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High-contrast Doppler-free transmission spectroscopy

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By applying Doppler-free saturated absorption spectroscopy in the regime of high integrated sample absorption, high-contrast Doppler-free laser transmission signals can be obtained as demonstrated in experiments on the sodium D lines. Natural linewidth background-free signals are observed.

In Doppler-free saturation spectroscopy,[1,2] the saturating beam ordinarily causes only a small fractional change in the transmitted probe intensity, in order to avoid power broadening. We find, however, that if the probe is almost completely attenuated by the absorbing gas, the saturating beam can produce a large increase in probe transmission at the Doppler-free wavelength. Strong transmission peaks on an essentially zero background permit direct, noise-free detection. In our experiments, which were performed on the sodium D lines, a linewidth approaching the natural radiative one could be attained. The method permits simplified high-resolution spectroscopy in certain cases. Laser frequency-stabilization applications can also be foreseen.

The basic principle of the present experiments is illustrated in Fig. 1(a). The atomic or molecular sample is optically thick enough so that radiation of moderate intensity impinging upon the sample is totally absorbed. Only at the line center does the sample bleaching by a strong counterpropagating saturating beam allow the probe beam to penetrate the sample. A detector for the probe beam is exposed to a direct laser beam of substantial intensity at the line center but does not receive any radiation at all for neighboring wavelengths. Thus a favorable signal-to-background-ratio situation is achieved, and the noise in the signal reflects only the fluctuations of the laser beam. In this respect there is a resemblance to fluorescence monitoring but with greatly increased signal levels. Transitions of high oscillator strengths are desirable in order to obtain an opaque sample while keeping the number density in the sample low enough that collisional broadening does not pose a problem. However, by using long absorbing paths with completely overlapping beams, the desired experimental requirements can frequently be met.

In Fig. 1(b) a particularly simple experimental setup is shown schematically. Such an arrangement was actually used in most of our measurements. Sodium cells of 15-mm diameter and lengths up to 30 cm were used. Heating was provided by inserting the cell into a piece of straight copper tubing, surrounded by a few turns of heating tape. A Coherent Radiation Model 599-21 single-mode dye laser with a stabilized linewidth of about 1 MHz was used. Normally the laser was operated at an output power of about 60 mW. The division ratio of the beam splitter used is governed mainly by the requirements to achieve proper saturation for a chosen laser-beam diameter. If a relatively low beam-splitter transmission (10–30%) is used, the reduced final throughput after double-passing the cell is largely compensated for by the correspondingly higher beam-splitter reflection (90–70%) of the beam into the detector. Further, low beam-splitter transmission reduces problems with feedback into the laser cavity, even off the Doppler-broadened lines. A simple silicon detector without any bias voltage was used, and the signal was directly recorded on an X–Y recorder.

In the optical arrangement of Fig. 1(b) the probe-beam intensity in the frequency region of the Doppler-free peaks is automatically reduced to a fraction of the pump-beam intensity, which ensures high-contrast signal transmission. A constant and freely selectable probe-beam intensity can be obtained by using the standard setup for saturated absorption spectroscopy with a beam splitter dividing up the primary laser beam into two beams, which are made to impinge upon the sample from opposite directions. For example, a constant, very weak probe beam can be chosen, or two
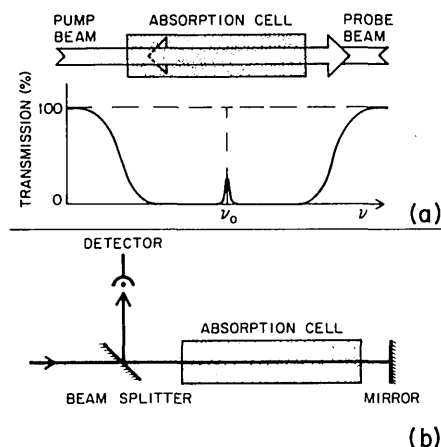

Fig. 1.   (a) Basic principle of high-contrast, Doppler-free transmission spectroscopy. (b) A simple experimental realization of Doppler-free transmission spectroscopy.
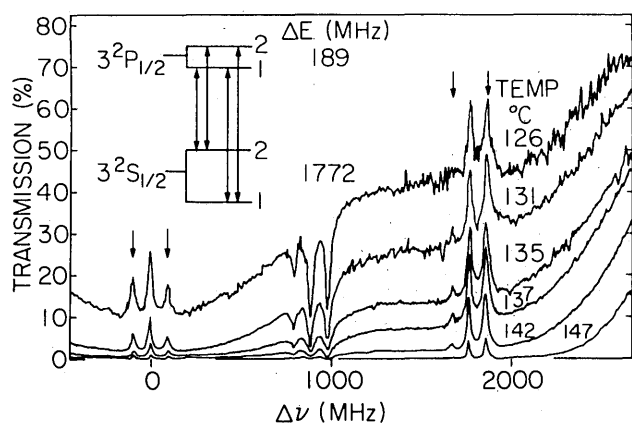
Fig. 2. Experimental recordings for the sodium D_1 line. A 30-cm-long absorption cell was used. A pump beam of a power of about 0.8 mW at the entrance of the cell was used, corresponding to a power density of about 30 mW/cm². The vertical scale indicates the transmission, i.e., the signal intensity in percent of the power obtained when the laser is completely tuned off the absorption line. A hfs transition diagram is included.
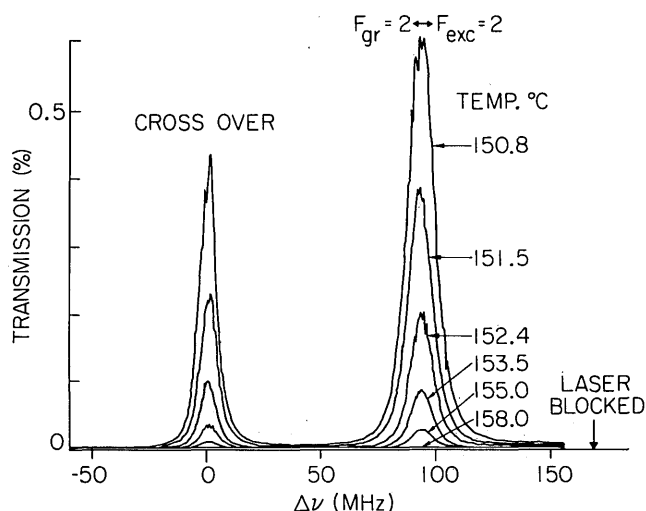

Fig. 3. Recordings of the $F_{gr} = 1 \rightarrow F_{exc} = 2$ D_1 line component and the adjacent crossover signal. The experimental conditions are the same as in Fig. 2.

equally strong, only partly saturating beams can be used that open up the cell for each other only at the line center.

In Fig. 2 recordings of the sodium D_1 line (3 $^2S_{1/2}$–3 $^2P_{1/2}$, λ = 589.6 nm) are shown for different cell temperatures. A setup as illustrated in Fig. 1(b) with a 30-cm cell was used. A diagram of the different hyperfine-structure (hfs) transitions is also included. For the lower cell temperatures all the different hfs components, corresponding to the 1772-MHz ground-state and the 189-MHz excited-state splitting, are shown. In addition, crossover signals, corresponding to transitions involving a common ground or excited substate, are obtained. As the temperature is increased the absorption in the central parts of the Doppler absorption profile starts becoming complete.

This occurs first at the $F_{gr} = 2 \rightarrow F_{exc} = 1, 2$ components and later at the $F_{gr} = 1 \rightarrow F_{exc} = 1, 2$ components. However, for frequencies corresponding to the Doppler-free transitions, for which the probe beam feels the strong bleaching by the pump beam, a strong transmission persists. A narrowing of these signals occurs for increasing temperatures. In Fig. 3 the $F_{gr} = 1 \rightarrow F_{exc} = 2$ component and the adjacent crossover signal are shown in the temperature regime of high absorption. A high signal contrast over the background is clearly shown. The residual background observed even after the signals have disappeared is due not to any light transmitted through the sample but rather to scattered light in the beam splitter and the cell window. Thus, in principle, this background, which reduces the contrast obtained, can be eliminated by spatial filtering or just by moving the detector for the transmitted laser beam far away. It can be seen in the figure that the signal outside the transmission peaks is essentially noise free, whereas the noise in the peaks reflects the fluctuations of the laser.

Further insight into these phenomena is provided from Fig. 4, in which data for Doppler-free signal transmission (background subtracted), signal-to-background ratio (contrast), and signal full width at half-maximum are presented as a function of integrated atom number density (volume density $\times$ path length). The behavior of the $F_{gr} = 1 \rightarrow F_{exc} = 2$ signal component is studied for two laser intensities that differ by a factor of 10, and the corresponding data curves fall in different domains of the figure, divided by the diagonal line. The transmission curves are
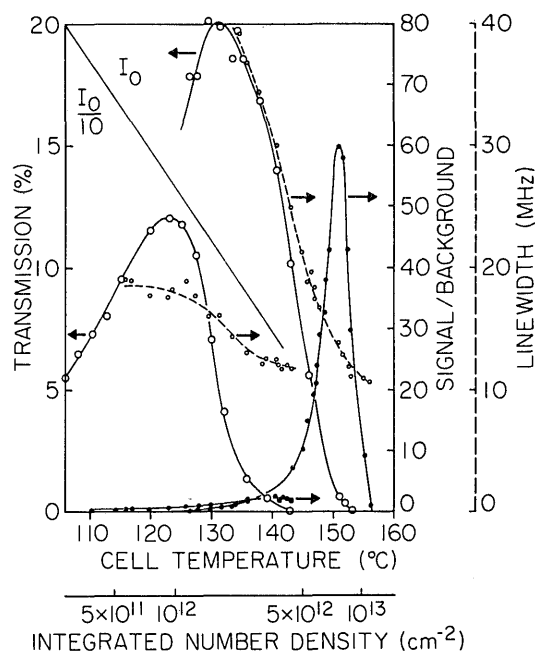

Fig. 4. Diagram for the $F_{gr} = 1 \rightarrow F_{exc} = 2$ D_1 line transition showing Doppler-free signal transmission, signal-to-background ratio (contrast), and signal half-width (FWHM) for two laser intensities. High- and low-intensity data fall in different domains of the figure, divided by a diagonal line. Arrows point toward scales to be used. I_0 is about 0.8 mW, corresponding to a power density of about 30 mW/cm².
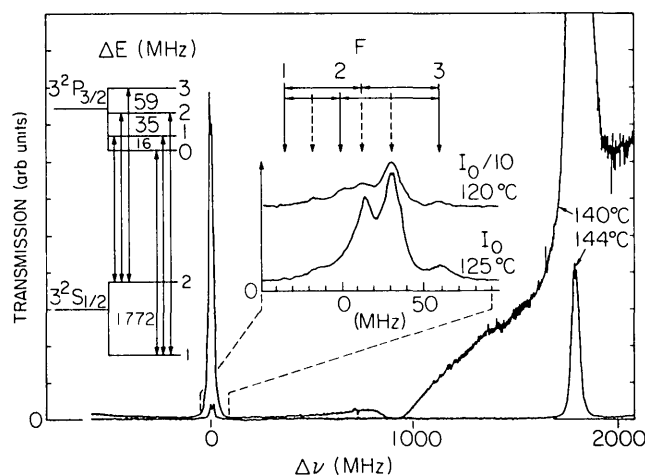
Fig. 5. Experimental recordings for the sodium D_2 line obtained for two different temperatures of a 10-cm long cell. A separated probe beam that had 2.5% of the pump-beam intensity was used. On an expanded frequency scale the hfs components connecting to the $F = 2$ ground level are shown. These latter recordings were taken with a 30-cm cell in an arrangement as shown in Fig. 1(b). Crossover signals between the regular three hfs transitions are observed.

drawn as solid lines with arrows pointing to the corresponding scale in the left-hand part of the diagram. The curves showing the contrast are also drawn with solid lines, now with arrows pointing to the corresponding scale in the right-hand part of the figure. Finally, the linewidth results are given as dashed lines to be evaluated against the dashed linewidth scale in the right-hand part of the figure.

Data were obtained for a 30-cm cell preceded by a beam splitter with a primary transmission of 10% in an arrangement illustrated in Fig. 1(b). For the higher laser power a maximum signal transmission of 20% was observed at 132°C. Then the signal-to-background ratio is about 1, a value that increases to 60 at 151°C, with a transmission of about 0.6%. The subsequent contrast falloff occurs because of the (temperature-independent) level of scattered background light, as discussed above, which in this case was about 0.003%. Further, from the figure it can be seen that the signal linewidth is reduced from more than 40 MHz (corresponding to the particular level of saturation broadening) to about 10 MHz. This value happens to be close to the natural radiation width corresponding to the excited-state lifetime by 16 nsec. The observed line narrowing is in a sense "artificial," since it is due to the difference in attenuation between the line center and the wings, which reflects the exponential nature of the Beer–Lambert law of absorption. It is worth noting that an interesting parameter combination of 5% Doppler-free-signal laser transmission, a contrast of 20, and a linewidth of 18 MHz can be achieved simultaneously. The data in Fig. 4 for the low-laser-inten-sity case demonstrate that contrast is quickly lost with weakened saturation. A maximum contrast of about 2 is observed, now at a much lower temperature than for the high-intensity case.

The sodium D_2 line ($3\,^2S_{1/2}$–$3\,^2P_{3/2}$, $\lambda = 589$ nm) was also studied. Here the excited state has a much smaller hfs. Very high transmission and contrast values were observed for unresolved signals recorded at substantial saturation conditions, as shown in Fig. 5. However, at lower laser power hfs components corresponding to transitions to the $F_{gr} = 2$ level can be partially resolved in spite of the presence of strong crossover signals, as demonstrated in the figure inserts. Signals for two laser intensities differing by a factor of 10 are shown.

The fact that the saturating beam is also gradually absorbed with increasing penetration depth into the sample leads to reduced bleaching at the rear end of the cell and to reduction of the signal transmission and contrast otherwise obtainable. By multipassing the cell and repumping the folded path with fresh laser beams, or, more simply, by slightly focusing the pump beam in a single pass to converge toward the back end of the cell, the saturation can be maintained at a high level with increased system performance.

By placing two setups of the type shown in Fig. 1(b) after another in a tandem arrangement, further contrast improvement and linewidth reduction can be expected as a consequence of the combined nonlinear effects. The first stage could then be considered a laser with atomic-resonance output control. For this first stage an expanded high-power laser beam would be required. By means of a beam compressor (inverted beam expander) between the two stages the correct saturation conditions can be maintained through the second stage. Subnatural linewidths for isolated components might be attainable.

The sharp and strong signals discussed in this Letter might be useful for stabilizing single-mode lasers. A simple stabilization of single-mode laser diodes to the strong resonance lines of K, Rb, and Cs, which fall in the wavelength region of such lasers operating at room temperature, might be accomplished.

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