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Polarization-modulated optical pumping spectroscopy of Na D_1 and D_2 lines

Jom Sool Kim, Hong Jin Kong, and Sang Soo Lee
Department of Physics, Korea Advanced Institute of Science and Technology, P.O. Box 150, Chongryang, Seoul 130-650, Korea

Jung Bog Kim
Department of Theoretical Physics and Chemistry, Korea Advanced Energy Research Institute, P.O. Box 7, Daeduk-danji, Choong-Nam, Korea

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Doppler-free spectra of linear and circular dichroisms for the hyperfine structures of Na D_1 and D_2 lines have been obtained by means of polarization-modulated optical pumping spectroscopy. The experimental results are compared with recent theoretical calculations based on velocity-selective optical pumping in a four-level atomic system.

I. INTRODUCTION

Velocity-selective optical pumping (VSOP) is an important technique for obtaining Doppler-free spectral resolution. This technique utilizes the optical pumping effect of a low-intensity light field on long-lived atomic states in combination with the high spectral resolution of a single-mode laser beam.[1–12] As in ordinary optical pumping,[13] optical anisotropy is induced by a pump laser beam. However, strong correlations between the velocities of atoms and their internal variables (orientation, alignment, etc.) are introduced by the selection of a single group of atomic velocity in the pumping process. When a weak counter-propagating probe beam is tuned so that it interacts with the pumped atoms, its polarization and intensity are modified. Consequently, by monitoring these probe-beam properties one can obtain information on the velocity distribution of the anisotropy.

Recently, the Doppler-free relative spectral intensities of the D_1 and D_2 lines of alkali-metal atoms were calculated by Nakayama, taking into account the rate equation and VSOP process in a four-level atomic system and arbitrary polarization configurations for the pump and probe beams. In his theory the four-level approximation was proposed as a method of calculating the optical anisotropy by selecting two-level principal resonance and three- and four-level crossover resonances among many Zeeman sublevels of the hyperfine levels.[14,15] In this paper we report the experimental results of polarization-modulated optical pumping spectroscopy of the Na D_1 and D_2 lines, and the resulting Doppler-free spectra of linear and circular dichroisms are compared with the theoretical calculations of Nakayama.

In the polarization-modulated optical pumping technique, the Doppler-free spectra of linear or circular dichroism of an absorbing gas can be obtained by using polarization modulation and phase-sensitive detection.[16] When the probe beam is linearly polarized ($\pi \equiv 0$) and the pump beam is modulated between parallel ($\pi \equiv 0$) and perpendicular ($\sigma^{\pm} \equiv \pm$) polarization, the probe beam experiences different absorption coefficients α_0^0 and $\alpha_{\pm}^0$ in the anisotropic gas. The phase-sensitive detection of the intensity modulation in the transmitted probe beam then gives rise to the signal proportional to $\alpha_0^0 - \alpha_{\pm}^0$, which shows linear dichroism. Similarly, probing with a circular polarization ($\sigma^+ \equiv +$) and modulating the pump beam between left ($\sigma^+ \equiv +$) and right ($\sigma^- \equiv -$) circular polarization yields the signal proportional to $\alpha_+^+ - \alpha_-^+$, which shows circular dichroism.

II. EXPERIMENTAL PROCEDURE

The experimental setup used is shown in Fig. 1. The dye laser (Spectra-Physics 375) with rhodamine-6G dye, pumped by an Ar^+ laser (Spectra-Physics 165, 4 W), produces up to about 20 mW of single-mode radiation tunable over several GHz with a linewidth of 10 MHz. The dye-laser frequency is stabilized to the maxima of a transmission fringe of a stable, high-finesse reference in-

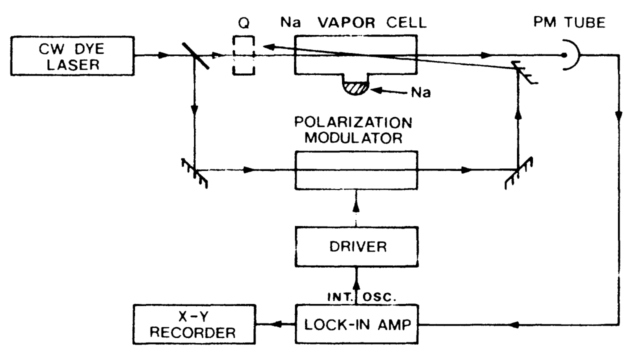

FIG. 1. Experimental setup for polarization-modulated optical pumping spectroscopy. When the probe beam is linearly (π) polarized, the pump beam is modulated between parallel (π) and perpendicular ($\sigma^{\pm}$) polarization by the electro-optic modulator. In the case where the probe beam is circularly (σ^+) polarized with the insertion of a $\lambda/4$ wave plate (Q), the pump beam is modulated between left (σ^+) and right (σ^-) circular polarization.

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terferometer, and the power is also stabilized by controlling the Ar^+ laser pump power. In front of a beam splitter, a Glan-Thompson prism polarizer is inserted into the laser output beam to ensure a well-defined polarization. The probe beam is thus linearly polarized in the absence of a $\lambda/4$ wave plate (Q), whereas circularly polarized through the Q. The pump beam polarization is modulated by an electro-optic modulator (Coherent Inc. 20). The modulator is driven at a frequency of 1 KHz by a square-wave voltage from the internal oscillator output of a lock-in amplifier (Stanford Research 530). The voltage amplitude is carefully adjusted to produce either half-wave retardation, corresponding to a modulation between π and $\sigma^{\pm}$ polarization or quarter-wave retardation, corresponding to a modulation between σ^+ and σ^- circular polarization. Throughout the present experiment, the pump and probe beams are adjusted to 2 and 1 mm in diameter, and 12 and 1.2 $\mu W/mm^2$ in intensity, respectively, and the angle between their propagation directions is maintained at less than 5 mrad.

The sodium is contained in a cylindrical pyrex cell, 25 mm in diameter, which has been evacuated to 10^{-7} torr before sealing off. No buffer gas is introduced. Heating coils are carefully counterwound around the cell to minimize magnetic field during heating. The cell is kept at a temperature of 150 °C. After passing the cell the probe beam is detected by a photomultiplier tube, and the signal taken is then fed into the lock-in amplifier. The resulting signal is recorded on a x-y recorder as a function of laser frequency. The frequency of the scan is calibrated by means of a confocal Fabry-Perot interferometer with a free spectral range of 150 MHz.

III. RESULTS AND DISCUSSION

Figure 2 shows the comparison of the experimental results and theoretical calculations for Doppler-free hyperfine signals of linear and circular dichroisms of the sodium D_1 line. The inset shows the relevant level structures with hyperfine splittings and resonances occurring between them, and the principal resonances are denoted as L and H, and the crossover resonances as X.[14] Each resonance in the theoretical curves is assumed to be a Lorentzian line shape with the linewidth of 16 MHz, considering the natural linewidth, the laser spectral width, and residual Doppler broadening. For the normalization of the signal intensities of the experimental and theoretical curves, L_1 is selected. Although Nakayama's VSOP theory is an approximation of the four-level system in low-intensity limit, the experimental results agree reasonably with the theoretical calculations. However, some discrepancies in the signal intensity are seen. For exam-

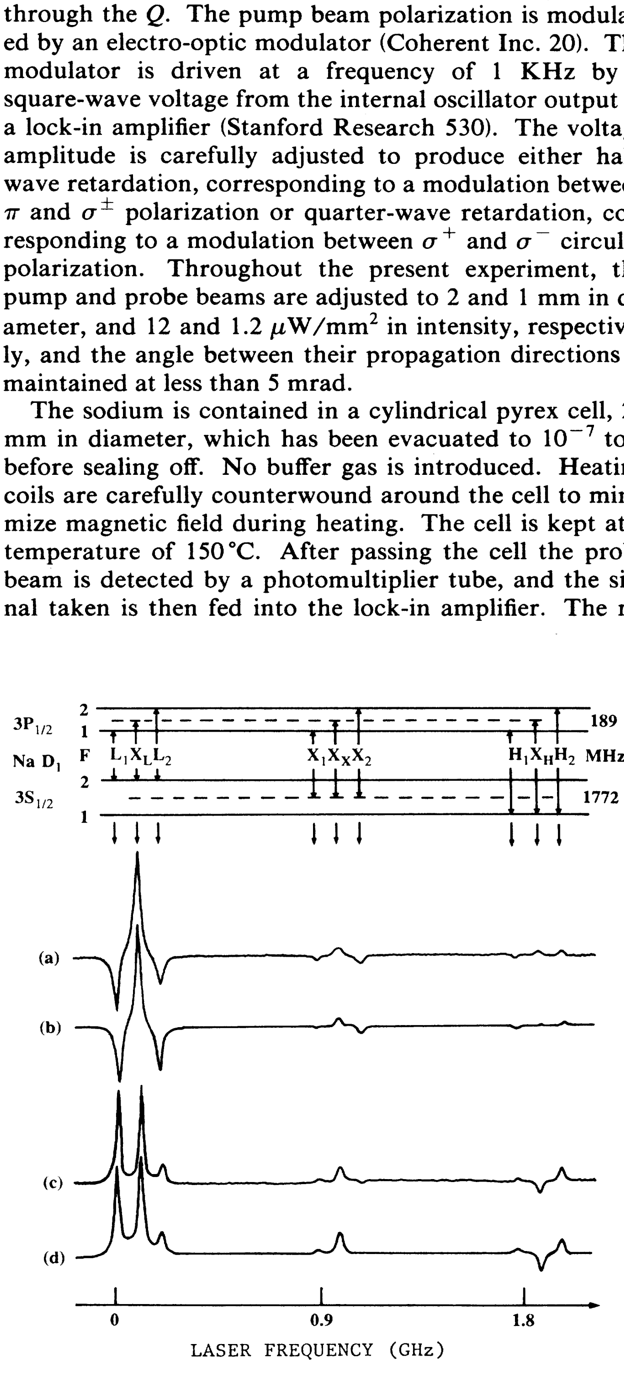

FIG. 2. Comparison of the experimental results and theoretical calculations (Ref. 14) for the D_1 line: the experimental (a) and theoretical (b) curves of linear dichroism, and the experimental (c) and theoretical (d) curves of circular dichroism. The inset shows the hyperfine structures and resonances of the D_1 line.

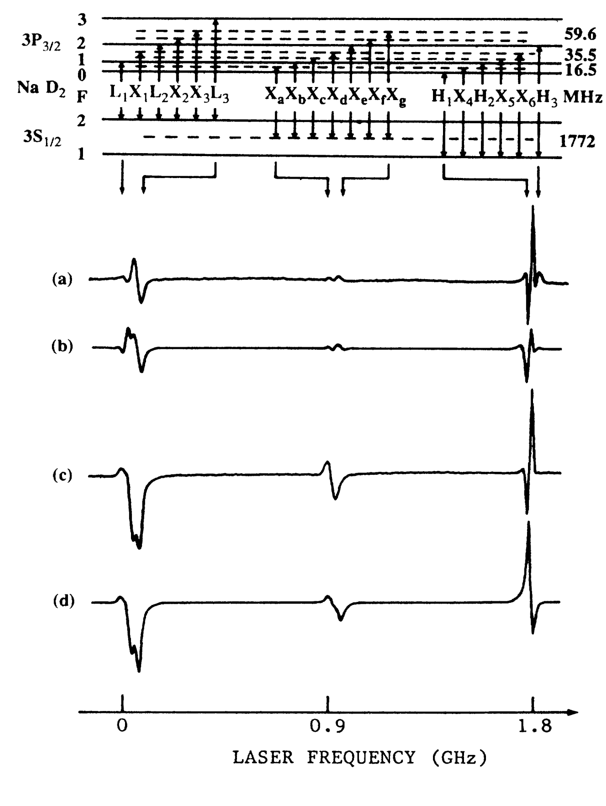

FIG. 3. Comparison of the experimental results and theoretical calculations (Ref. 15) for the D_2 line: the experimental (a) and theoretical (b) curves of linear dichroism, and the experimental (c) and theoretical (d) curves of circular dichroism. The inset shows the hyperfine structures and resonances of the D_2 line.

ple, X_H in Fig. 2(a) is observed larger than the theoretical prediction in Fig. 2(b), and X_2 in Fig. 2(c) is observed experimentally but is not predicted theoretically in Fig. 2(d).

Several causes of such discrepancies are considered. In fact the pump-beam intensity used in the present experiments is comparable with optical pumping saturation intensity ($I_0=2I_s\tau/T$, where τ is the radiative lifetime of the excited state, T is the light-atom interaction time, i.e., effective ground-state lifetime, and I_s is the usual saturation intensity with no optical pumping.[2] Since $\tau\sim 10$ nsec, $T\sim 1.8$ μsec, and $I_s\sim 64$ μW/mm^2, $I_0\sim 7$ μW/mm^2 for the D_1 line). So the contribution of a system of more than four levels to optical pumping and saturation effect should be considered. Such consideration is confirmed by the fact that the hyperfine signals of circular dichroism of the D_1 line experimentally obtained by Aminoff *et al.*[9] in the linear regime of optical pumping and with the sufficiently low intensity to avoid any saturation show excellent agreement with Nakayama's calculations. In addition, although the probe beam intensity is much weaker than the pump beam intensity, optical pumping effect of the probe beam alone cannot be neglected. The possibility is also considered that such discrepancies are somehow due to the effects of residual, inhomogeneous or slightly misaligned magnetic field and incomplete polarization of the pump and probe beams. In connection with the above considerations, the detailed analysis of optical pumping caused by both the pump and probe beam, as well as the probe beam alone as a function of the atomic transit time for various magnetic fields may be found in Ref. 4.

Figure 3 compares the experimental results with the theoretical calculations for the D_2 line. The signals consist of the six hyperfine (L_1-L_3 and H_1-H_3) and 13 crossover resonances (X_1-X_6 and X_a-X_g).[15] For the normalization L_3 is selected. The low- (L_1-L_3 and X_1-X_3) and center- (X_a-X_g) frequency side signals show reasonable agreement with the experiment and theory, but there is a large discrepancy in the intensity and sign for the high-frequency side signals (H_1-H_3 and X_4-X_6). Such discrepancy is considered to be the additional effect due to the excited hyperfine levels separated by only 16.5 or 35.5 MHz. As six resonances occur within the narrow width of 52 MHz of H_1 to H_3, they are separated by less than the natural or laser spectral linewidth. Therefore, even though the laser is tuned to a given transition, neighboring resonances can simultaneously occur, which is different from the crossover resonance due to the experimental geometry. In order to analyze the effect, the rate equation should be modified to include the case when two or more transitions share in common the same ground-state level.

In conclusion, we have reported the experimental results of polarization-modulated optical pumping spectroscopy of the sodium D_1 and D_2 lines. For each line, Doppler-free hyperfine spectra of linear and circular dichroisms are obtained. The experimental results agree reasonably with the theoretical calculations of Nakayama. A small discrepancy is considered to be the effect due to the use of the pump beam intensity comparable with optical pumping saturation intensity. However, there remains a large discrepancy for the high-frequency side signals of the D_2 line, which is primarily regarded as the additional effect resulting from several closely lying resonances separated by less than the natural or laser spectral linewidth.

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