In-resonator laser spectroscopy with semiconductor lasers

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In-resonator absorption spectra have been measured with the help of diode lasers and suppression of loss selection. This suppression was achieved with the help of a specially developed laser diodes with Brewster-angle surfaces. The sensitivity found experimentally is lower than that expected theoretically. Ways to improve the sensitivity are discussed. © *1995 American Institute of Physics.*

In-resonator laser spectroscopy is currently the most sensitive method for absorption spectral analysis. The sensitivity in the detection of weak absorption lines is[1] 10^{-11} cm^{-1}. In-resonator spectrometers are widely used for spectroscopic research on forbidden and multiphoton transitions,[2] on the kinetics of chemical reactions,[3] on plasmas, etc. This method is based on the high sensitivity of the spectrum of the emission by a wide-band laser to the spectrum of the losses in the resonator. This sensitivity is characteristic of lasers for which the homogeneous width of the gain band is much larger than the characteristic period of the spectral dependence of the losses caused by absorption of the test substance in the resonator. If a dip by a factor of e in the output spectrum is to be manifested in an absorption line with a coefficient k (in cm^{-1}), the length of the generation pulse must be $t = 1/kc$, where c is the velocity of light. If the generation pulse has a length of 1 ms, for example, a dip by a factor of e in the spectrum is manifested in a line with an absorption coefficient of 3×10^{-8} cm^{-1}. In other words, the laser resonator in a study of in-resonator absorption is equivalent to a cell with an effective length $l = ct$. Limitations are imposed on the increase in the sensitivity with increasing duration of the generation pulse by nonlinear processes and by spontaneous emission from the active medium. Sensitivities in the detection of absorption ranging from 10^{-5} to 10^{-11} cm^{-1} have been achieved in various types of lasers. In particular, the figure for cw dye lasers is currently 10^{-11} cm^{-1}.

The lasers which have been used so far in in-resonator laser spectroscopy have used neodymium glass, solutions of organic dyes, and alkali halide crystals with color centers. All these lasers require either intense flashlamp pumping or coherent pumping. These requirements are fundamental difficulties in the effort to develop convenient instruments based on in-resonator spectrometers.

Semiconductor lasers look promising for use in in-resonator spectroscopy. These lasers offer a wide gain band with a short transverse-relaxation time, so the band can be regarded as being uniformly broadened. Semiconductor lasers span a broad region in the IR spectrum which cannot be spanned by other condensed-media lasers and which con-

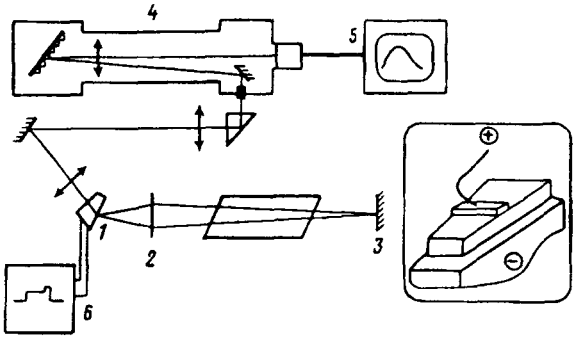

FIG. 1. Schematic diagram of the experimental apparatus. The inset is a schematic diagram of the emitting diode.

tains fundamental absorption bands of molecules. By working with fundamental bands instead of overtones one can improve the concentration sensitivity at a given sensitivity in terms of absorption. Back in the early years of the development of in-resonator laser spectroscopy, attempts were made to use semiconductor lasers. These attempts were complicated by the particular properties of the resonators of semiconductor lasers. The active medium of a semiconductor laser has a refractive index ~ 3.5, and facets of the crystal serve as the resonator mirrors. An additional mirror of the outer resonator does not substantially alter the Q of the resonator. The slight additional losses in the outer resonator due to the absorption under study have only a small effect on the emission spectrum. The effect is determined by the structure of the modes of the resonator formed by the crystal. The emission spectrum thus represents the mode structure of the semiconductor crystal. A brightening coating was applied to the facets of a semiconductor crystal in Ref. 4 to eliminate the effect of reflection. This coating reduced the reflection coefficient to a few hundredths of 1%. However, because of the difference between the dispersion curves of the semiconductor and of the titanium dioxide brightening layer, exact brightening could be achieved at only one point in the spectrum, and only for normal incidence; the output beam had a divergence of more than 10°. The output spectrum of the laser in Ref. 4 thus consisted of discrete lines corresponding to natural modes of the inner resonator. Modes which were overlapped by absorption lines of potassium vapor were not seen in this spectrum.

In this letter we are reporting the first observations of the spectrum of in-resonator absorption in experiments using a semiconductor laser. In an effort to counter the selective properties of the crystal surface, we developed some special laser diodes whose output surface was inclined at the Brewster angle with respect to the active region of the $p-n$ junction. In addition, a brightening coating was applied to the output surface of some of the crystals. We studied various resonator layouts, with two mirrors and also with three or four mirrors. We also tried using microscope objectives inside the resonator. Although the output facets of the crystal were inclined at the Brewster angle, a "parasitic" structure was seen in the emission spectrum of the laser. This parasitic structure was apparently caused by scattering by microscopic inhomogeneities at the crystal surface. By varying the resonator geometry and the pumping conditions, we were able to vary the depth of this parasitic modulation. The resonator layout shown in Fig. 1 turned out to be the optimum one for our conditions. A GaAs crystal *1* (shown in larger scale in

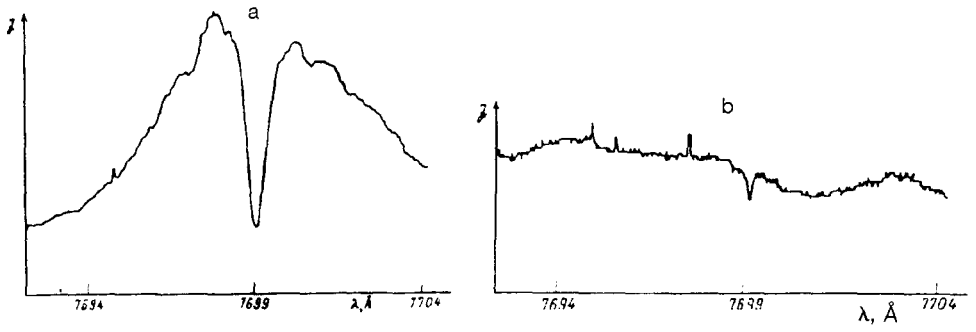

FIG. 2. a—Spectrum of the in-resonator absorption of potassium; b—absorption spectrum of potassium outside the resonator.

the inset in Fig. 1) was polished in the shape of a trapezoidal prism, whose rear facet was perpendicular to the channel of the $p-n$ junction and served as the output mirror. The second facet was inclined at the Brewster angle with respect to the active region. The light leaving through this facet was focused by microscope objective *2* onto a plane dielectric mirror *3* with a reflection coefficient of 99%. The astigmatism of the inclined Brewster surface was cancelled to some extent by rotating the microscope objective. The lasing thresholds of various diodes with this layout were 40–100 mA. It was found possible to achieve pumping at two or three times the threshold. The light generated, which exited through the normal surface of the crystal, was focused on the slit of spectrograph *4*, which was based on a UF-90 autocollimation chamber ($F = 130$ mm) and a 300-line/mm diffraction grating, operating in the eighth order of diffraction. The spectrum was measured by a Princeton Applied Research optical multichannel analyzer (*5*). The dispersion was 0.046 Å /channel, and the resolution was 2 channels.

The sensitivity of this spectroscopy is determined by the length of the generation pulse in the vicinity of the absorption line under study. We used pulsed pumping with a pulse length on the order of 1 μs. However, the crystal warmed up during the generation, causing frequency shifts of both individual modes and the entire generation spectral band. To partially cancel this temperature shift, we developed a special laser pumping unit *6*. This unit generated a two-step pump pulse. First, the crystal was heated by a current pulse below the threshold to a quasiequilibrium temperature; then the current was increased to a level above the threshold. As we have already mentioned, a residual structure was seen in the generation spectrum. We believe that this structure is due to light scattering by the Brewster facet of the crystal. The modulation depth associated with this structure differed from that in all previous studies in being less than 50%, even less than 20% in the better samples. We were thus able to obtain an in-resonator absorption spectrum with the help of a semiconductor laser.

Figure 2a shows the absorption spectrum of potassium vapor in a cell in the resonator of GaAs laser. Shown for comparison in Fig. 2b is the absorption spectrum of the same cell in the case of an in-resonator position. We see that the in-resonator position raises the sensitivity by two orders of magnitude. Still, this increase is much less than that

expected theoretically. A comparison with the single-pass absorption shows that the effective length of the absorbing layer is on the order of 10^3 cm. This figure corresponds to a duration on the order of 30 ns, which is far shorter than the length of the generation pulse (1 μs) and shorter than the duration of the generation near the absorption line. Possible reasons for this limitation are a high level of spontaneous noise associated with the low resonator Q, high losses in the focusing of the astigmatic beam reflected from the outer mirror into the active region of the crystal, and nonlinear effects in the active region. The low level of the feedback from the external mirror, which carries information on the in-resonator absorption, and the high level of single-pass gain, accompanied by a high level of spontaneous emission, tend to fill in the dips in the spectrum. Further research will be required to identify the reasons for the limitation on the sensitivity and to find ways to improve this sensitivity.

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