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Sub-Doppler Spectroscopy of Xenon Laser Lines with a Saturated-Amplification Method. Resolution up to the Natural Width

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A saturated-amplification method has been applied to the spectroscopic study of several infrared laser lines of xenon. The experiment is characterized by the use of two lasers. Linewidth at very low pressure, isotope shift, and hyperfine structure have been measured.

We report here ultrahigh-resolution measurements of atomic structures by means of a two-laser saturated-amplification method. Saturated-absorption techniques were first extensively used in molecular spectroscopy for which homogeneous widths are very weak[1,2]; in atomic spectroscopy, although lifetimes of excited levels are generally short and induce larger homogeneous widths, the gain in effective resolution remains important and a few experiments have been performed.[1,3,4]

We have studied several laser lines of xenon in the near infrared. For these lines, we have observed saturated absorption at high pressures but also saturated amplification at low pressures; both situations are fundamentally equivalent in their principle. In order to obtain the best possible resolution, we have systematically performed the experiments at pressures as low as possible, i.e., in saturated amplification; then the homogeneous width is nearly equal to the natural one and is very weak compared to

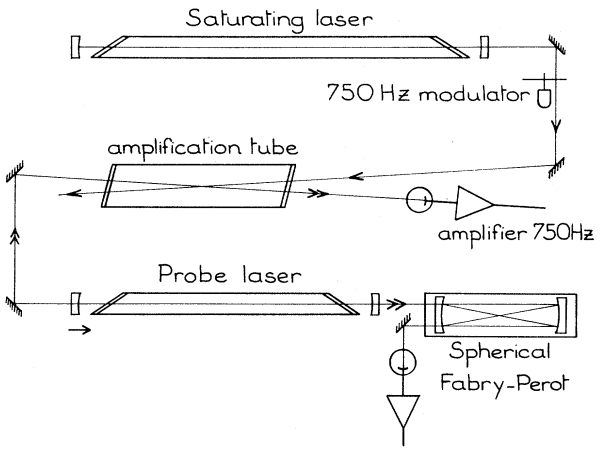

FIG. 1. Simplified block diagram of the experiment.

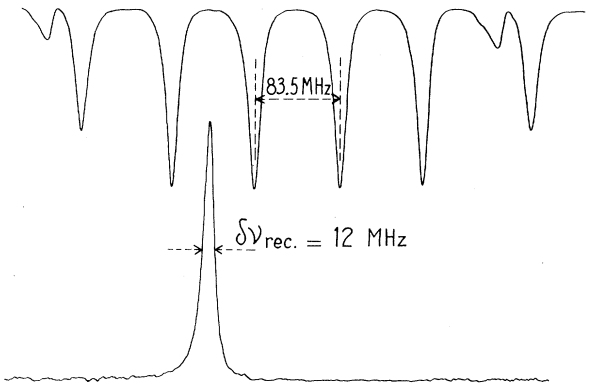

FIG. 2. $\lambda = 3.51\ \mu$m. Linewidth measurements. Typical recording obtained for a pressure $\simeq 3\times10^{-3}$ Torr, discharge current = 25 mA, amplification rate $\simeq 20\%$, and saturation rate $\simeq 0.25$.

the Doppler one. To improve the versatility of the experiment we have used two lasers[5,6]; the first one provides the saturating beam and the second one provides the probe beam. The frequency independence of the two beams allows the study of atoms for which the longitudinal velocity is nearly equal to zero (as in one-laser saturated-absorption experiments) as well as the study of atoms for which the longitudinal velocity is different from zero[7]; furthermore, the low power which is suitable for the probe beam allows the use of a short laser and yields a relatively large scanning range.

A schematic diagram of the setup is shown in Fig. 1. For the measurements reported here, the saturating laser is of single mode and stabilized at the Lamb dip. The probe laser is tunable over a few Doppler widths; its intensity is stabilized to a constant value during the scanning.[8] An external Fabry-Perot etalon is used to calibrate the frequency scale. The two beams travel in opposite directions along the amplification tube in the central part of which they are superposed. As in other experiments, the saturating beam is chopped by a mechanical modulator; a lockin detector is used to record only the modulated part of the probe beam which is induced by nonlinear effects in the amplification tube. As is well known, at low saturation power, the resonance signal has a Lorentzian profile and, in one-laser experiments, its width is nearly equal to the homogeneous one. In our experiment for which only the frequency of the probe beam is tunable, the recorded width is nearly twice the homogeneous one; however, the effective resolution is not affected since all structures are also expanded by the same factor.

In order to test the quality of our experiment, we have performed three kinds of measurements on infrared lines of Xe I.

Linewidth measurements ($\lambda = 3.51\ \mu$m; $5p^5 5d[\frac{7}{2}]_3 \rightarrow 5p^5 6p[\frac{5}{2}]_2$).—We have recorded the saturation signal of a pure even isotope at pressures as low as possible in order to reduce the broadening by collisions. The measurements have been done using 99% pure ^{136}Xe in the amplification tube as well as in the two lasers. A typical recording is shown in Fig. 2. The lower trace shows the homogeneous profile; the upper trace shows Fabry-Perot fringes whose spectral interval is 83.5 MHz. The constant amplitude of the fringes shows the intensity stabilization of the probe laser and indicates the limits of the scanning range which here is broadened up to 300 MHz by adding helium into the probe laser; consequently the scanning range is not centered on the saturation profile.

Various recordings have been performed for different saturation and amplification rates. The smallest recorded width, $\delta\nu_{rec} = 10.5 \pm 1$ MHz, corresponds to a pressure of xenon approximately equal to 10^{-3} Torr. The homogeneous width $\delta\nu_{hom}$ (natural width increased by collision effects) can be deduced from the following expression[9,10]:

$$\delta\nu_{hom} = \delta\nu_{rec}[1 + (1+\chi)^{1/2}]^{-1},$$

where χ is the saturation parameter[10] which here is evaluated as 0.25. This leads to $\delta\nu_{hom} = 5.0 \pm 0.5$ MHz. The natural width $\delta\nu_{nat}$ which can be

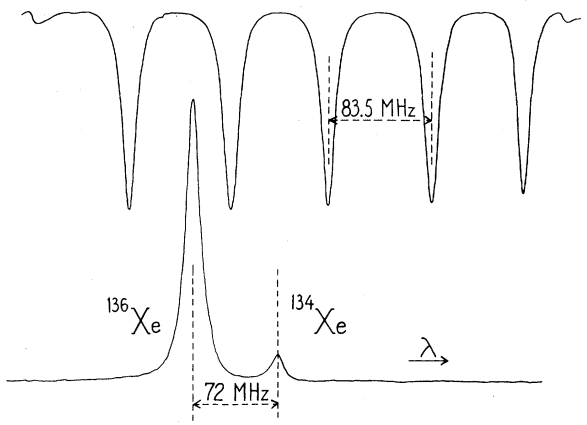

FIG. 3. $\lambda = 3.51\ \mu$m. Isotope shift between ^{136}Xe and ^{134}Xe.

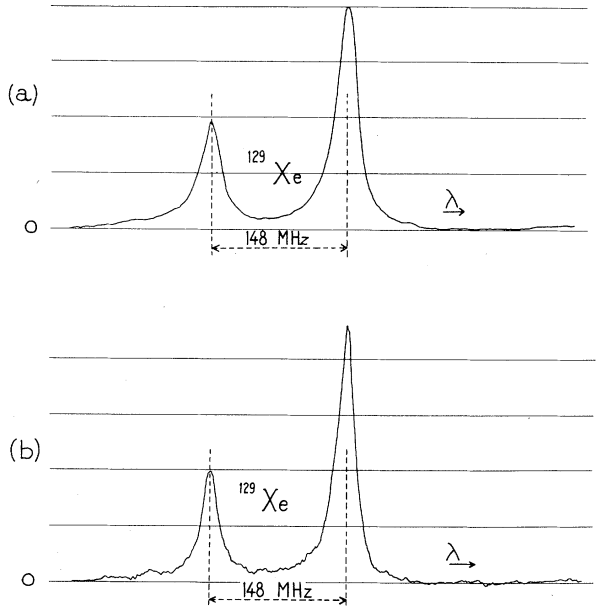

FIG. 4. $\lambda = 3.36\ \mu$m. Hyperfine structure of ^{129}Xe ($I = \frac{1}{2}$) for two saturation rates: (a) $\chi \simeq 4$ and (b) $\chi \simeq 0.5$. Notice the variation of relative intensities.

deduced from lifetime measurements and theoretical evaluations[11-13] is $\delta\nu_{nat} = 4.6 \pm 0.7$ MHz. The small difference between $\delta\nu_{hom}$ and $\delta\nu_{nat}$ is not significant and indicates that, at such low pressures, one practically reaches the natural width.

Isotope-shift measurements ($\lambda = 3.51\ \mu$m).—These have been done with a mixture of ^{136}Xe (84%) and ^{134}Xe (15%) in the amplification tube and pure ^{136}Xe in the two lasers. A typical recording is shown in Fig. 3; it has been obtained under the same conditions as the linewidth measurements. The lower trace shows the structure. Here the frequency of the saturating beam corresponds to the center of the Doppler profile related to ^{136}Xe and to the wings of the profile related to ^{134}Xe; consequently the intensity of the ^{134}Xe component is reduced. When this effect is taken into account, the ratio of intensities is equal to the ratio of isotopic abundances. We have verified that this result is independent of the saturation parameter. The recorded value of the structure is 72 MHz corresponding to the isotope shift

$$\delta\nu(136, 134) = 36.0 \pm 0.6 \text{ MHz}.$$

This direct measurement, which is allowed by the high resolution of the experiment, is in good agreement with a value deduced from earlier measurements: 37.5 ± 2.5 MHz.[14]

Hyperfine-structure studies ($\lambda = 3.36\ \mu$m; $5p^5 5d[\frac{5}{2}]_2 \rightarrow 5p^5 6p[\frac{3}{2}]_1$).—For hyperfine studies, we have used 98% pure ^{129}Xe in the amplification tube as well as in the two lasers. For the line at 3.36 μm, earlier measurements and calculations[15-17] indicate that the hyperfine constant of the upper level is not well determined. Typical recordings of the structure are shown in Fig. 4; only the two intense components can be recorded because the frequency of the saturating beam is largely outside the Doppler profile of the third component. Normally the measured structure is hidden by Doppler broadening and cannot be obtained by means of a classical experiment.

The value of the splitting is $\sigma\nu((\frac{5}{2},\frac{3}{2})-(\frac{3}{2},\frac{1}{2})) = -74 \pm 1.5$ MHz. If one assumes, in accord with the theoretical results proposed in Refs. 16 and 17, that the hyperfine constant of the lower level is $A(5p^5 6p[\frac{3}{2}]_1) = -1320$ MHz, our measurements yield a value for the constant of the upper level of $A(5p^5 5d[\frac{5}{2}]_2) = -822$ MHz, in agreement with Ref. 17.

The two recordings shown in Fig. 4 have been done for the first one at high saturation ($\chi \simeq 4$), and for the second one at low saturation ($\chi \simeq 0.5$); one can notice the broadening by saturation and overall the evolution of relative intensities; as already quoted,[18] the weaker component is reduced at low saturation.

Conclusion.—In this class of experiments, one can perform ultrahigh-resolution atomic spectroscopy measurements on excited levels. The preliminary results which we report here show that the resolution of our experiment is practi-

cally limited only by the natural width of the transitions. This resolution has been obtained with high signal-to-noise ratios.

Linewidth, isotope shift, and hyperfine structure have been measured; we plan now to study Zeeman patterns at low magnetic fields and pressure effects at very low pressures. Furthermore we plan to apply the experiment to the study of atoms with large longitudinal velocities and to the study of three-level systems, studies which are allowed by the use of two-laser saturated-amplification techniques.

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Measurement of the Stark Effect in Sodium by Two-Photon Spectroscopy*

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We have observed the Stark effect in the $5s\,^2S_{1/2}$ and $4d\,^2D_{3/2,5/2}$ levels of sodium. Shifts as well as splittings were measured by use of two-photon spectroscopy in a short atomic beam. It was possible to separate uniquely the contributions from the neighboring p and f levels. The polarizabilities were found to be, in MHz/(kV/cm)^2, (a) $5s\,^2S_{1/2}$, $\alpha_0 = 5.2(3)$, (b) $4d\,^2D_{3/2}$, $\alpha_0 = 155.3(1.7)$ and $\alpha_2 = -38.5(7)$, and (c) $4d\,^2D_{5/2}$, $\alpha_0 = 156.1(1.3)$ and $\alpha_2 = -53.2(5)$. From this the oscillator strengths $f_{4d\to 5p} = 0.274(29)$ and $f_{4d\to 4f} = 0.018\,85(24)$ are obtained.

In this Letter we report the first direct observation of Stark shifts and splittings in highly excited states of the alkalis with resolution high enough to observe the individual fine-structure components. The $4d$ or $5s$ level in sodium was excited by use of a two-photon process which eliminates Doppler broadening and allows the fine structure or hyperfine structure to be resolved.[1,2] It was possible to use comparatively small electric fields to produce an easily resolved Stark splitting or shift of these levels. Absolute frequency shifts were then measured by use of a thermally isolated 1.2-m confocal interferometer as a frequency reference. Previous work utilizing the level-crossing technique in crossed electric and magnetic fields[3] and in pure electric fields[4] has yielded information about differential Stark splittings. Highly excited states have been studied with a two-step process.[5] In the present work, however, both the Stark shifts and the splittings may be obtained. This additional information allows the contributions from the p and f states to be uniquely separated in the Stark shifts of the $4d$ level. Also states such as the $5s$, which