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Experimental investigation of the sub-Doppler transmission spectroscopy in a thin vapour layer at room temperature*

Zhao Yan-Ting(赵延霆)†, Zhao Jian-Ming(赵建明)‡, Huang Tao(黄 涛), Xiao Lian-Tuan(肖连团), and Jia Suo-Tang(贾锁堂)

State Key Laboratory of Quantum Optics and Quantum Optics Devices, College of Physics and Electronics Engineering, Shanxi University, Taiyuan 030006, China

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The sub-Doppler transmission spectrum in a thin vapour layer (about 150 μm) was observed at room temperature using the wavelength modulation technology. The absorption signal and its second-order harmonic were detected with an external-cavity diode laser. A sub-Doppler spectrum corresponding to resonant transitions of the caesium D_2 line ($6S_{1/2} \rightarrow 6P_{3/2}$) was demonstrated. The dependence of the transmission signal on the intensity of the laser was also investigated.

Keywords: thin vapour layer, sub-Doppler technique, harmonic detection
PACC: 3270, 9575F

1. Introduction

The sub-Doppler technique is widely applied in the fields of atomic hyperfine structure, nonlinear process and interface physics, especially in Doppler-broadened systems. Various velocity-selective techniques, such as saturated absorption spectroscopy, provide linewidth narrowed down to natural width limited. As a sensitive detection method with sub-Doppler technology, the selection reflection spectroscopy is usually used to monitor the interaction properties between vapour and medium surface.[1] When the absorption spectroscopy of thin vapour (with a thickness of 10–1000μm) is studied, the atomic flight time from wall to wall is less than the hyperfine energy pumping time, which will create a physical mechanism to select the slow-velocity atomic excitation. So the Doppler effect in the direction of laser pumping can be diminished. The velocity selection is realized in one dimension in a hot atomic environment.

In recent years, the Bloch and Ducloy group have made many experimental studies to testify the regime where the inhomogeneous transit time induces a velocity selection and the signals were affected by optical pumping to a non-absorbing state.[2] Izmailov, Zambon and Nienhuis, Vartanyan and Lin, and Zhao *et al* also gave some theoretical calculation to approve this effect of spatial dispersion due to the atomic motion and electronic quenching with the different thickness of thin cell.[3] Briaudeau *et al* used transmission spectroscopy through an ultra thin Cs cell with a single light beam[2] and Sarkisyan *et al* reported their results obtained with a new thin cell of about 300nm thickness filled with natural Cs or Rb,[4] and investigating the sharp resonance originating from the coherent Doppler narrowing. These experiments are done in the high temperature (above 100°C) for improving the atomic density and the signal to noise ratio (SNR).

In this paper, the transmission spectrum in a thin vapour layer (about 150μm) is measured at room temperature, and wavelength modulation technology is used to enhance the sensitivity. The dependence of the transmission signal on the intensity of the laser is investigated simultaneously.

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†E-mail: zhaoyt@sxu.edu.cn
‡E-mail: zhaojm@sxu.edu.cn

2. Experimental apparatus

The experimental setup is presented in Fig.1. We use a Littman–Metcalf external-cavity laser (NewFocus 6017 model) as a source of tunable radiation, which is temperature stabilized and can be tuned to the D_2 line of caesium atoms. The free-running linewidth is less than 500kHz (<50ms). The linearly polarized laser beam is expanded and through a thin vapour cell under normal incidence. Cs vapour pressure is about 10^{-4}Pa and its density is about 3×10^{10} atoms/cm^3 at room temperature. We use the optical attenuator to control the incidence power and avoid the saturation of avalanche photodiode (Hamamatsu Si APD, S3884). A saturated absorption spectrum of caesium vapour is simultaneously recorded to provide a frequency reference. The function generator (HP33120A) provides a ramp voltage and a sine voltage signal provided by the lock-in amplifier (Model SR 830) is added to the high-speed current modulation input of laser head. The harmonic detection technique is used to increase the sensitivity. The signal is fed into a lock-in amplifier, digitized by an A/D convertor and stored in a personal computer for further processing.

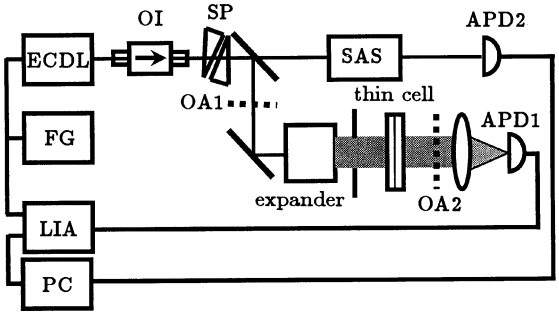


Fig.1. Experimental setup: ECDL—external cavity diode laser; FG—function generator; LIA—lock-in amplifier; OA—optical attenuator; OI—optical isolator; SAS—saturated absorption spectrum; SP—shaping prism.

3. Experimental result and analysis

The transmission spectrum in a thin vapour layer shows the sub-Doppler structure, which provides a novel way to analyse each hyperfine component of a resonance line. When the atomic flight time from wall to wall is less than the hyperfine energy pumping time, only the atoms with small velocity components perpendicular to the walls will have sufficient time for optical transition. The interaction between an atom and coherent radiation is influenced mainly by collisions with the closely spaced end wall. The effect of the laser power on the transmission spectrum of the single beam is investigated and the result is shown in Fig.2.

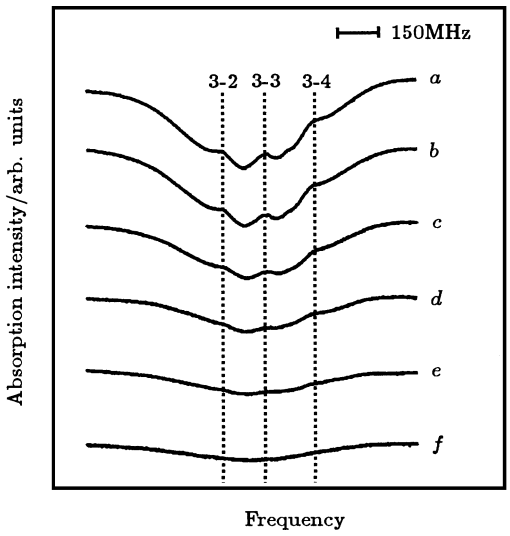


Fig.2. Transmission spectrum of single beam in a thin vapour layer (150μm), resonant transitions caesium D_2 line ($6S_{1/2}$ (F=3) $\rightarrow$ $6P_{3/2}$ (F'=2, 3, 4)); the dependence of the spectral profile on the laser power is demonstrated in the different curves. The laser intensity is (a) 3.56mW, (b) 2.16mW, (c) 1.12mW, (d) 0.43mW, (e) 0.22mW, (f) 0.15mW. The laser beam diameter is 1cm.

When the incident power is increased, the hyperfine structures are demonstrated clearly. The thin cell has a thickness in the laser beam direction, so there are some atoms which can off-resonantly absorb the laser radiation when the laser frequency is scanning over the transition line. When the laser intensity is increased, the number of these atoms also is increasing, and more atoms are optically pumped to the other hyperfine level. When the incidence laser beam power is 3.56mW (the beam diameter is 1cm), the typical transmission spectrum of the $6S_{1/2}$ (F=3) $\rightarrow$ $6P_{3/2}$ (F'=2, 3, 4) is obtained (shown by curve a). The hyperfine transition line disappears when the power is less than 0.15mW (shown by curve f). The sub-Doppler spectrum has no crossover lines; this is coincident with our prediction. The theoretical description has been reported by Otake *et al.*[5] In their model, the optical pumping process and surface-induced population relaxation between the hyperfine sublevels are taken into account. In our experimental condition, the contribution for the power absorption coefficient of each hyperfine component $F = 3 \rightarrow F'$=2, 3, 4 is

given by

$$\alpha(\omega) \propto \sum_{i=2,3,4} \int_{-\infty}^{+\infty} \frac{R_i^2 \left(N_3 - N_3^*\right)}{\left(\omega - \omega_i + kv_z\right)^2 + \dfrac{1}{(2\tau)^2}} \mathrm{d}v_z,$$

where R_i is the Rabi frequency of each transition; N_3 is the steady-state velocity distribution in the hyperfine ground state F=3 and N_3^* is the velocity distribution in thermal equilibrium; ω is the frequency of the laser and ω_i is the resonant frequency of each hyperfine component of the D_2 line; τ is the spontaneous emission lifetime of the $6P_{3/2}$ state.

The atomic relaxation lifetime is determined by the flight time between two closely spaced end walls of the thin cell. The group of atoms with a smaller velocity component perpendicular to the end walls is more efficiently accumulated in the ground state (F=3). The spectrum near resonance is notably affected by the condition that atoms depart from the surface mainly in their ground state, and will have traversed some distance before the induced dipole moments of the vapour atoms reach their steady state.[3] In Briaudeau's experiments, the intensity of the single beam is sufficiently low to ensure the linear absorption.[2] Under that condition, only atoms with small velocity components can complete hyperfine pumping during the wall-to-wall flight. The interaction of the laser and atom is velocity-dependent. The thickness of the layer is less than the mean free path of atoms. The velocity-dependent mean free time is essential for producing the non-Maxwellian velocity distribution (the mean free path for the velocity-changing collision is 1.4m),[5] so the special velocity distribution is compacted in the laser transmission direction and appears insensitive to the Doppler shift. It has been theoretically shown that a major contribution to the signal could come from atoms that are slower than those selected by the usual Doppler selection for a given transition, which can lead to the observation of super-slow atoms.[6]

Because of the special line profile, Lorentz structure under Doppler background will lead to much narrower profile by using the wavelength modulation and harmonic detection.[3] We can contrast the direction absorption signal and its second-order harmonic signal (see Fig.3.).

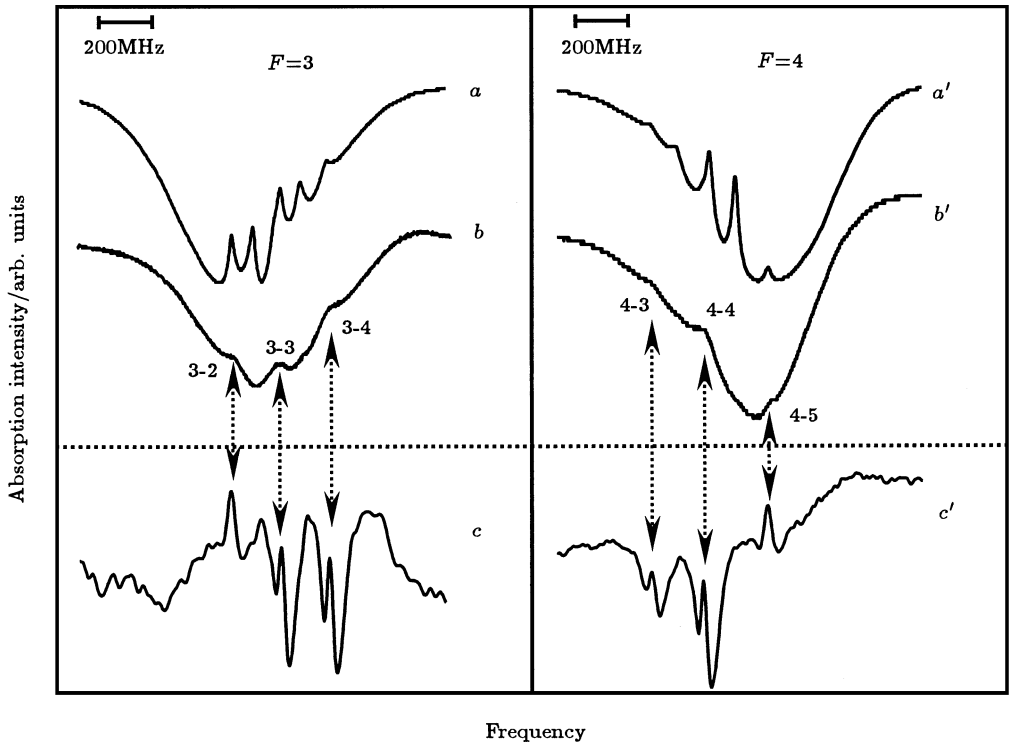


Fig.3. The comparison between the transmission spectrum and its second-order harmonic signal. This higher sensitive detection technology is used to investigate the sub-Doppler hyperfine structure. The laser intensity is 1.5mW, curves a and a' are the saturated absorption spectra, curves b and b' are the transmission spectra and curves c and c' are the second-order harmonic signal. The left graph is the transition spectrum of F=3 $\rightarrow$ F'=2, 3, 4 and the right graph is the transition spectrum of F=4 $\rightarrow$ F'=3, 4, 5.

Curve a is the saturated absorption spectrum (F=3), curve b is the direction absorption spectrum without modulation and curve c is the second-order harmonic of curve b. For detecting curve c, the modulation index is 0.8, modulated frequency is 50kHz and the integral time constant is 3 μs. Higher sensitivity and SNR can be obtained if the parameters are suitable.[7] At the same time, the narrower linewidth is demonstrated when the modulation amplitude is small (contrast curves b and c). If we want to improve the SNR, the modulation amplitude should be larger and the linewidth becomes broadened. The transition spectrum from $6S_{1/2}$ (F=4) $\rightarrow$ $6P_{3/2}$ (F'=3, 4, 5) has the same result, as shown in the right panel of Fig.3.

4. Conclusion

In conclusion, direct absorption experiments with caesium D_2 line for the $6S_{1/2} \rightarrow 6P_{3/2}$ transition in a thin vapour cell are carried out at room temperature. The sub-Doppler spectral structure caused by the velocity-dependent transient optical pumping is demonstrated. The Doppler effect in the direction of laser pumping is diminished and the velocity selection is realized at one dimension. The second-order harmonic of the signal is detected to improve the observation and the effect of the laser power on the signal is investigated simultaneously. This is a convenient and powerful tool for many applications, such as the study of gas–surface interaction,[1] application in long-term frequency stabilization of laser systems,[8] and image processing by far fewer photons than other nonlinear materials,[9] even for potential application in electromagnetic induced transparency medium.[10]

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