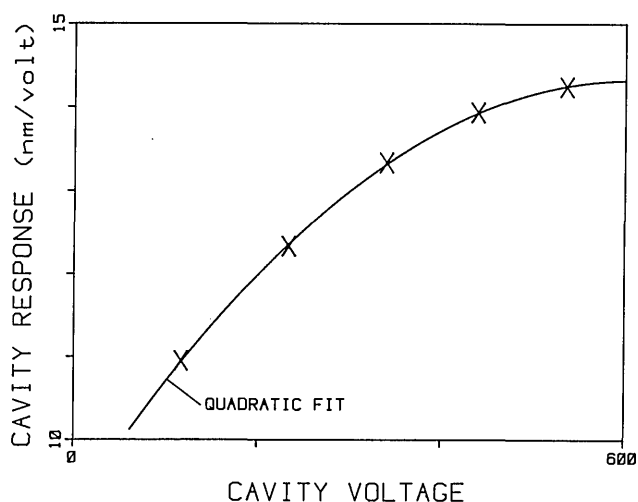


Fig. 13. Laser cavity response as a function of cavity voltage. Note the much greater curvature of the cavity response in comparison with the etalon response shown in Fig. 11.

of each other. The total number of cavity mode hops between the two features is easily counted and added to the fractional difference in positions within one cavity FSR. In this way, the FSR can be found to within 1 MHz if sharp features are known with sufficient accuracy. As a final check of both these numbers, a long range continuous scan is made between known features. Since any error will be cumulative, the difference between the actual and predicted frequency separation indicates the value of the actual cavity FSR.

At this point, the calibration procedure is complete. Once the laser and diagnostics are aligned and stable, the entire calibration can be performed in ~ 1 h. The calibration is stable unless any of the optics are realigned. Typically, a temperature drift will shift the etalon frequency a few gigahertz overnight, however, the resultant change is almost always a simple shift of the tables. From the position of a single known feature, suitable offsets to all tables can be readily implemented. We have found the tables to remain accurate for periods exceeding weeks. To summarize, the grating table is accurate to 1 GHz, the etalon table to 200 MHz, and the cavity table to 10 MHz, provided the spectrometer is not realigned.

V. Operating Procedures and Examples

The previous sections have concentrated on the production of calibrated, continuous single-mode scans. Clearly, much more is involved in the operation of the spectrometer such as control of data acquisition, data presentation, and data analysis. Operator interactions with the system are all made through a command language interpreter. After the control program has been entered, a standard prompt (FCL>) appears at the terminal. The operator then chooses the particular operation to be carried out by supplying a two-letter command code from a library of fifty commands and, if appropriate, up to five additional numeric or alphabetic parameters to complete the command specification. For the more complex operations such as calibration or spectrum plotting, a series of operator supplied instructions are then required.

The operating system provides considerable flexibility in control of the laser. For example, it is often convenient to make survey scans over large fractions of the operating range of the laser rapidly with lower resolution. This can be readily done by disabling the laser cavity scan and thus tracking only etalon and grating. The laser then scans by hopping cavity modes in steps of ~ 0.01 cm^{-1} . Because these hops are only about two-thirds the Doppler width for light molecules, there is no danger of missing an absorption line. Such a survey scan of NO is presented in Fig. 14.

Data acquisition is normally carried out in such a way that %T or absorbance is readily obtained. This is done by acquiring a signal proportional to laser power using the reference detector; then the absorption signal channel is divided by the reference channel signal. To eliminate drift in the dc detector channels, the laser beam is flagged off by a computer actuated solenoid during the settling time of an etalon flyback and thus

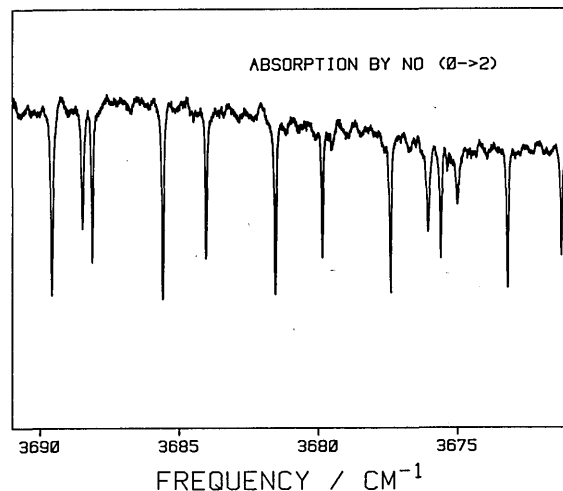


Fig. 14. Survey scan of nitric oxide using cavity mode hopping scan.

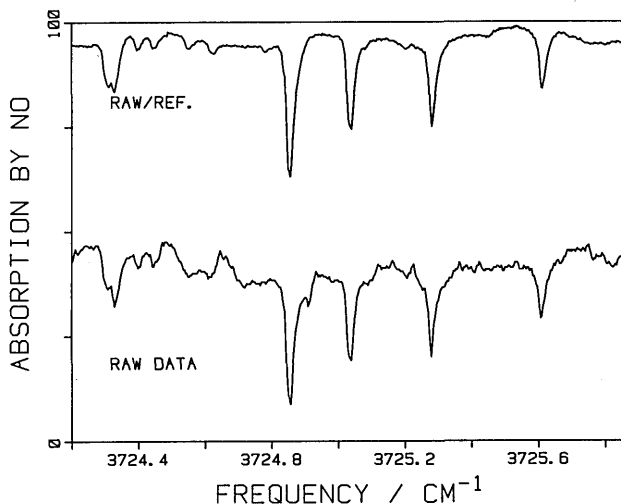


Fig. 15. NO Q-branch region scanned in continuous single-mode operation; upper trace, normalized by reference detector; lower trace, unnormalized by reference detector.

the zero point detector output is obtained. After the appropriate arithmetic, the spectrum is presented with a known 0%T, and a flat base line between absorption features which is set as 100%T by the operator.

This normalization process markedly increases the SNR for simple absorption spectroscopy because the laser intensity shows variations over small wavelength regions due to the cavity and etalon scanning process. In Fig. 15, the NO Q-branch region is shown before and after normalization by the reference detector signal and clearly illustrates the SNR improvement. When examined closely, the base line after normalization is still rolling slightly. We believe that this is the result of slight changes in the direction of the output laser beam upon tuning. If the laser beam is displaced slightly, the amount of displacement at the signal and reference

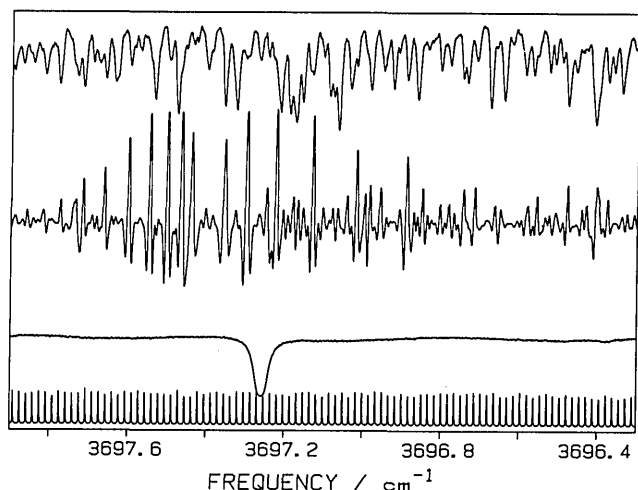


Fig. 16. Expansion of a portion of a 10-cm^{-1} scan of the OH stretch of methanol showing that detailed information from any portion of such a long range scan is accessible: upper trace, methanol absorption; next trace, 100-kHz Stark modulation in parallel polarization; next trace, NO reference spectrum; bottom trace, marker cavity signal.

detectors is different because they are located at different distances from the laser. Since the detector areas are quite small, this results in a differential change in signal.

Several 10-cm^{-1} scans of the OH stretching region of methanol near the band origin have been taken. Figure 16 shows a 1.6-cm^{-1} portion of such a scan. The four channels presented there include the NO reference, the cavity markers, the simple absorption spectrum of methanol, and the Stark modulation spectrum. A more detailed discussion of these observations on methanol will be published.²⁷ Here we wish to show that scans over wide regions which include all the information necessary for calibration of the spectrum are possible and that the means are available to focus on a particular region and examine it in detail without having to acquire more data.

VI. Summary

The conversion of a Burleigh FCL-20 color center laser into a long range high resolution spectrometer using computer control has been described. The spectrometer is capable of acquiring extremely high resolution spectra which can be precisely measured to $\sim 0.003\text{ cm}^{-1}$. If desired, 10 cm^{-1} can be covered in a single continuous scan.

The laser spectrometer has two levels of resolution determined by the tuning elements used in the laser cavity. Survey scans covering the entire 300-cm^{-1} region of stable operation of a single crystal can be made by scanning the grating and etalon simultaneously. The laser operates in a single longitudinal mode with the resolution limited by cavity mode hopping ($\sim 300\text{ MHz}$). A 3-MHz resolution (free running laser jitter) can be achieved by simultaneously scanning the cavity length, etalon, and grating.

The keys to computer control of the laser are in the development and use of cross-reference tables which relate the position of each tuning element to frequency and in the introduction of computer-controlled feedback. During a scan, each tuning element is sequentially positioned to track with the others with feedback introduced to overcome the interactions between the elements and their nonidealities. Once the cross-reference tables have been constructed, they remain accurate for weeks provided the laser is not totally realigned.

The authors would like to thank P. Carrick and E. Koester for invaluable technical assistance. This work was supported by the National Science Foundation and the Robert A. Welch Foundation (grant C-071).

References

1. O. Redi, *Phys. Today* **34**, 26 (1974).
2. C. R. Pollock, J. V. V. Kasper, G. K. Ernst, W. E. Ernst, S. Blit, and F. K. Tittel, *Appl. Opt.* **18**, 1907 (1979); C. M. Marshall, R. E. Stickel, F. B. Dunning, and F. K. Tittel, *Appl. Opt.* **19**, 1980 (1980).
3. M. Stuke and E. Marinero, *Appl. Phys.* **16**, 303 (1978).
4. H. Gerhardt and A. Timmerman, *Opt. Commun.* **21**, 343 (1977).
5. Moletron Corp., Sunnyvale, Calif. 94086.
6. T. Suzuki, H. Kato, Y. Taira, Y. Adachi, N. Konishi, and T. Kasuya, *Appl. Phys.* **24**, 331 (1981).
7. M. Endeman and R. L. Byer, *J. Opt. Soc. Am.* **70**, 1582 (1980).
8. M. J. Coulombe and A. S. Pine, *Appl. Opt.* **18**, 1505 (1979).
9. Laser Analytics, 25 Wiggins Ave., Bedford, Mass. 01730.
10. G. Litfin, C. R. Pollock, J. V. V. Kasper, R. F. Curl, and F. K. Tittel, *IEEE J. Quantum. Electron.* **QE-16**, 1154 (1980).
11. L. F. Mollenauer and D. H. Olson, *J. Appl. Phys.* **46**, 3109 (1975).
12. L. F. Mollenauer, "Color Center Lasers," in *Methods of Experimental Physics*, C. L. Tang, Ed. (Academic, New York, 1979), Vol. 15, Part B, pp. 1-54.
13. G. Litfin and R. Beigang, *J. Phys. E* **11**, 984 (1978).
14. K. R. German, *Appl. Opt.* **18**, 2348 (1979).
15. Deviations due to the nonreproducible drive train of the unmodified Burleigh are well in excess of this and may approach 150 GHz.
16. H. Schroder, H. Dux, and H. Welling, *Appl. Phys.* **17**, 21 (1973).
17. The FSR of earlier FCL-500 etalons was 26.23 GHz due to a different hole burning mode spacing.
18. The linewidth is limited by noise on the cavity PZT amplifier.
19. P. W. Smith, *IEEE J. Quantum. Electron.* **QE-1**, 343 (1965).
20. A. Henry, M. F. LeMoal, P. Cardinet, and A. Valentin, *J. Mol. Spectrosc.* **70**, 18 (1978); C. Amiot, R. Bacis, and G. Guelachvili, *Can. J. Phys.* **56**, 251 (1978).
21. S. L. Lee and J. L. Hall, "A Travelling Michelson Interferometer with Phase-Locked Fringe Interpolation," in *Laser Spectroscopy III*, J. L. Hall and J. L. Carlsten, Eds. (Springer, Berlin, 1977), pp. 421 and 422.
22. M. Hercher, *Appl. Opt.* **7**, 951 (1968).
23. The range for single-mode operation is critically dependent on alignment and pumping power as well as the specific laser device.
24. This assumes that the linkage is sufficiently tight and reproducible.

25. A. Savitsky and M. J. E. Golay, "Smoothing and Differentiation of Data by Simplified Least Squares Procedures," *Anal. Chem.* **36**, 1627 (1964); J. Steinier, Y. Termonia and J. Deltour, *Anal. Chem.* **44**, 1906 (1972).
26. Since the cavity FSR is 300 MHz, not all cavity modes can be swept through a marker. It is thus necessary to shift the etalon to a cavity mode which does scan through a marker. Alternatively, the PSD signal can be used instead of the marker signal. Although this is in some ways more convenient, it is less accurate.
27. E. Koester, P. G. Carrick, J. V. V. Kasper, and R. F. Curl, to be published.

Applied Optics

BOOKS

Books for review should be submitted to the Managing Editor, P. R. WAKELING, WINC, 1613 Nineteenth Street N. W., Washington, D. C. 20009

Laser Spectroscopy of Solids. Edited by W. M. YEN and P. M. SELZER. Springer-Verlag, New York, 1981. 310 pp. \$58.50.

That the development of lasers has revolutionized solid-state spectroscopy is indicated by the title of this book. An appropriate subtitle might have been "Point Defects in Insulators," since for the most part these are the systems whose properties are considered. The optical properties of such systems have been studied for many years, and several important books and review articles in this area have appeared during the past quarter of a century. Examples of these are D. L. Dexter's 1958 review article in the *Solid State Physics* series edited by F. Seitz and D. Turnbull, and B. DiBartolo's 1974 book *OPTICAL PROPERTIES OF IONS IN SOLIDS*. It was through these earlier studies that much of the basic understanding of the optical response of defects in insulators was achieved, including the important effects of the electron-lattice interaction.

The present volume concentrates on those aspects of the spectroscopy of point defects in insulators for which the laser makes a difference. Most of the treatments are related to defects for which the electron-lattice coupling is weak; this includes rare-earth and transition-metal ions but excludes most of the traditional color centers in insulating solids. Thus there is not much overlap of this with the content of *PHYSICS OF COLOR CENTERS*, edited by W. B. Fowler (1968) or *F-CENTERS IN ALKALI HALIDES* by J. J. Markham (1966).

The editors of this monograph have chosen their topics well and, together with the authors, have produced a comprehensive and well-integrated treatment of the field. Cross-references between articles are abundant. Such coherence is no mean task with ten authors contributing to the seven articles which comprise this volume. Treatments are current with references running through 1979 and in some cases into 1980. The book is well-produced and illustrated with but a handful of (obvious) typographical errors. It should remain current and be a valuable reference for some time.

Chapter 1 by Imbusch and Kopelman sets the stage for the following articles by a general and well-referenced, albeit brief, review of the optical properties of the types of defect which are later treated in more detail.

Holstein, Lyo and Orbach's chapter is an account of the microscopic theoretical basis for the various mechanisms of phonon-assisted excitation transfer that have been observed in disordered systems. This chapter is clear and thorough and ties together theory and experiment very well.

This is followed by a chapter by Huber on the dynamics of incoherent transfer. The temporal aspects of fluorescence line narrowing and the effects of traps on the integrated fluorescence are considered in detail. It is shown how valuable information on excitation transfer rates can be obtained from the experimental data.

Next, Selzer presents a brief summary of experimental techniques used in laser spectroscopy. Experimental details of several approaches, including fluorescence line narrowing, hole burning, polarization spectroscopy, and coherent transient spectroscopy are given. Again, abundant references are provided.

The final three chapters provide and discuss the experimental results associated with laser spectroscopy in various situations. Yen and Selzer treat ions in crystals, a topic which has been among the most widely studied. They consider effects associated with isolated ions as well as concentration-dependent phenomena such as energy transfer. They also discuss effects observed in concentrated and ordered ionic crystals, where excitons or magnons may play a large role.

Ions in glass are treated by Weber. The special results introduced by the nonperiodic structure are discussed, and experimental techniques and results are presented. Of particular interest is the attempt to use fluorescence line-narrowing data to probe the structure of the glass itself. This has provided useful information in a few instances.

In the final article Francis and Kopelman consider the dynamics of excitations in molecular solids. Special theoretical effects associated with such systems are given along with an abundance of experimental results. The nature of excitation migration in disordered crystals is treated in some detail in the context of various theories.

W. B. FOWLER

Lasers and Applications. Edited by W. O. N. GUIMARES, C.-T. LIN, and A. MOORADIAN. Springer-Verlag, Heidelberg, 1981. 335 pp. \$34.00.

Sergio Porto was a world-renowned scientist in the field of laser applications, in particular the use of Raman scattering. The present book, the proceedings of the International Conference on Lasers and Applications that was held in 1980 to commemorate Porto, contains thirty-seven papers presented by international laser scientists. The topics covered include the use of lasers in light scattering studies, spectroscopy, nonlinear optics, photochemistry, fiber optics, surgery, and optical bistability. Thus, the book contains a reasonably broad assortment of contemporary laser research in both the basic and applied sciences and should be useful for the reader who wants a broad knowledge of current directions in laser applications. Also, the papers in the book are generally readable by the nonspecialist, and are self-contained with enough reference citations for the readers that demand more. To give some indication of the materials covered, examples of some interesting articles in the book are given below.

Fleury and Lyons describe the great usefulness of narrow-banded lasers for high-resolution Brillouin and Raman spectroscopy to study phase transitions (such as ferroelectric transitions, cooperative Jahn-Teller transitions, etc.) in solids. Chang *et al.* present an excellent overview of surface-enhanced Raman scattering, a subject being actively investigated by numerous laboratories. Owyong and Esherrick report some recent developments of the inverse Raman spectroscopic technique which is highly sensitive, capable of sub-Doppler resolution, and can be used in hostile environments. Shen *et al.* give an inspiring account of nonlinear optics at surfaces, including harmonic generation and CARS, and the surface-enhancement possibilities of these effects. Rand *et al.* describe the observation of extremely narrow optical linewidths (~1 kHz) which they explain in terms of magic-angle line narrowing in solids. Patel *et al.* review the simple but powerful technique of laser optoacoustic spectroscopy which can be used to detect weak linear or nonlinear optical absorptions in condensed matter. Hellmuth *et al.* discuss the angular distribution and polarization dependence in multiphoton

continued on page 292