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## High-Resolution Saturation Spectroscopy of the Sodium *D* Lines with a Pulsed Tunable Dye Laser\*

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The sodium *D* resonance lines have been studied in saturated absorption with a repetitively pulsed tunable dye laser. The hyperfine splitting of the  $3^2S_{1/2}$  and  $3^2P_{1/2}$  states of  $\text{Na}^{23}$  is resolved. Measurements with a delayed probe reveal a remanent hole burning in the velocity distributions of the two ground-state levels, caused by a velocity-selective optical pumping cycle. A time-resolved observation of the collisional-velocity thermalization in the presence of Ar buffer gas is reported.

In the past the techniques of laser saturation spectroscopy<sup>1-5</sup> that eliminate Doppler broadening by spectral hole burning have essentially been restricted to either gas-laser transitions or molecular transitions in accidental coincidence. Although organic-dye lasers have been tuned to atomic resonance lines,<sup>6,7</sup> they generally did not provide the required spectral resolution and stability. The present paper reports on a study of the sodium resonance lines  $D_1$  at 5896 Å and  $D_2$  at 5890 Å in saturated absorption using a repetitively pulsed tunable dye laser with an instrumental bandwidth of 7 MHz and a pulse length of 30 nsec. The use of a pulsed laser source is made possible by a recently introduced sensitive technique of saturation spectroscopy.<sup>4,5</sup> Two light beams of equal frequency are sent in opposite directions through the resonant atomic vapor, and the change in absorption for one of the beams, the probe, caused by the other, the bleaching or saturating beam, is recorded. A

saturation signal is observed if the laser is tuned close to an atomic resonance frequency, so that both light waves interact with the same atoms, i.e., those with essentially zero axial velocity.

A simplified scheme of the experimental setup is shown in Fig. 1. The narrow-band tunable dye

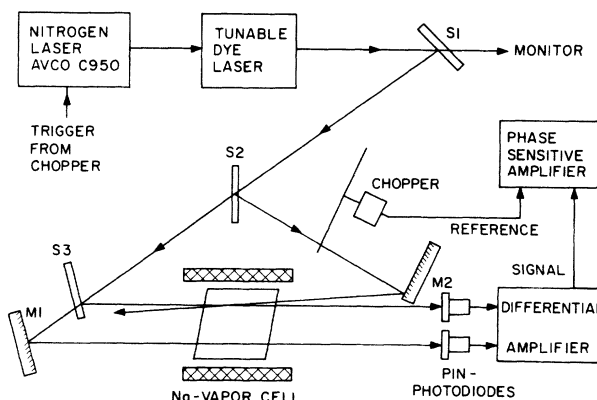


FIG. 1. Experimental setup.

laser, side-pumped by a nitrogen laser,<sup>8,9</sup> will be described elsewhere in detail.<sup>10</sup> A  $5 \times 10^{-3}$ -mole/liter solution of a 20:1 mixture of rhodamine 6G and rhodamine B is the active medium in the present experiments. A piezoelectrically tunable confocal Fabry-Perot interferometer (free spectral range, 2 GHz) serves as an ultra-narrow passband filter outside the dye-laser cavity. It reduces the linewidth from 300 to 7 MHz full width at half-maximum (FWHM) and stretches the pulses from 5 to 30 nsec FWHM. Peak powers on the order of several watts are obtained in this way up to repetition rates of 100 pulses per second in a nearly diffraction-limited output beam. During the measurements the dye-laser frequency is continuously scanned, within about five minutes, over a frequency range of several GHz. This is accomplished by tilting an intracavity Fabry-Perot etalon with the help of a motor-driven micrometer and simultaneously tuning the external passband filter by applying a proper voltage ramp. The fringe system displayed by an additional fixed Fabry-Perot interferometer is used to monitor the laser spectrum and to provide a frequency calibration.

The Na vapor cell of 10 cm length and 2.5 cm diameter is made from Pyrex, and the windows are tilted with respect to the optical axis to avoid back reflection. The cell is heated by an electric oven. The temperature of a glass finger containing the metallic Na supply is kept at 110°C, corresponding to a vapor pressure of about  $3 \times 10^{-7}$  Torr or a  $1/e$  absorption length of 15 cm at line center. With beam splitters and mirrors, a probe beam and a saturating beam are sent in nearly opposite directions through the absorption cell so that they cross at a small angle ( $< 10$  mrad). Attenuators reduce the peak powers to about 0.5 and 50 mW, respectively. The beam diameters are typically 4 mm. In a differential detection scheme the intensity of the probe after absorption is compared with that of a second dummy probe that also passes the Na vapor, but does not cross the bleached region. In this way the noise due to random laser amplitude fluctuations is greatly reduced. The pulse energies are measured by means of two *p-i-n* photodiodes, connected to the input terminals of a differential pre-amplifier with a pulse-stretching passband filter (10–100 Hz). Every second saturating pulse of light is blocked by a rotating chopper to which the dye-laser pump, operating at 80 pulses per second, is synchronized. The resultant ac modulation of the probe pulse amplitude is measured

by a lock-in amplifier. Relative probe-intensity changes down to  $10^{-3}$  can thus be registered with 1-sec time constant. The saturation signal is finally recorded by an *x-y* plotter versus the tuning voltage at the laser passband filter.

Figure 2(a) shows a saturation spectrum of the Na  $D_1$  line. The zero point of the frequency scale is arbitrary. The neighboring  $D_2$  line, if plotted on the same scale, would appear some 10 m to the right. Both the  $^2S_{1/2}$  ground state and the excited  $^2P_{1/2}$  state of Na<sup>23</sup> are split into two hyperfine components.<sup>11</sup> The splittings are 1772 and 192 MHz, respectively. The corresponding possible hyperfine transitions with their relative oscillator strengths are shown at the bottom [Fig. 2(c)]. All four resonance lines are clearly resolved in the observed spectrum. Additional structures appear halfway in between the closely spaced lines. There are atoms with an appropriate axial velocity for which, at these laser frequencies, the saturating beam appears to coincide with one transition and the probe appears to be at the other. The observed signal is interpreted as the expected crossover bleaching due to a common ground-state level.<sup>12</sup> A corresponding crossover signal is also expected near the center of Fig. 2(a), i.e., in between lines that start from different ground-states levels, but share a common upper level. The actually observed signals appear inverted, however. This seems to indicate that the crossover bleaching is here overcompensated.

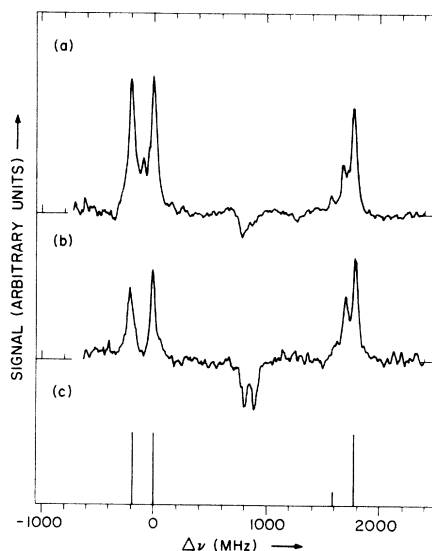


FIG. 2. (a) Saturation spectrum of the Na  $D_1$  line without delay. (b) Like (a), but with a probe delay of 56 nsec. (c)  $D_1$  hyperfine transitions with relative oscillator strengths.

sated by the effect of a velocity-selective optical pumping and fluorescence cycle. When the atoms absorb saturating radiation and subsequently radiate spontaneously, some of them return to their original level, but some end in the other ground-state level. Thus the number of atoms in the other level is increased and additional absorption is observed by the probe. Such a model implies that the holes and peaks in the velocity distribution of the stable ground-state levels should be remanent many radiative lifetimes after the bleaching, until the atoms move out of the probed region. This is confirmed by the second spectrum [Fig. 2(b)] which was obtained by delaying the probe pulse 56 nsec with respect to the bleaching pulse by means of a folded optical delay line. The bleaching at the resonance frequencies remains observable and the inverted signals near the center appear more pronounced after the time delay that allows for spontaneous emission.

The narrowest observed linewidth of one component of the  $D_1$  line is  $40 \pm 4$  MHz (FWHM). The natural linewidth, corresponding to 16 nsec radiative lifetime, is 20 MHz for comparison. The difference is ascribed to the final laser bandwidth and to residual Doppler broadening due to the finite crossing angle of the two light fields. Distinct power broadening is observed at higher pump powers. It is expected that a simple hole-burning model<sup>4</sup> with an appropriate interpretation

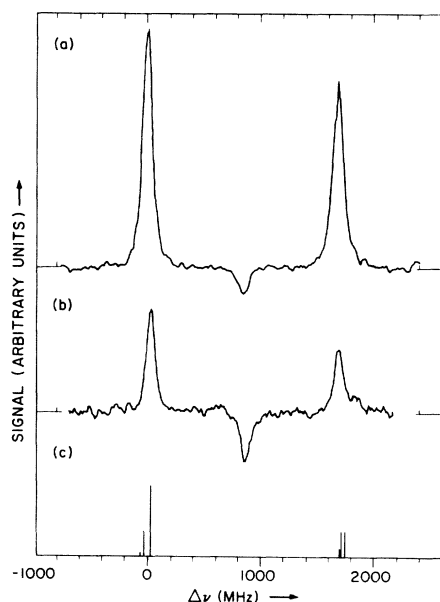


FIG. 3. (a) Saturation spectrum of the Na  $D_2$  line without delay. (b) Like (a), but with a probe delay of 700 nsec. (c)  $D_2$  hyperfine transitions.

of the time constants involved still provides a reasonable description in the present limit of small light intensities, although transient effects<sup>6</sup> certainly are no longer negligible for pulse widths comparable to the radiative lifetime.

The corresponding results for the sodium  $D_2$  line are shown in Fig. 3. Here the hyperfine splitting of the upper  $^2P_{3/2}$  state [Fig. 3(c)], which can hardly be resolved even with optical rf double-resonance and level-crossing experiments,<sup>13,14</sup> leads to a line broadening in the present experiment. The second spectrum [Fig. 3(b)] has been obtained with 700 nsec time delay between bleaching and probing. Despite the long delay the spectrum appears almost without deterioration and shows a very pronounced enhanced absorption at the center.

Atomic collisions in pure Na vapor can certainly be neglected at the present vapor temperature. The situation is different in measurements that are obtained in the presence of 3.1 Torr Ar buffer gas (pressure at room temperature). Here, the lines broaden with time because of elastic Na-Ar collisions that tend to thermalize the velocity distribution of the Na ground-state atoms. In Fig. 4 averaged values of the observed linewidth are plotted versus the delay time. After a relatively rapid initial increase, the width finally approaches the value expected for thermal equilibrium. Considering the present time resolution of 30 nsec, measurements at a much lower buffer-gas pressure are more desirable for a detailed quantitative analysis. At any rate the present measurements permit a rather direct observation of collisional-velocity changes. In the previous studies of pressure broadening by steady-state saturation spectroscopy,<sup>1,3</sup> on the other hand, it is generally quite difficult to distinguish the effect of velocity-changing colli-

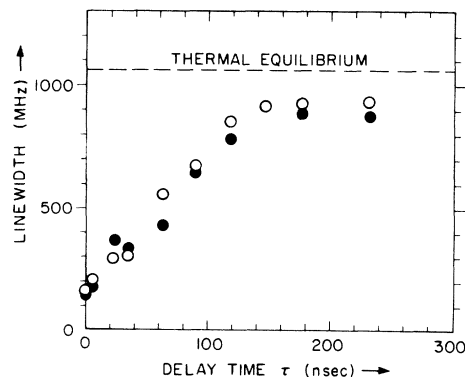


FIG. 4. Collision-broadened linewidth of the  $D_2$  components versus probe delay time at 3.1 Torr Ar.

sions from the line broadening due to a dephasing of the atomic oscillators. There were indications, however, that the collisional diffusion in velocity space can in fact dominate the observed pressure broadening of Lamb dips.<sup>3</sup>

Relatively simple theoretical models for the Brownian motion of particles in velocity space can be given in the two limiting cases of "hard" collisions<sup>5</sup> and of very "weak" collisions, where the Boltzmann integral equation can be replaced by a Fokker-Planck differential equation for a diffusion process.<sup>15</sup> The total elastic scattering cross section for Na-Ar collisions is  $401 \text{ \AA}^2$ , as determined from atomic-beam experiments.<sup>16</sup> The corresponding mean time between two collisions is 3.1 nsec. A comparison with the observed line-broadening rate seems to favor a diffusion model. Such an assumption is also in qualitative agreement with the known differential cross section for a far-reaching van der Waals potential that strongly favors small-angle scattering.<sup>16</sup> In the diffusion approximation the velocity distribution of atoms with a given initial velocity  $v_0$  can be given in an analytic form. At a later time  $t$  it is a Gaussian, centered at  $v_0 \times \exp(-\eta t)$  with a width  $\Delta v = \Delta v_D [1 - \exp(-2\eta t)]^{1/2}$ , where  $\eta$  is the dynamic friction coefficient and  $\Delta v_D$  the width in thermal equilibrium.<sup>15</sup> The corresponding expected linewidth in the present saturation experiment is  $\Delta\nu = \Delta\nu_D \{ [1 - \exp(-\eta t)] / 2 \}^{1/2}$ , and a tentative fit to the experimental data yields a dynamic friction coefficient  $\eta = (3 \pm 1) \times 10^{-6} \text{ sec}$  although unresolved hyperfine structure, natural linewidths, and laser bandwidth would need to be incorporated in the analysis for an accurate value.

In the present experiments highly resolving laser saturation spectroscopy is, for the first time, applied to atomic resonance lines. The hyperfine splitting of the  $3^2P_{1/2}$  and  $3^2P_{3/2}$  states of Na<sup>23</sup> is resolved and measurements with a delayed probe permit novel studies of elastic atomic collision processes. More detailed investigations with the present technique are appropriate to reveal a possible difference in elastic-scattering behavior for atoms in the two hyperfine levels of the Na ground state. Measure-

ments with different light polarization can yield information on correlations between velocity-changing and disorienting collisions.<sup>3</sup> The remanent hole burning in the ground-state levels should permit the construction of ultranarrow-band atomic absorption filters. Moreover it is demonstrated with the experiment described here that it is now possible, by using narrow-band tunable dye lasers, to apply the powerful techniques of laser saturation spectroscopy in principle to any atomic or molecular transition even between highly excited states throughout the visible spectrum.

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