Vol 10 No 8, August 2001
1009-1963/2001/10(08)/0716-04
Chinese Physics
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and IOP Publishing Ltd

PRECISE MEASUREMENT OF DENSITY-DEPENDENT SHIFT BY WAVELENGTH MODULATION REFLECTION SPECTROSCOPY*

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(Received 15 February 2001)

The density dependence of the line shift of the cesium D_2 line is studied with sub-Doppler selective reflection spectroscopy. By use of wavelength modulation and sixth-harmonics detection we observed the coefficient of the pressure-induced shift of the $6S_{1/2}(F=4) \rightarrow 6P_{3/2}(F'=3)$ hyperfine transition of the cesium D_2 line $\Delta\delta/\rho = -0.9(5) \times 10^{-8}\mathrm{cm}^3$. In the limit ρ=0 a frequency shift about −3MHz remains, which may be attributed to long-range atom–surface interactions. The experimental results can be used in measurements of the local field-induced frequency shift at high atomic densities with sufficient accuracy.

Keywords: selective reflection, sub-Doppler, wavelength modulation spectroscopy, harmonic detection

PACC: 3290

I. INTRODUCTION

The optical properties of dense atomic vapours have received notable attention in recent years, partly motivated by possible application in quantum electronics, such as intrinsic optical bistability.[1]

In dense atomic vapours, conventional absorption spectroscopy can be applied only to the wings of an atomic line because multiple scattering of resonance photons in the gas sample strongly distorts the line centre.[2] For an investigation of the line centre in optically dense vapour, one can use the Doppler-free frequency modulation selective reflection (SR) spectroscopy with the first-harmonic detection technique.[3−5] The self-broadening of the hyperfine structure (hfs) components of the D_2 line (λ=852nm) has been measured with this method.[3] Subsequent accurate studies of Doppler-free SR spectroscopy have demonstrated the need to take into account the influence of long-range atom–surface interactions on the resonance lines.[4] It has been shown that a van der Waals (VW)-type attraction potential is responsible for pressure-independent frequency redshifts and lineshape asymmetry of the SR resonances observed in Cs and Rb.[5] In this paper, we investigate the shift of the cesium D_2 line components induced by Cs–Cs collisions by use of wavelength modulation SR spectroscopy and harmonics detection. Here the SR resonance line is determined mainly by residual Doppler broadening. This technique allows the monitoring of the interaction between the atoms and the reflecting surface.

II. HARMONICS DETECTION METHOD

The principal differences between frequency modulation spectroscopy (FMS) and wavelength modulation spectroscopy (WMS)[6−8] are slight and the terminology is somewhat misleading since in both cases it is the optical frequency of the laser that is modulated. They can be understood as the two limiting cases of the same technique. In FMS, the modulation index of the laser is small, but the ratio of the modulation frequency to the absorption linewidth is large; as a result the absorption feature of interest is probed by a single isolated sideband. In WMS, the ratio of the modulation frequency to the absorption linewidth is small; but the modulation index of the laser is large.

When WMS is performed with small modulation indices, and detection is performed at a harmonic of the modulation frequency and the derivative spectra are obtained. For a Gaussian line shape the Nth

*Project supported by the National Natural Science Foundation of China (Grant No. 19774039, 60078009) and the Natural Science Foundation of Shanxi Province.

derivative, $g^N(\nu)$, can be written as

$$g^N(\nu) = \frac{g(\nu)}{\sqrt{\pi}} \frac{1}{(\overline{\Delta\nu})^{2N+1}} \cdot \sum_{K=0}^{[N]/2} C_{2K,N} (\overline{\Delta\nu})^{2K} (\nu - \nu_0)^{N-2K}, \quad (1)$$

where

$$C_{2K,N} = (-1)^K \frac{N!}{(N-2K)!} \frac{(-2)^{N-2K}}{K!}$$

$$\text{and} \quad [N] = \begin{cases} N-1 & \text{for} \quad N \quad \text{odd}, \\ N & \text{for} \quad N \quad \text{even}, \end{cases}$$

$$\text{and} \qquad g(\nu) = \exp\left(\frac{-(\nu-\nu_0)^2}{(\Delta\nu)^2}\right).$$

Here, $\Delta\nu$ is the Doppler width given by

$$\Delta\nu = 2\left(\frac{2\kappa T \ln 2}{Mc^2}\right)^{1/2} \nu_0, \quad \text{and} \quad \overline{\Delta\nu} = \frac{\Delta\nu}{\sqrt{4\ln 2}}.$$

Hence, using a combination of Fourier and Taylor series expansions, the signal detected in a wavelength modulation experiment employing Nth-harmonic detection, can be written as

$$S_N(\nu) = -I_0(\nu) L (-1)^{[N]/2} \sum_{\upsilon=0}^{\infty} \left(\frac{\beta}{2}\right)^{2\upsilon+N} \cdot \frac{1}{\upsilon!(\upsilon+N)!} g^{2\upsilon+N}(\nu). \quad (2)$$

Here, $I_0(\nu)$ is the intensity of the laser, which passes though a cell of length L filled with an absorbing gas. υ is an integer, $g^{2\upsilon+N}$ is the $(2\upsilon+N)$th derivative of the Gaussian line shape, and β is the magnitude of the frequency-modulation amplitude (in Hz). In the limit of a not very large modulation index $m = \beta/\Delta\nu$, the υ=0 term dominates and we can obtain a signal which is proportional to the Nth-derivative of the line shape function of the absorbing feature. Therefore, the υ=0 term leads to derivative spectroscopy.

The even-order harmonic signal peaks are at the absorption line centre frequency, but the odd-order harmonic signal values are then zero. Here we only use the even harmonic detection. We have demonstrated experimentally how the sensitivity can be increased by the use of harmonics detection.[7] It has also been shown that the application of higher-order harmonics detection can result in an increase in wavelength resolution.[6,8]

Ⅲ. EXPERIMENTAL SET-UP

A schematic diagram of the experimental arrangement is shown in Fig.1. A highly coherent external cavity diode laser (New Focus Model 6017) is used as a source of tunable radiation. The power of the collimated laser beam is about 2 mW, while the linewidth does not exceed 0.2 MHz. We modulate the laser frequency with 1 kHz. The laser beam (divergence∼2 mrad) was sent onto a cesium vapour cell in which the reflecting interface was formed by the optical glass window and the vapour.

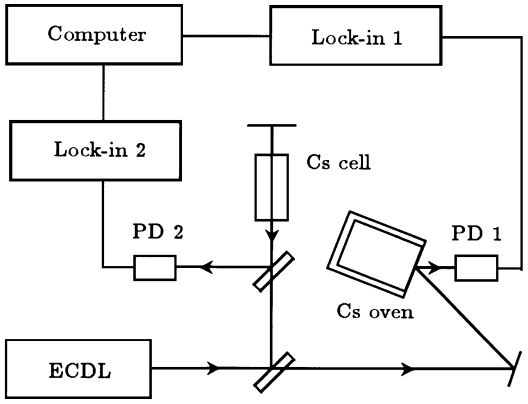


Fig.1. A schematic diagram of the experimental set-up.

The Cs-filled glass cell was set in an oven with two sections, one containing the body of the cell, the other the Cs metal reservoir. The temperature of the reservoir of the cell was measured and the Cs pressure was determined by using the Langmuir–Taylor formula.[9] For temperatures between 90°C and 160°C we obtain atom concentrations between 10^{13}cm^{-3} and $3\times10^{14}\text{cm}^{-3}$.

A reflected light beam was sent onto a photodetector, and the signal was demodulated with Nth-harmonics detection by a digitized lock-in amplifier (Model SR 830) and recorded with a PC for off-line analysis.

A saturated absorption spectrum of cesium vapour at room temperature was simultaneously recorded to provide a frequency reference.

To avoid nonlinear effects such as optical pumping, we work with low beam intensity ($0.2\,\text{mW/cm}^2$).

We use the cell with the incidence angle θ of 60 mrad. The width of the SR resonance is $\Gamma_{\min} \approx \gamma + \Delta\nu_{\text{D}}\theta \approx 35$ MHz and is determined mainly by residual Doppler broadening $\Delta\nu_{\text{D}}\theta \approx 30$ MHz. The reason that we do not use the normal incidence is that the observed line has an anti-symmetric dispersive

Lorentzian shape and the harmonic signal peaks are not exactly at the absorption line centre frequency.

IV. RESULTS

In the present work we study the frequency shift of the $6S_{1/2}(F{=}4) \rightarrow 6P_{3/2}(F'{=}3)$ hyperfine transition of the cesium D_2 line (wavelength λ=852nm, natural width $\gamma_r/2\pi$=5.3MHz). Random samples of other hfs components show essentially the same behaviour.

Figure 2 shows the experimental results (second-, fourth-, sixth-harmonic detection) obtained for the $6S_{1/2}(F{=}4) \rightarrow P_{3/2}(F'{=}3)$ hyperfine transition of the cesium D_2 at low density ($\rho{=}10^{14}\mathrm{cm}^{-3}$, collision broadening $\gamma_{\mathrm{col}} < \gamma_r$). It is seen that the peak is sharper when the higher harmonic is detected. We did not measure the higher-harmonic signals (eighth, tenth, ...) because the signal-to-noise ratios (SNRs) decrease with the increasing harmonic.[7] The resonance frequency of the unperturbed line has been obtained from the sixth harmonic of the saturation spectrum recorded simultaneously at very low cesium vapour density (room temperature). We examined the frequency shift of the SR line centre, measured relative to the resonance frequency.

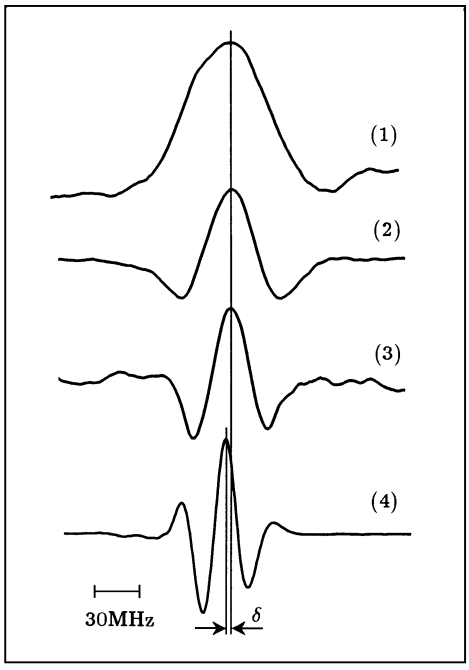


Fig.2. Measured $6S_{1/2}(F{=}4) \rightarrow 6P_{3/2}(F'{=}3)$ of Cs D_2 line with (1) second-, (2) fourth- and (3) sixth-harmonic reflection spectra at m=3.3. (4) is the sixth harmonic of the saturated absorption dip in an additional low-pressure cell. The oven temperature T is about 140°C.

In Fig.3 the observed shift δ_{SR} with the sixth-harmonic detection is plotted against the atomic density ρ. Since the contribution of the Lorentz shift is reduced by the ratio of homogeneous to Doppler width, the Lorentz shift is negligible compared to the collisional shift in the density region studied here ($\gamma/\Delta\nu_{\mathrm{D}} < 0.1$). As expected for the collision-induced shift, we find good agreement with a linear function, $\delta_{\mathrm{SR}} = \delta_0 + \alpha\rho$, where $\delta_0 = -(3.0 \pm 0.4)$MHz accounts for the shift that remains for $\rho \rightarrow 0$ and $\alpha = -(0.9 \pm 0.4) \times 10^{-8}\mathrm{cm}^3\mathrm{Hz}$ is the coefficient for the collisional shift. For the cesium D_2 line in the moderate density region ($\rho \leq 10^{15}\mathrm{cm}^{-3}$), Chevrollier *et al.*[4] have observed a density-dependent shift which did not vary systematically and which was different for different hyperfine components. The order of magnitude of the shift coefficient is the same as the one observed here. The redshift of approximately 3 MHz which remains for $\rho \rightarrow 0$ has already been observed for the cesium D_2 line[4] and rubidium D_1 line.[10] These shifts have been explained by long-range atom–surface interactions. Consistent with this interpretation is the fact that we have found no change in the redshift δ_0 for intensities between 0.1mW/cm^2 and 50mW/cm^2.

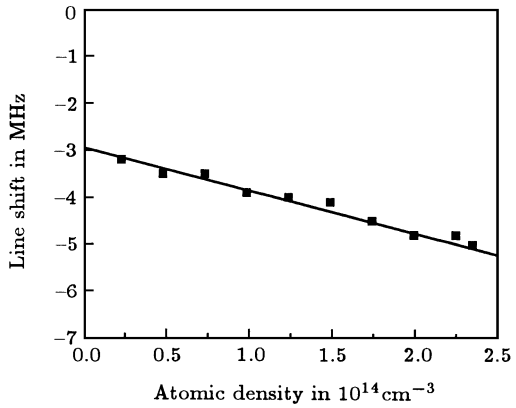


Fig.3. Line shift of the $6S_{1/2}(F{=}4) \rightarrow 6P_{3/2}(F'{=}3)$ component relative to a saturation line as a function of the atomic density ρ.

V. CONCLUSION

The SR of light from the interface between a dielectric and an atomic vapor is a spectroscopy tool that differs in various respects from ordinary absorption spectroscopy. The results given show that WMS, when used with a higher (than first) harmonic detection, is a particularly useful tool for analysing the optical properties of dense atomic vapours. We have measured the self-broadening coefficient for the hfs components of the cesium D_2 line. Our findings are in good agreement with theoretical predictions and con-

firm previous measurements in the atomic line wings. It can be useful for the determination of the local field-induced shift at high atomic densities. The experimental information about collision-induced shift of the hfs in optically dense vapours may be important for a better understanding of new nonlinear effects in an optically dense gas medium.[11]

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