Measurement of the wavelength modulation indices with selective reflection spectroscopy

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The wavelength modulation indices are measured based on harmonic amplitude ratio of $4f_{\text{amp}}/6f_{\text{amp}}$ ($4f_{\text{amp}}$ and $6f_{\text{amp}}$ are the 4- and 6-th harmonic central peak amplitudes correspondingly) with the Doppler-free selective reflection modulation spectroscopy. The experiments for the $6S_{1/2}(F=4) \rightarrow 6P_{3/2}(F'=5)$ transition of cesium D_2 line with 30-MHz linewidth were carried out. The $4f$- and $6f$-harmonic signals were detected with two digital lock-in amplifiers separately. The maximum error for modulation indices measurement was ± 0.1 within the range of m from 3 to 6. The non-linear modulation behaviour of an external cavity diode laser induced by voltage tuning was studied with this method. The method for modulation indices measurement does not require a solid etalon as usual for measuring the wavelength modulation depth and the absorption linewidth correspondingly.

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Wavelength modulation spectroscopy (WMS) with diode lasers has been widely used by many researchers to perform sensitive measurements in gaseous media[1−6]. The absorption spectrum allows a particular atom or molecule to be identified and its concentration and temperature to be measured. WMS with detection at the N-th harmonic of the modulation frequency (Nf-harmonic, i.e. to modulate the wavelength of the diode laser at one frequency, f, and then detect the absorption by use of a lock-in amplifier with it's reference signal having a frequency of N-times of this frequency) would maintain the absorption line derivative like properties. Although the maximum signal amplitude is obtained for $2f$-harmonic (compared with other even harmonics, such as $4f$- and $6f$-harmonic) detection, sometimes residual amplitude modulation of the diode laser can result in a large background variation in the $2f$-harmonic signal and will limit its sensitivity[6,7]. Thus, $4f$- and $6f$- harmonics are preferable with the bigger signal-to-noise (SNR) because the signals are less sensitive to amplitude modulation.

When the modulation center of the laser frequency is tuned to the absorption center of sample, the laser frequency is expressed as $\nu = \nu_0 + \Delta\nu \cos(2\pi f t)$, where ν_0 is the absorption-center frequency, $\Delta\nu$ is the wavelength modulation depth (in hertz), γ is the half-width at half-maximum (HWHM) of the absorption line. The wavelength modulation index m is defined as $m = \Delta\nu/\gamma$. m is one of the most important parameter in modulation spectroscopy with which one can obtain the independent characteristic of the laser or the sample by use of harmonic detection[1,5−7]. Deriving a value for the modulation index m usually requires knowing the frequency range of the laser when the current of the diode laser is modulated at a specific amplitude and the width of the spectral line being probed by use of a solid etalon (free spectral range of GHz or so)[1,5]. Unfortunately, the measurements are difficult to apply to little range (MHz or even less) tuning of laser diodes.

In this work we propose a method for measuring m which will be referred to that we discuss below by use of the harmonic amplitude ratio $4f_{\text{amp}}/6f_{\text{amp}}$. We illustrate the method by making measurements of the hyperfine structure (HFS) of the Cs D_2 line using Doppler-free wavelength modulation reflection spectroscopy[8,9], which provides a novel way to analyse separately for each HFS component collisional processes on a resonance line, while the ordinary saturated absorption technique is plagued with velocity changing collision.

The experimental setup is shown in Fig. 1. A highly coherent external cavity diode laser (New Focus Model 6017)[10] was used as the source of tunable radiation in the experiment. The power of the collimated laser beam is about 5 mW, while the linewidth does not exceed 0.3 MHz. We modulated the laser frequency with 1-kHz sine signal from the internal oscillator sine wave of a digital lock-in amplifier (lock-in 1, SRS, SR830). The wavelength modulation is achieved with sine voltage tuning by means of scanning the piezoelectric stepper motor, which rotates the end mirror inside the laser cavity. 1-Hz triangular wave from a function generator (Hp 33120A) was combined (using Mini Circuit module: ZFRSC-2050) with the sine modulation voltage to scan the piezo too. There is a gain factor set of 25 for input voltage signal's amplification inside the laser controller. The laser beam was sent at a incidence angle θ of 60 mrad onto a Cs vapor cell in which the reflecting interface was formed by an optical glass window and the Cs vapor. The reflected signal was demodulated for $4f$- and $6f$-harmonic signals correspondingly with two digital lock-in amplifiers (lock-in 2 and lock-in 3, SRS, SR830) and recorded with a PC for off-line analysis.

The pure Cs filled glass cell was set in an oven with the temperature of 130 °C. And the Cs density was about 10^{14} cm^{-3} (at the low density collision broadening $\gamma_{\text{col}} < \gamma_r$, the natural width $\gamma_r/2\pi = 5.3$ MHz).

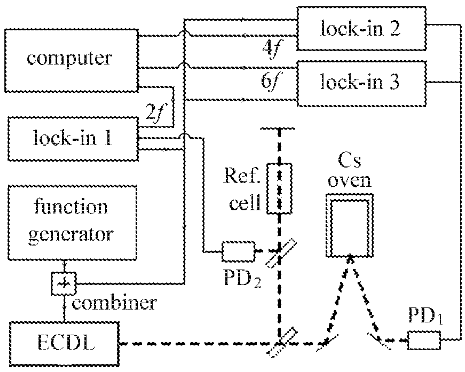

Fig. 1. Experimental setup. The laser beam was from a external cavity diode laser used to pump the $6S_{1/2}(F=4) \rightarrow 6P_{3/2}(F'=5)$ transition. The laser frequency was calibrated using Cs saturation absorption spectrum.

A saturated absorption spectrum with $2f$-harmonic detection of Cs vapor at room temperature was simultaneously recorded with lock-in 1 to provide a frequency reference.

The linewidth of the reflection HFS resonance of the Cs D_2 line determined by residual Doppler broadening[11] was $\Gamma_R = \Delta\nu_D\theta = 30$ MHz (the Doppler linewidth $\Delta\nu_D$ is about 500 MHz). Random samples of other HFS components show essentially the same behavior.

To a Gaussian absorption line, the individual harmonic components can be measured with a lock-in amplifier, and the Nf- harmonic (N = even) amplitudes are given by[7]

$$Nf_{\rm amp}(m) \propto \int_0^{\pi} \exp\left[-\ln 2(m\cos(\omega t))^2\right]\cos(N\omega t){\rm d}(\omega t) \quad (1)$$

with $\omega = 2\pi f$.

Figure 2 shows the theoretical results about the central peak amplitude of $4f$- and $6f$- harmonic signals ($4f_{\rm amp}$ and $6f_{\rm amp}$, dash) as a function of modulation index m. From both of the curves we gave out the curve of harmonic amplitude ratios R ($R = 4f_{\rm amp}/6f_{\rm amp}$) versus m. The curve fitted with a second-order exponential decay to the theoretical R datum reads

$$m = 1.15 + 91.27\exp(-R/0.44) + 12.25\exp(-R/1.20) \quad (2)$$

with R and m in arbitrary unit.

From Eq. (2), we can acquire m with the measured R.

The optimal value for m with the maximum amplitude to harmonic signal has been shown to be 3.6 and 5.2 for the $4f$- and $6f$-harmonic detections, respectively, to a Doppler line shape[1,5,6]. The two points A ($m = 3.6$) and B ($m = 5.2$) in Fig. 2 can be recognized as calibration points by use of checking the maximum amplitude for the $4f$- and $6f$-harmonic signal, respectively, in practice.

Figure 3 shows experimental results of $4f$- and $6f$-harmonic detection traces obtained form the $6S_{1/2}(F=4) \rightarrow 6P_{3/2}(F'=5)$ hyperfine transition of the cesium D_2. Here the amplitude of sine voltage for modulation

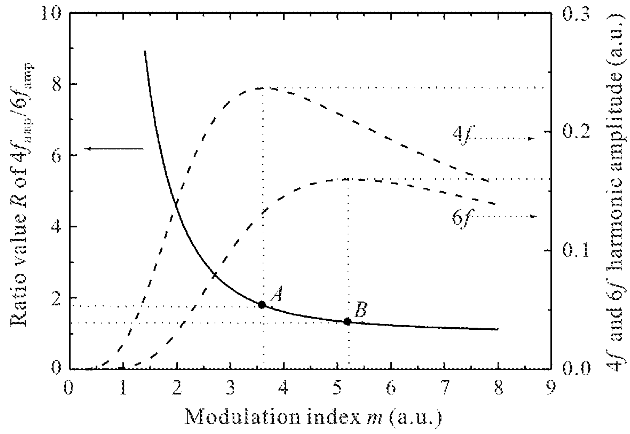

Fig. 2. The theoretical curve of the $4f$- and $6f$- harmonic central peak amplitude ($4f_{\rm amp}$ and $6f_{\rm amp}$, dash) and the harmonic amplitude ratios ($4f_{\rm amp}/6f_{\rm amp}$, solid) as a function of modulation index m for Gaussian reflection (or absorption) line. The points A and B are according to the m = 3.6, 5.2 for the $4f$- and $6f$-maximum peak signals respectively.

was 33 mV rms and all of the lock-in amplifiers with a time constant of 30 ms. The lock-in amplifiers phase settings were chosen to maximize the harmonic signal amplitudes. The ratios of signal (peak-to-peak amplitude) to noise (3σ, where σ is the standard deviation of the noise) were $\sim$ 110 for $4f$-harmonic detection and $\sim$ 70 for $6f$-harmonic detection.

We can infer corresponds the $m = 3.08 \pm 0.05$ with Eq. (2) by use of the measured value $R = 2.21 \pm 0.01$ of $4f_{\rm amp}/6f_{\rm amp}$ in Fig. 3. The error of R is mainly from the ± 1 °C uncertainty of the Cs temperature measurement. With the 30-MHz linewidth, we could also infer that the modulation depth should be $\Delta\nu = \gamma_R \times m = 1/2\Gamma_R \times m = 1/2 \times 30 \times (3.1 \pm 0.1) = 46 \pm 2$ (MHz).

We have measured the modulation index while changing the amplitude of the sine modulation voltage. Figure 4 shows m and modulation depth variation as a function of modulation voltage in the system. For the fixed modulation frequency of 1 kHz, m increases (here also means modulation depth increasing with the fixed resonance HFS linewidth) as the modulation voltage increases. However, we note that the relationship of m and the

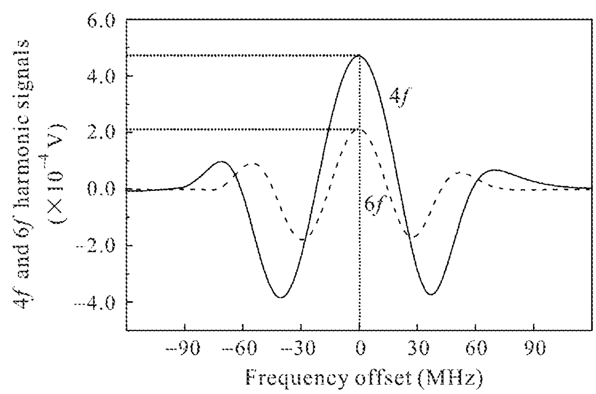

Fig. 3. The $4f$- (solid) and $6f$- (dashed) harmonic traces of the modulation reflection spectroscopy of the Cs D_2 line HFS $6S_{1/2}(F=4) \rightarrow 6P_{3/2}(F'=5)$ (modulated voltage 33 mV rms).

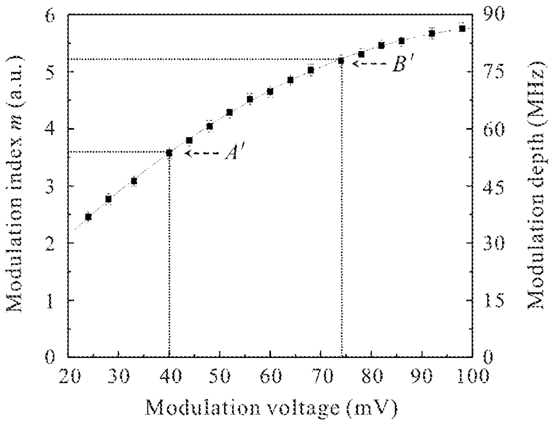

Fig. 4. Modulation characteristics of the external cavity diode laser as a function of the modulation voltage (square, experimental data). The m at A' and B' are measured to be 3.6 ± 0.1, 5.2 ± 0.1 with the maximum peak of $4f$- and $6f$-harmonic detection respectively. The curve is the fitted result of the experiment data.

modulation depth dependence on modulated voltage is not linear which just likes the nonlinear characteristics of m versus modulation frequency with a fixed voltage we have measured before[6]. Both of the nonlinear characteristics were caused by the nonlinear motion of the end mirror piezoelectrically tuned. The experimentally measured modulation indices with maximized amplitudes are $m = 3.6 \pm 0.1$ (A' in Fig. 4) and $m = 5.2 \pm 0.1$ (B' in Fig. 4) for the $4f$- and $6f$-harmonic signals respectively, which equal to the optimum indices earlier recommended for Doppler-broadened absorption line detection in conventional WMS. The maximum error for modulation indices measurement is $\sim \pm 0.1$ within the range of m from 3 to 6. We did not measure the m less than 3 and more than 6 because of the lower SNR of the $4f$- and $6f$- harmonic detection. Moreover, the influence from other HFS of the Cs D_2 line nearby should be considered for the measurement of m more than 6.

We did not use the even lower ($2f$) or higher ($8f$, $10f$, $\cdots$) harmonic signals here for the measurement only considering of the SNR ratios[5]. In our experiment, $2f$-harmonics have slightly higher amplitude signal levels but exhibit significantly higher noise terms; and even higher harmonics have lower background noise terms, but the peak signal level is significantly lower.

In conclusion, we have presented a useful method for the measurements of wavelength modulation indices using the harmonic central peak amplitude ratios $4f_{\mathrm{amp}}/6f_{\mathrm{amp}}$ instead of a solid etalon. To the 30-MHz linewidth of the $6S_{1/2}(F = 4) \rightarrow 6P_{3/2}(F' = 5)$ transition of cesium D_2 line of the Doppler-free selective reflection spectroscopy, the modulation depths were also inferred at the same time. The maximum error for modulation indices measurement is $\sim \pm 0.1$ within the range of m from 3 to 6. The nonlinear modulation behavior of the diode laser dependence on modulated voltage has been shown by use of this method. The method can also be utilized to other symmetric line shapes such as Lorentzian and Voigt profile within the linewidth from kHz to GHz. In addition, possible applications of the method for determining the optimum modulation index which is required in practice[12], or the linewidth of absorption (or deflection) line[13] in the range of kHz to GHz could be of interest.

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