Doppler-Free Laser Polarization Spectroscopy*

C. Wieman† and T. W. Hänsch
Department of Physics, Stanford University, Stanford, California 94305
(Received 25 March 1976)

We have demonstrated a sensitive new method of Doppler-free spectroscopy, monitoring the nonlinear interaction of two monochromatic laser beams in an absorbing gas via changes in light polarization. The signal-to-background ratio can greatly surpass that of saturated absorption. Polarization spectra of the hydrogen Balmer-β line, recorded with a cw dye laser, reveal the Stark splitting of single fine-structure components in a Wood discharge.

We report on a sensitive new technique of high-resolution laser spectroscopy based on light-induced birefringence and dichroism of an absorbing gas. Polarization spectroscopy is related to saturated-absorption[1] or saturated-dispersion[2] spectroscopy, but offers a considerably better signal-to-background ratio. It is of particular interest for studies of optically thin samples or weak lines and permits measurements even with weak or fluctuating laser sources. We have studied the Balmer-β line of atomic H and D near 4860 Å by the new method, using a single-frequency cw dye laser. The spectra reveal for the first time the Stark splitting in the weak axial electric field of a Wood-type gas discharge.

For saturation spectroscopy it is well known that the signal magnitude depends on the relative polarization of saturating beam and probe.[3,4] The possibility and advantage of using the resulting optical anisotropy in a sensitive polarization detection scheme seem to have gone unexplored, however. On the other hand, the phenomena of light-induced birefringence and dichroism are quite common in optical-pumping experiments with incoherent light sources.[5] Recent related experiments[6,7] encourage us to expect that high-resolution polarization spectroscopy will also prove useful for studies of two-photon absorption and stimulated Raman scattering in gases.

The scheme of a polarization spectrometer is shown in Fig. 1. A linearly polarized probe beam from a monochromatic tunable laser is sent through a gas sample, which is shielded from external magnetic fields to avoid Faraday rotation. Only a small fraction of this beam reaches a photodetector after passing through a nearly crossed linear polarizer. Any optical anisotropy which changes the probe polarization will alter the light flux through the polarizer and can be detected with high sensitivity. Such an anisotropy can be induced by sending a second, circularly polarized, laser beam in nearly the opposite direction through the sample. In the simplest case both beams have the same frequency ω and are generated by the same laser. As in conventional saturation spectroscopy, a resonant probe signal is expected only near the center of a Doppler-broadened absorption line where both beams are interacting with the same atoms, those with essentially zero axial velocity.

For a quantitative description we can decompose the probe into two circularly polarized beams, rotating in the same (+) and in the opposite (−) sense as the polarizing beam. As long as the probe is weak these two components can be considered separately. The polarizing beam in general induces different saturation, i.e., changes in absorption coefficient, $\Delta\alpha^+$ and $\Delta\alpha^-$, and in refractive index, Δn^+ and Δn^-, for these components. A difference $\Delta\alpha^+ - \Delta\alpha^-$ describes a circular dichroism which will make the probe light elliptically polarized, and a difference $\Delta n^+ - \Delta n^-$ describes a gyrotropic birefringence which will rotate the axis of polarization. As long as these polarization changes are small, the complex field amplitude behind the blocking polarizer is given by

$$E = E_0[\theta + (\omega/c)(\Delta n^+ - \Delta n^-)l/2 - i(\Delta\alpha^+ - \Delta\alpha^-)l/4], \tag{1}$$

where E_0 is the probe amplitude, θ is some small angle by which the polarizer is rotated from the

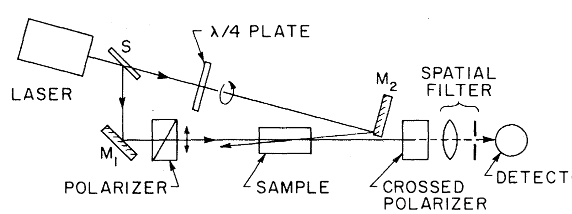

FIG. 1. Scheme of laser polarization spectrometer.

perfectly perpendicular position, and l is the absorption path length.

For low intensities, within third-order perturbation theory[8] and in the limit of large Doppler widths, the interaction of two counterpropagating circularly polarized beams with two levels of angular momentum J and J' can be described simply in terms of a velocity-selective hole burning[1] for the different degenerate sublevels of orientational quantum number m, where the axis of quantization is chosen along the direction of propagation. As in conventional saturation spectroscopy[1] the absorption change as a function of the laser frequency is a Lorentzian function with the natural linewidth γ_{ab}:

$$\Delta\alpha^+ = \Delta\alpha^-/d = -\tfrac{1}{2}\alpha_0 I/I_{\text{sat}}(1+x^2). \qquad (2)$$

Here, α_0 is the unsaturated background absorption, I is the intensity of the polarizing beam, I_{sat} is the saturation parameter, and $x=(\omega-\omega_{ab})/\gamma_{ab}$ describes the laser detuning from resonance. The absorption change corresponds to the imaginary part of a complex third-order susceptibility, whose real part results in a concomitant change in refractive index, $\Delta n^{\pm} = -\frac{1}{2}\Delta\alpha^{\pm} xc/\omega$, in agreement with the Kramers-Kronig relation.

The magnitude of the anisotropy is described by the parameter d and depends on the angular momenta and the decay rates γ_J and $\gamma_{J'}$ of the involved states. If spontaneous re-emission into the lower state is ignored, the steady-state signal contributions of the different orientational sublevels can be added with the help of simple sum rules[4] to yield

$$d \equiv \Delta\alpha^-/\Delta\alpha^+ = \begin{cases} 1-5/(4J^2+4J+2) & \text{for } J=J', \\ (2J^2+5rJ+3)/(12J^2-2) & \text{for } J=J'+1, \end{cases} \qquad (3)$$

where $r=(\gamma_J-\gamma_{J'})/(\gamma_J+\gamma_{J'})$.

By inserting these results into Eq. (1), we obtain the light flux at the probe detector

$$I = I_0[\theta^2 + \theta\tfrac{1}{2}sx/(1+x^2) + (\tfrac{1}{4}s)^2/(1+x^2)], \qquad (4)$$

where I_0 is the unattenuated probe power and $s = -\frac{1}{2}(1-d)\alpha_0 lI/I_{\text{sat}}$ gives the maximum relative intensity difference between the two counter-rotating probe components. In practice we have to add an amount $I_0\xi$ to account for the finite extinction ratio ξ of the polarizer.

For a perfectly crossed polarizer, i.e., $\theta=0$, the combined effects of dichroism and birefringence lead to a Lorentzian resonance signal. The signal magnitude is proportional to s^2, i.e., it drops rapidly for small s. Small signals can be detected with higher sensitivity at some small bias rotation $\theta \gg s$. The last term in Eq. (4) can then be neglected and the birefringent polarization rotation produces a dispersion-shaped signal on a constant background. If, as in many practical situations, laser intensity fluctuations are the primary source of noise, a figure of merit for the sensitivity is the signal-to-background ratio. Compared to saturated-absorption spectroscopy, this ratio is improved by a factor $(1-d)\theta/4(\theta^2+\xi)$ which reaches its maximum $(1-d)/8\sqrt{\xi}$ for $\theta=\sqrt{\xi}$.

In addition to the improved sensitivity, such a dispersion-shaped signal is of obvious interest for the locking of the laser frequency to some resonance line. It is also noteworthy that its first derivative has a linewidth smaller than half the natural width, which can greatly facilitate the spectroscopic resolution of closely spaced line components.

For the alternative scheme of a linearly polarized saturating beam, rotated 45° with respect to the probe polarization, it can be shown in analogous fashion that the signal always remains Lorentzian, independent of the polarizer angle θ.

As in saturated-absorption spectroscopy, crossover signals are expected halfway in between two resonance lines which share a common upper or lower level. A third-order nonlinear susceptibility tensor, applicable to this situation, has actually been calculated previously.[3] It predicts that the ratio $\Delta\alpha^-/\Delta\alpha^+=d$ for certain angular momentum states can exceed 1, unlike the expression (3), and hence give rise to signals with inverted polarization rotation.

For the experimental study of the hydrogen Balmer-β line we used a cw jet-stream dye laser (Spectra-Physics Model No. 375) with 7-diethylamino-4-methyl-coumarin in ethylene glycol, pumped by an uv argon laser (Spectra-Physics Model No. 171). Single-frequency operation was achieved with an air-spaced intracavity etalon (free spectral range 30 GHz; mirror reflectivity 30%) and two additional fixed uncoated quartz etalons (thicknesses 0.1 and 0.5 mm). At 1 W pump power, the laser provides single-mode output of about 10 mW near 4860 Å with a linewidth of about 20 MHz. The laser can be scanned continuously over ~4 GHz by applying linear ramp voltages to the piezotransducers of the cavity end mirror and the air-spaced etalon.

As in the previous saturated-absorption experiment,[9] the hydrogen atoms were excited to the absorbing $n = 2$ state in a Wood-type discharge tube (1 m long, 8 mm in diameter, 0.2 Torr, 3 mA dc current). The laser light is sent through a 40-cm-long center section of the positive column. Probe and polarizing beam are each about 1 mm in diameter and have powers of 0.1 and 1 mW, respectively. A mica sheet serves as a $\lambda/4$ retarder for the polarizing beam. To avoid residual Doppler broadening due to a finite crossing angle we operated with collinear beams, replacing mirror M_2 in Fig. 1 with an 80% beam splitter which transmits part of the probe.

Standard Glan-Thompson prism polarizers (Karl Lambrecht) were employed for the probe beam. Birefringence in the quartz windows of the gas cell due to internal strain was reduced by squeezing the windows gently with adjustable clamps. We achieve extinction ratios of 10^{-7} or better in this way, and the possible improvement over saturated-absorption spectroscopy in signal-to-background ratio is on the order of 100–1000. The probe light which passes the blocking polarizer is sent through a spatial filter to eliminate incoherent light emitted by the gas discharge and scattered light from the polarizing beam. Its intensity is monitored with a photomultiplier.

Figure 2 shows a portion of the Balmer-β spectrum plotted versus time during a laser scan of about 5 min duration. To record the derivative of dispersion-shaped resonances, the dye laser was frequency modulated by adding a small audio-frequency voltage to the cavity-mirror tuning ramp, and the resulting signal modulation was detected with a phase-sensitive amplifier. The three strongest theoretical fine-structure transitions in this region are shown on top for comparison. Hyperfine splitting is ignored. The positions of possible crossover lines due to a common upper or lower level are indicated by arrows. Obviously, the polarization spectrum reveals many more components. These have to be ascribed to the Stark splitting in the axial electric field of the positive discharge column.

We have calculated the theoretical Stark pattern for an axial field of 10 V/cm by diagonalizing the Hamiltonian.[10] The positions of the strongest Stark components and their respective crossover lines are indicated in Fig. 2 and agree well with

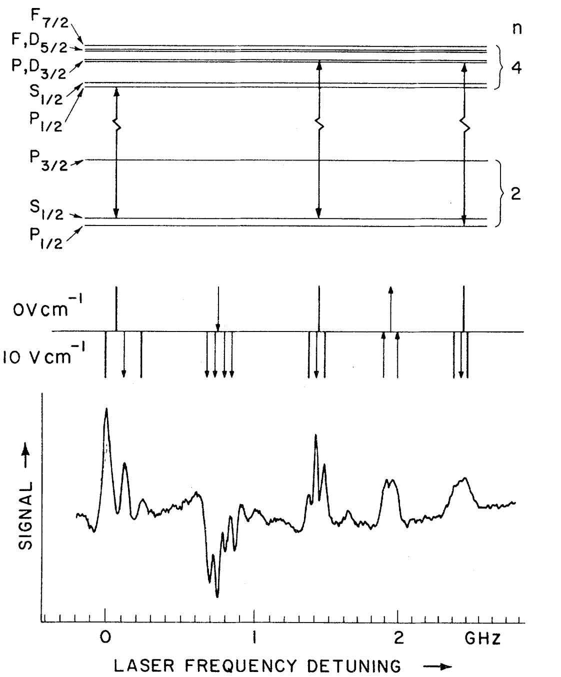

FIG. 2. Polarization spectrum of a portion of the deuterium Balmer-β line. The three strongest fine-structure components and the positions of the strongest Stark components for an axial electric field of 10 V/cm are shown on top for comparison. Crossover lines due to a common upper (↑) or lower (↓) level are indicated by arrows.

the observed spectrum. The splitting of the upper $4P,D_{3/2}$ level is essentially a linear function of the electric field, and its magnitude can easily be determined to within a few percent from the observed spectrum. The theoretically expected line strengths and the signs of the crossover lines are also in satisfactory agreement with the experiment. The polarization spectrum of light hydrogen looks almost identical to that of deuterium and shows no indication for the 170-MHz hyperfine splitting of the $2S$ state; the components originating in the $F = 0$ state are missing, because atoms in this level cannot be oriented.

The observed Stark pattern changes quite drastically if the laser beams are displaced from the tube axis, indicating the presence of additional radial electric fields due to space and surface charges of cylindrical symmetry. Polarization spectroscopy of the hydrogen Balmer lines thus opens new possibilities for sensitive plasma diagnostics.

The spectrum in Fig. 2 also clearly reveals the different natural linewidths of components originating in the short-living $2P$ state and the longer-living $2S$ state. The narrowest observed components have a width of about 40 MHz, corresponding to a resolution of about 6 parts in 10^8, i.e., they exhibit more than an order of magnitude improvement over our earlier pulsed-laser saturation spectra.[9] A substantial improvement in the accuracy of the $1S$ Lamb shift is expected, if such a polarization spectrum is used as a reference for the $1S$-$2S$ two-photon spectrum.[9] A still further improvement in resolution should be possible if the laser linewidth is reduced by frequency stabilization. At low electric fields the natural linewidth of the quasi-forbidden $2S$-$4S$ transition is only about 1 MHz. A measurement of the H-D isotope shift to better than 0.1 MHz would confirm or improve the important ratio of electron mass to proton mass, and an absolute wavelength or frequency measurement to better than 6 MHz would yield a new improved value for the Rydberg constant. We are presently exploring these and other possibilities for new precision measurements.

We are indebted to Professor A. L. Schawlow for his stimulating interest in this work, and we thank J. Eckstein for his help in calculating Eq. (3).

*Work supported by the National Science Foundation under Grant No. NSF 14786, and by the U. S. Office of Naval Research under Contract No. N00014-75-C-0841.

†Hertz Foundation Predoctoral Fellow.

[1]P. W. Smith and T. W. Hänsch, Phys. Rev. Lett. 26, 740 (1971).

[2]C. Borde, G. Camy, B. Decomps, and L. Pottier, C. R. Acad. Sci., Ser. B 277, 381 (1973).

[3]T. W. Hänsch and P. Toschek, Z. Phys. 266, 213 (1970).

[4]M. Sargent, III, M. O. Scully, and W. E. Lamb, Jr., *Laser Physics* (Addison-Wesley, London, 1974).

[5]W. Happer, Prog. Quantum Electron. 1, 53 (1970).

[6]P. F. Liao and G. C. Bjorklund, Phys. Rev. Lett. 36, 584 (1976).

[7]D. Heiman, R. W. Hellwarth, M. D. Levenson, and G. Martin, Phys. Rev. Lett. 36, 189 (1976).

[8]M. Dumont, thesis, University of Paris, 1971 (unpublished).

[9]S. A. Lee, R. Wallenstein, and T. W. Hänsch, Phys. Rev. Lett. 35, 1262 (1975).

[10]J. A. Blackman and G. W. Series, J. Phys. B 6, 1090 (1973).

Analogy between the Laser and Second-Order Phase Transitions: Measurement of "Coexistence Curve" and "Susceptibility" for a Single-Mode Laser near Threshold*

Mario Corti† and Vittorio Degiorgio‡
Centro Informazioni Studi Esperienze, Segrate, Milano, Italy

(Received 21 January 1976)

We report the first experimental determination of the laser "coexistence curve" and "susceptibility" below threshold. Both quantities are derived from an accurate measurement of the average output intensity as a function of the normalized net gain. Our data confirm the predictions of laser theory and give a quantitative support to the recently proposed analogy between the laser near threshold and second-order phase transitions.

It has been recently shown that the theoretical treatment of the threshold region of a laser bears a close analogy with the mean-field approach to a second-order phase transition, if one identifies the order parameter with the slowly varying amplitude of the laser electric field and the temperature with the unsaturated population inversion.[1,2] We report here the first measurement of the laser "coexistence curve" and "susceptibility" below threshold. Our data confirm the predicted mean-field-theory behavior of the laser transition in the threshold region.

The statistical properties of single-mode laser radiation near threshold have been extensively investigated in the past few years.[3] Various moments of the photon probability distribution and time-dependent correlations of both amplitude and phase fluctuations have been measured as functions of the average output intensity I of the laser. Our experiment concerns instead the dependence of I on the difference between the unsaturated population inversion σ and the threshold population inversion σ_t.

From the viewpoint of the proposed analogy, the quantity I does not always have the same meaning. Specifically, in the region above threshold where amplitude fluctuations are relatively small, I is the square of the laser order param-