

Modulation transfer spectroscopy in a lithium atomic vapor cell

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Abstract: We have investigated modulation transfer spectroscopy of D₂ transitions of ⁷Li atoms in a vapor cell. The role of the intensity of the probe beam in the spectrum is important, we have seen unique characteristics of the signal in the crossover peak. In order to find the best signal for laser locking, the slope and frequency offset of the zero-crossing signal are determined. The dependence of the modulation transfer spectra on polarizations of pump and probe beam is demonstrated. The residual amplitude modulation in the system is also considered, and the distortion of the spectra due to the modulation is analyzed. It was found that the crossover peak is more suitable for frequency stabilization due to its better residual amplitude modulation compensation.

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1. Introduction

Laser frequency stabilization is crucial for precision measurement with atoms. Modulation transfer spectroscopy is a good candidate for laser frequency stabilization, where the phase of pump beam is modulated by an electro-optic modulator (EOM) or an acousto-optic modulator (AOM), and then is transferred to the probe beam. Due to the nonlinear interaction between the light and atoms, modulation transfer spectroscopy is Doppler free, its spectral zero point is not affected by magnetic field and the stability of the baseline is not affected by laser intensity, laser polarization, and temperature. This spectroscopy was first proposed in 1982 [1]. Since then, it has been investigated experimentally and theoretically. In 1995, Eickhoff and Hall stabilized a frequency-doubled Nd: YAG laser by modulation transfer spectrum (MTS) of molecular iodine [2]; Jaatinen proposed a scheme to optimize the MTS [3]. In 2001, Bertinetto et al. stabilized the frequency of a diode laser using the MTS of cesium [4]. In 2003, Zhang et al. displayed the MTS of rubidium D_2 transition line [5]. In 2011, Li et al. calculated the MTS for non-cycling transitions in a two-level system [6]; Noh et al. further studied the MTS of rubidium theoretically [7]. Recently, Negnevitsky and Turner obtained MTS of rubidium and stabilized the frequency of a laser using the spectra [8]. The above MTS signals are explained by four wave mixing (FWM) [9]. Usually the cycling transition resonances are paid more attention than the crossover ones. In this paper, we investigate the modulation transfer spectrum of lithium,

and focus on the crossover peaks as well as the cycling transitions.

Lithium is attracting a wide attention in precision measurement physics due to its small mass and large photon-recoil momentum. In 2011, Hohensee and Müller [10] proposed that ^6Li and ^7Li are particularly suitable for the weak equivalence principle test. Recently, sub-Doppler cooling of lithium has made breakthrough progress [11–14]. However, the spacing between the hyperfine ground states of lithium is less than 1 GHz, where the crossover peak of the ground state plays a key role in the spectrum. Meanwhile the spacing among the hyperfine excited states is as small as the natural linewidth, the conventional spectroscopy, such as saturated absorption spectroscopy (SAS), frequency modulation spectroscopy (FMS), cannot supply a stable locking point for laser frequency stabilization. This is due to Doppler broadening, and the dependence of the stability lock point on baseline, magnetic field, intensity, polarization and temperature. Although, the MTS can't resolve the complicated hyperfine spectra of lithium D_2 line, it gives a much steeper slope at the zero-crossing of the dispersion signal due to its nonlinear feature. This zero-crossing is further immune to Doppler broadening, baseline ambiguity, ambient magnetic field, intensity, polarization and temperature. Thus MTS results in a much tighter locking for lasers used in laser cooling and trapping of lithium atoms. But the MTS in lithium has not been studied before. Here we present experimental investigation of MTS of ^7Li for the first time. Because of the unique structure of the energy levels, the MTS of ^7Li displays special characteristics. In this work, we carefully examine the dependence of spectra not only on the intensity of the probe beam but also on polarizations of the probe and pump beams [15, 16]. Different combinations, such as the parallel linear polarizations $\text{lin}\parallel\text{lin}$, perpendicular linear polarizations $\text{lin}\perp\text{lin}$, the same circular polarizations $\sigma^+-\sigma^+$ ($\sigma^- - \sigma^-$) and the opposite circular polarizations $\sigma^+-\sigma^-$ ($\sigma^- - \sigma^+$), are demonstrated. The residual amplitude modulation (RAM) in the system is studied thoroughly and the distortion of modulation transfer spectra due to RAM is analyzed using a fitting routine.

The results indicate that MTS is suitable for laser frequency stabilization in atoms like lithium where the small hyperfine splitting make the frequency stabilization due to the conventional techniques more uncertain. .

2. Theory

The phase-modulated light can be expressed by

$$E = E_0 \sin(\omega_0 t + \delta \sin \omega_m t). \quad (1)$$

Eq.(1) can be expanded by Bessel function as

$$E = E_0 \left[\sum_{n=0}^{\infty} J_n(\delta) \sin(\omega_0 + n\omega_m)t + \sum_{n=0}^{\infty} (-1)^n J_n(\delta) \sin(\omega_0 - n\omega_m)t \right], \quad (2)$$

where ω_0 is the carrier wave frequency, ω_m is the modulation frequency, δ is the modulation index, and $J_n(\delta)$ is the n -th order Bessel function. According to FWM theory [1], the beat signal of the probe beam and its sidebands on the detector is in the form

$$S(\omega_m) = \frac{C}{(\Gamma^2 + \omega_m^2)^{1/2}} \sum_{n=-\infty}^{\infty} J_n(\delta) J_{n-1}(\delta) [(L_{(n+1)/2} + L_{(n-2)/2}) \cos(\omega_m t + \Phi) + (D_{(n+1)/2} - D_{(n-2)/2}) \sin(\omega_m t + \Phi)], \quad (3)$$

where $L_n = \frac{\Gamma^2}{\Gamma^2 + (\Delta - n\omega_m)^2}$, $D_n = \frac{\Gamma^2(\Delta - n\omega_m)}{\Gamma^2 + (\Delta - n\omega_m)^2}$, Γ is the natural linewidth, Δ is the frequency detuning, Φ is the phase of the modulation field applied to the pump beam and C is a constant.

Eq. (3) indicates that the MTS is composed of a sine term which represents the quadrature component, and a cosine term which describes the in-phase component. The phase-sensitive detection is used to recover the two terms by setting the phase to 0 and $\pi/2$, respectively.

3. Experimental setup

The experimental setup for modulation transfer spectroscopy of Li atoms is shown in Fig. 1. The 671 nm seed beam is provided by a diode laser (New Focus, DL). The seed beam is injected into a homemade Tapered amplifier (TA) via a single mode PM fiber. Out of the 470 mW beam, about 3 mW of power is used for the MTS experiment through PBS1. The probe and pump beams are made to coincide in the lithium vapor cell [17]. We use three mutually orthogonal coil pairs to compensate the stray magnetic flux densities to about 10 mG for the area of the interaction between atoms and laser. The vapour cell is heated to 460°C to get sufficient vapours of Lithium for the experiment. A telescope system [18], consisting of a lens with focus of -50 mm and a lens with focus of 100 mm, is used to expand the laser beam size from 1 mm to 2 mm in diameter. In addition, a $\lambda/2$ and a $\lambda/4$ wave plate are added on both sides of the vapor cell to adjust the beams polarization. The phase of pump beam is modulated by an EOM (EO-AM-NR-C1, Thorlabs), which is driven by a 6.42 MHz rf signal from a direct digital synthesis (DDS). The modulation index is 0.5. A photodiode (PD) with two channels is used to record the spectrum, the AC channel is for MTS, and the DC channel is for saturated absorption spectrum (SAS). The AC signal is amplified and mixed with the demodulation signal by a frequency mixer (Mini-Circuits, ZX05-1-S+). The output from the frequency mixer is filtered by a 30 kHz low pass filter and amplified by a homemade amplifier. An oscilloscope is used to monitor the final MTS signal.

The lineshape of the MTS signal depends on two processes, the incoherent process (modulated hole burning) [1] and the coherent process (FWM), both of which are determined by the modulation frequency ω_m . As mentioned in Ref. [8], for low frequency modulation, the incoherent process is predominate, the signal resembles a Doppler-free frequency modulation spectrum (FMS); for high frequency modulation, the coherent process works in the formation of MTS. To reduce the effect of incoherent process, high frequency modulation was chosen for our experiment.

The lower limit of ω_m is decided by the mean transit time for atoms in the vapor which will depend on the velocity and hence temperature of atoms and the size of the laser beam. Under our experimental conditions with temperature of 460 and a beam diameter of 2 mm, this lower limit is around $f = 750$ kHz. In order to get an optimal signal as [19], the best modulation frequency value is around 0.7Γ where Γ is the linewidth of the transition involved. For ^7Li , the natural linewidth is 5.9 MHz, corresponding to the best modulation frequency of 4.13 MHz. However as in the experiment, because of the influence of saturation broadening of the spectrum, linewidth of the spectral lines, is often larger than the natural line width, so the corresponding optimal modulation frequency also increases. Further higher modulation frequency is beneficial for suppressing the amplitude noise of the laser beams [20], allowing better signal to noise ratio. However much higher modulation frequency can make the spectral profile worse [18]. For these reasons, the modulation frequency was chosen as 6.42 MHz which is near to the value of the natural linewidth.

4. Experimental results and discussion

4.1. Intensity of the probe beam

In order to increase the signal to noise ratio of the spectral lines, the intensity of pump beam I_{pump} is usually regulated a lot more than the probe beam in the MTS experiments. At the

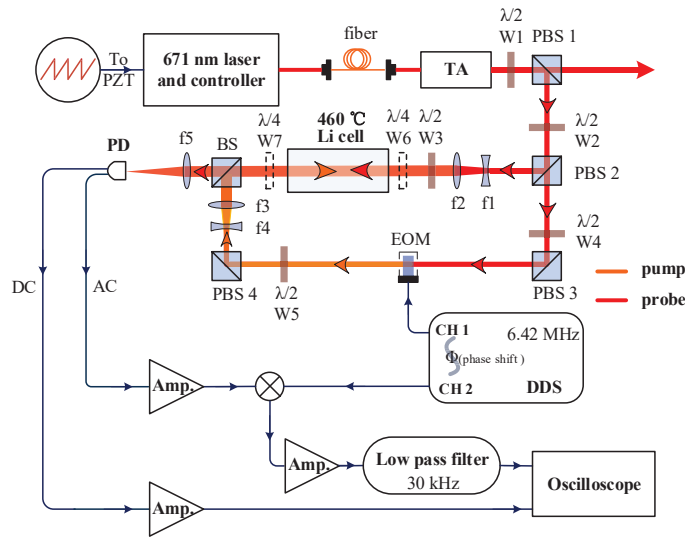


Fig. 1. Experimental setup of MTS measurement. Frequency modulation of 6.42 MHz is transferred from the pump to the probe beam within the lithium vapor cell. The beat of the probe frequency components at a two-path photodetector PD is demodulated to produce an error signal. f1, f2, f3, f4, and f5 are lenses with focuses of -50, 100, 100, -50, and 100 mm, respectively. EOM, electro-optic modulator; W, wave plate; PBS, polarization beam splitter; DDS, direct digital synthesizer; TA, tapered amplifier.

same time, the probe light power I_{probe} should be weaker than the saturation intensity. In our experiment, we study the role of the intensity of the probe beam in the spectrum when the intensity of pump beams is high. We choose the $\sigma^+-\sigma^+$ ($\sigma^--\sigma^-$) polarization combination of the probe beam and pump beams, for the improvement of the signal of MTS corresponding to the closed transition. Different kinds of polarization configurations will be discussed in the following pages. By adjusting the phase, we could change all the three peaks as dispersion line shape which will be useful for amplitude comparison.

Figure 2 shows the influence of different intensities of the probe beam on the spectra when the intensity of pump beams stay the same ($I_{pump}=8I_{sat}$, $I_{sat}=5.1 \text{ mW/cm}^2$). By modulating the voltage applied to the piezo actuator and the current applied to laser diode, the laser is scanned over a frequency range of 800 MHz that covers two resonance transition peaks [1, F'] and [2, F'] and a crossover [1-2, F']. Figure 2(a) displays the signal of MTS when the intensity of the probe beam is $0.5I_{sat}$, I_{sat} and $2I_{sat}$ respectively. From this figure we see, when $I_{probe}=0.5I_{sat}$ at the spectrum of MTS in lithium shows the same character as in other alkali atoms (such as rubidium and potassium) that the main peak of the spectrum corresponds to the cycling transition, and other peaks are significantly inhibited. However, with the increase of I_{probe} , the amplitude of all the three peaks increases. In order to get more systematic study for the influence of the intensity of the probe beam on the spectrum, we changed I_{probe} from $0.25I_{sat}$ to $11I_{sat}$. Figure 2(b) and Fig. 2(c) show changes of the peak-peak amplitude of the three peaks and their proportions in the whole spectrum. From the figures we see that, the vertical dashed line, representing the position of the saturate intensity, divides the whole process into two parts. When $I_{probe} < I_{sat}$, in the beginning, the peak-peak amplitude and proportion of two resonance peaks is greater than

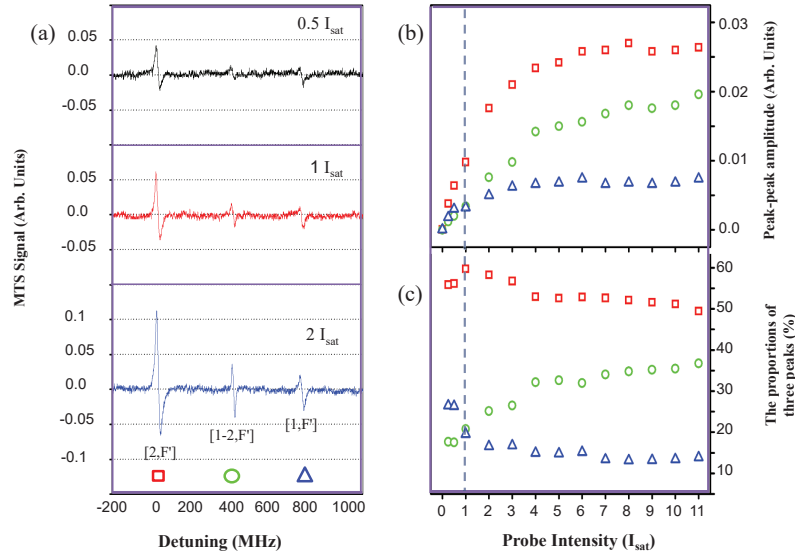


Fig. 2. The influence of different intensities of the probe beam on the spectra when the intensity of pump beams stay the same ($I_{pump} = 8I_{sat}$, $I_{sat} = 5.1 \text{ mW/cm}^2$). (a) The signals of MTS when the intensity of the probe beam I_{probe} is $0.5 I_{sat}$, $1 I_{sat}$ and $2 I_{sat}$ respectively; (b) The peak-peak amplitude of the three peaks in the spectrum; (c) The proportions of three peaks in the whole spectrum. The position of the saturation intensity is shown by a vertical dashed line.

the crossover one. But with the increase of I_{probe} , the amplitude of $[2, F']$ and $[1-2, F']$ continue to increase linearly, but the increase rate of $[1, F']$ begins to decline which can be seen from Fig. 2(b). When $I_{probe} = I_{sat}$, the peak-peak amplitude of $[1, F']$ and $[1-2, F']$ are basically the same, while the proportion of $[2, F']$ reached a peak. When $I_{probe} > I_{sat}$, the peak-peak amplitudes of $[1, F']$ and $[2, F']$ are increased gradually and leveled off and their proportions decrease and leveled off too. In contrast, both the peak-peak amplitude and the proportion of $[1-2, F']$ have been on the rise, and gradually become stable. This can be seen from the chart, the position where the amplitude begins to stabilize is in $I_{probe} = 4I_{sat}$. In fact, in the following experiment, the intensity of the probe beam is the value of choice.

4.2. The comparison of MTS and FMS

The modulation transfer spectra of lithium with $\text{lin} \perp \text{lin}$ polarization configuration are shown in Fig. 3. According to Fig. 3, the profile changes with the phase shift. The in-phase and quadrature components correspond to $\Phi = 0^\circ$ and $\Phi = 90^\circ$, respectively. The detuning of resonance peak $[2, F']$ is defined as zero, then the detuning of $[1, F']$ is about 800 MHz and the crossover peak is at 400 MHz. The spectra of lithium show that there is a crossover dispersion peak, which is different from the work reported before (rubidium [5, 18], potassium [21]), this is related to the unique energy level structure of lithium atoms [22]. The crossover dispersion peak is intense and steep, it is suitable for stabilizing laser frequency. It is worth noting that, the shape of resonance peak in the spectra is asymmetric, while the shape of crossover peak is symmetric

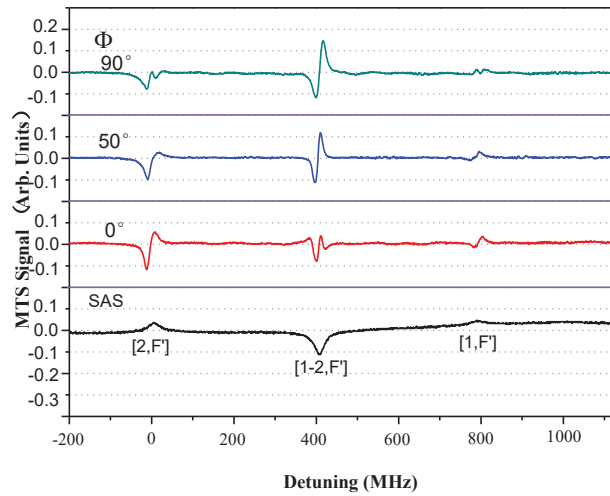


Fig. 3. The MTS of lithium with lin⊥lin polarization configuration and the SAS for reference.

ust when we choose the phase at about 50° , this phenomenon is influenced by the residual amplitude modulation (RAM). To well understand the MTS, we compare it with FMS, which has been widely used in laser locking. For MTS, the frequency modulation is applied to the pump beam; while for FMS, the frequency modulation is applied to probe beam, which is similar as SAS, this avoids additional noise caused by modulating laser current directly. What we should do is just move the EOM crystal from the path of pump beam to the probe beam.

Figure 4 shows the FMS of D_2 transitions in ^7Li under same parameters as the MTS. The spectrum shows good dispersion character which resembles the differential signal of SAS. There are several significant differences between FMS and MTS. Firstly, the Doppler background still exists in the FMS due to incoherent process. Secondly, the profile of MTS changes with phase shift Φ , while the profile of FMS does not change with Φ , the sign of the slope for resonance peaks is opposite with that of crossover. Thirdly, from the inset at the top right, the slope of the dispersion peak in the MTS is steeper than that of FMS. Finally, the profile of MTS changes with polarization, the profile of FMS does not. The detailed comparison between the slope and frequency offset of the zero-crossing signal is shown in Table 1. Table 1 shows that the FMS has less steeper slope and larger frequency offset due to Doppler background, the resonant peak $[2, F']$ (0°) and the crossover peak $[1-2, F']$ (90°) in MTS are worse than $[1-2, F']$ (50°) due to residual amplitude modulation.

Table 1. The slope and frequency offset of the signal zero-crossing for MTS and FM. $[2, F']$ is the resonance peak, $[1-2, F']$ is the crossover peak, and the slope is relative value in any units.

	MTS			FMS	
	$[2, F']$ (0°)	$[1-2, F']$ (90°)	$[1-2, F']$ (50°)	$[2, F']$	$[1-2, F']$
Slope	0.625(8)	0.465(9)	0.702(17)	0.392(8)	0.520(5)
Offset (MHz)	0.83(28)	0.69(12)	0.36(14)	5.04(42)	1.29(19)

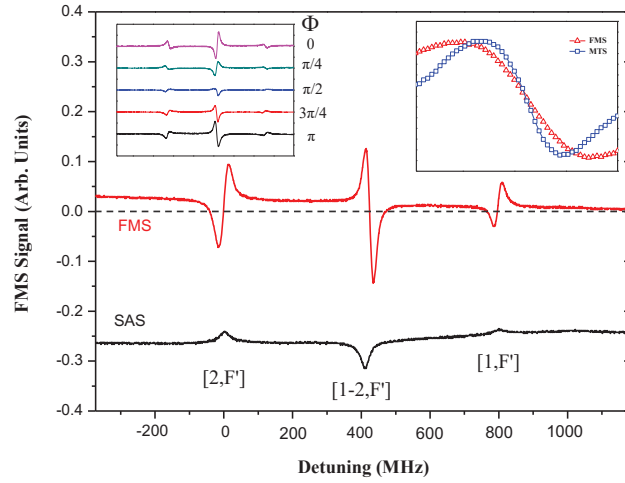


Fig. 4. FMS of D_2 transitions in ^7Li . The inset at the top left shows that the FMS varies with the phase shift $\Phi(0, \pi/4, \pi/2, 3\pi/4$ and π from top to bottom). The inset at the top right shows the comparison of the slopes of the crossover dispersion peaks between MTS and FMS.

4.3. Different polarization combinations

To investigate the dependence of MTS on laser polarizations, we consider the different polarization combinations, such as $\text{lin}\parallel\text{lin}$, $\text{lin}\perp\text{lin}$, $\sigma^+-\sigma^+$ ($\sigma^--\sigma^-$), and $\sigma^+-\sigma^-$ ($\sigma^--\sigma^+$). We remove the $\lambda/4$ wave plates on both sides of the vapor cell to make sure that the probe beam and pump beam are linear polarized, and choose suitable angles of $\lambda/2$ wave plates for the configuration of $\text{lin}\parallel\text{lin}$, $\text{lin}\perp\text{lin}$. For the circular polarization configuration, the $\lambda/4$ wave plates are inserted back to the light path, the $\lambda/2$ and $\lambda/4$ wave plates are rotated to satisfy the polarization configurations of $\sigma^+-\sigma^+$ ($\sigma^--\sigma^-$) and $\sigma^+-\sigma^-$ ($\sigma^--\sigma^+$).

The modulation transfer spectra under different polarization configurations are shown in Fig. 5, where Fig. 5(a) and Fig. 5(b) display the in-phase and quadrature components, respectively. According to Fig. 5(a), the amplitude of resonance peaks $[2, F']$ and $[1, F']$ in the $\text{lin}\perp\text{lin}$ configuration are weaker than that of $\text{lin}\parallel\text{lin}$ configuration; the amplitude of resonance peaks $[2, F']$ and $[1, F']$ in $\sigma^+-\sigma^-$ ($\sigma^--\sigma^+$) configuration is lower than that of $\sigma^+-\sigma^+$ ($\sigma^--\sigma^-$) configuration; the amplitude of the two resonance peaks $[2, F']$ and $[1, F']$ in the $\sigma^+-\sigma^-$ ($\sigma^--\sigma^+$) configuration is lower than in other configurations. The amplitude of the resonance peak $[2, F']$ in the $\sigma^+-\sigma^+$ ($\sigma^--\sigma^-$) configuration is higher than that in other configurations, while the amplitude of the resonance peak $[1, F']$ is not sensitive to polarization configurations, this is different from that mentioned in [16] because the spacings among the hyperfine states of the excited state of ^7Li are too small to be distinguished. Figure 5(b) shows that the amplitude of crossover peak is not sensitive to polarization configuration.

Even the frequency of laser is tuned to the closed transition in lithium, atoms may still decay out of the close loop, because of the unresolved excited state. Relative to the “completely closed transition”, in this kind of closed transition the spontaneous emission branch ratio is less than 1, thus the MTS signal is weakened [6]. For this reason, the closed transitions in lithium are

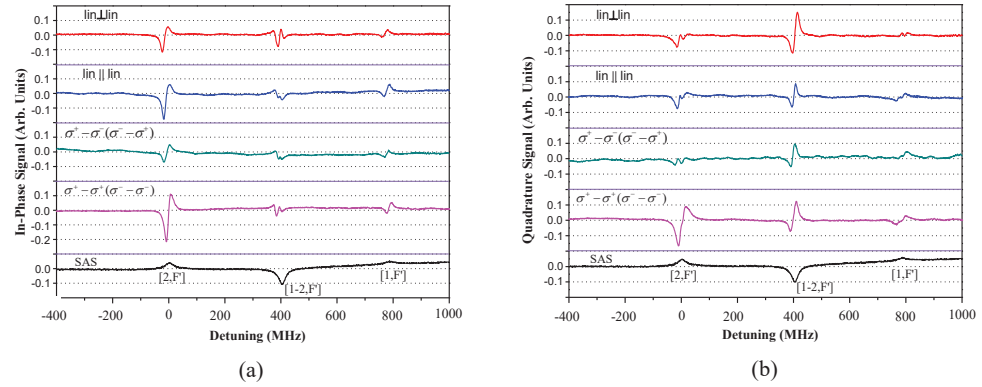


Fig. 5. The in-phase (a) and quadrature (b) components of modulation transfer spectra under different polarization configurations.

weaker than those of other elements, such as rubidium. These transitions are “weakly closed transition”.

The resonance signal in the MTS can be interpreted by the FWM mechanism, the amplitude is determined by atom population and transition probabilities (branch ratios). Figure 5(a) shows the spontaneous emission branch ratios in the transition of $F_g=2 \rightarrow F_e=3$, and Fig. 5(b) and Fig. 5(c) reveal the FWM processes in different polarization configurations. P, C and S denote the probe, carrier and sideband beams, respectively. The new light beam generated by FWM process is represented by the curve with an arrow. The numbers on the lines of magnetic sublevels are relative populations.

As shown in Fig.6 (b), radiated by the pump beams with circular polarization of σ^- (σ^+), all atoms are populated in $F_g = 2, m_F = -2$ ($F_g = 2, m_F = 2$). When the probe beam polarization is the same as pump beams (Fig. 6(b) [i]), namely σ^- (σ^+), then all of the atoms are confined in the transition of $F_g=2, m_F = -2 \rightarrow F_e=3, m_F = -3$ ($F_g = 2, m_F = 2 \rightarrow F_e = 3, m_F = 3$) which is a completely closed transition, and the spontaneous emission branch ratios is 1, the amplitude of the corresponding MTS peak is the largest. However, when the probe beam polarization is opposite (Fig. 6(b) [ii]), namely σ^+ (σ^-), atoms transit from $F_g = 2, m_F = -2$ ($F_g = 2, m_F = 2$) to $F_e = 3, m_F = -1$ ($F_e = 3, m_F = 1$), in which the transition probability is too low (the relative transition probability is only 1/15), so that the amplitude of the corresponding MTS signal is very weak.

Figure 6(c) shows the situations that the probe and pump beams are all linearly polarized. The FWM process contains the probe and pump beams, and the amplitude of the MTS signal is related to the spontaneous emission branch ratio γ_1 , atomic population η and the transition probability ρ , so that the MTS signal

$$S(\omega_m) \propto \eta \cdot \rho_C \cdot \rho_S \cdot \gamma_1 \cdot \rho_P \quad (4)$$

where ρ_C , ρ_S and ρ_P are the relative transition probabilities under the action of carrier, sideband and probe beams, respectively. For the lin||lin configuration (Fig. 6(c) [i]), five “weakly closed transitions” are contained in the system, so that the MTS signal is formed by five FWM processes. As a result of the “weakly closed transitions”, the whole amplitude of MTS signal is weaker than that in “completely closed transitions”. The whole MTS signal can be expressed by

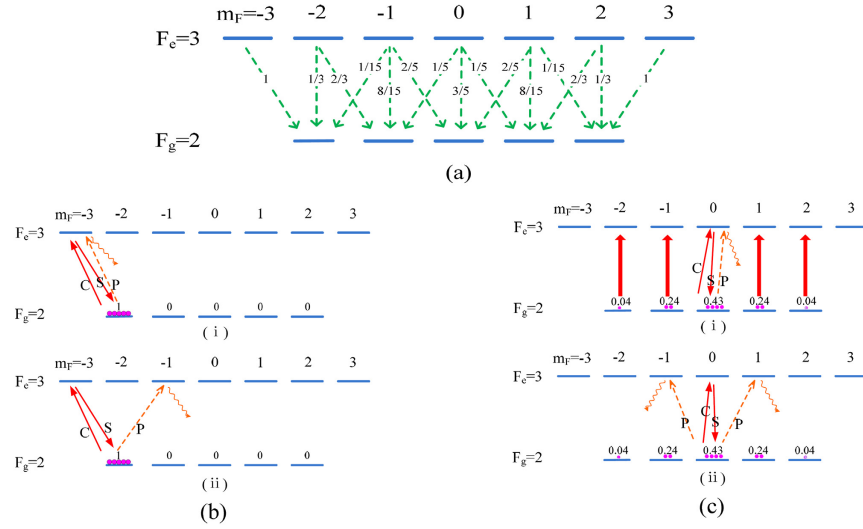


Fig. 6. (a) The spontaneous emission branch ratios in the transition of $F_g=2 \rightarrow F_e=3$. (b) The diagram of FWM processes in the transition of $F_g=2 \rightarrow F_e=3$ when the pump beams are circular polarization of σ^- and the probe beam is circular polarization of σ^- (i) or σ^+ (ii). (c) The diagram of FWM processes in the transition of $F_g=2 \rightarrow F_e=3$ when the polarization configuration is $\text{lin} \parallel \text{lin}$ (i) or $\text{lin} \perp \text{lin}$ (ii).

$$S(\omega_m)_{\parallel} = S(\omega_m)_{-2}^{-2} + S(\omega_m)_{-1}^{-1} + S(\omega_m)_0^0 + S(\omega_m)_1^1 + S(\omega_m)_2^2, \quad (5)$$

where $S(\omega_m)_A^B$ means an FWM, A denotes a magnetic sublevel of the ground state, B is a sublevel of the excited state.

For the $\text{lin} \perp \text{lin}$ configuration (Fig. 6(c) [ii]), the linear polarization is decomposed as σ^+ and σ^- , the MTS signal is expressed by

$$S(\omega_m)_{\perp} = \frac{1}{2} [S(\omega_m)_{-2}^{-3} + S(\omega_m)_{-2}^{-1} + S(\omega_m)_{-1}^{-2} + S(\omega_m)_{-1}^0 + S(\omega_m)_0^{-1} + S(\omega_m)_0^1 + S(\omega_m)_1^0 + S(\omega_m)_1^2 + S(\omega_m)_2^1 + S(\omega_m)_2^3]. \quad (6)$$

Different from Eq. (5), a factor of 1/2 is added to Eq. (6) because of the attenuation of light intensity. Inserting Eq. (4) into Eq. (5) and Eq. (6), we get that $S(\omega_m)_{\parallel} > S(\omega_m)_{\perp}$, which is consistent with experimental result.

The crossover signal in MTS of lithium can be interpreted by a new scheme [16]. Figure 7 shows the energy level diagrams for the crossover signal under the parallel (a, c) and perpendicular (b, d) linear polarization configurations. The green dash line represents the spontaneous emission process.

The component of $F_g=1, 2 \rightarrow F_e=1$ (a, b) results from both probe components $F_g=1 \rightarrow F_e=1$ (i) and $F_g=2 \rightarrow F_e=1$ (ii), where the pump beam is tuned to $F_g=2 \rightarrow F_e=1$ (i) and $F_g=1 \rightarrow F_e=1$ (ii), respectively. When the pump beam is tuned to $F_g=2 \rightarrow F_e=1$, atoms on the levels of $|F_g=2, m_F=0, \pm 1\rangle$ are excited to $|F_e=1, m_F=0, \pm 1\rangle$, and then decay to $|F_g=1, m_F=0, \pm 1\rangle$. For the $\text{lin} \parallel \text{lin}$ configuration, the transition $|F_g=1, m_F=0\rangle \rightarrow |F_e=1, m_F=0\rangle$ is forbidden, atoms in $|F_g=1, m_F=0\rangle$ does not participate in the MTS process; while for $\text{lin} \perp \text{lin}$, all magnetic sublevels of $F_g=1$ contribute to the MTS signal, so that the amplitude of

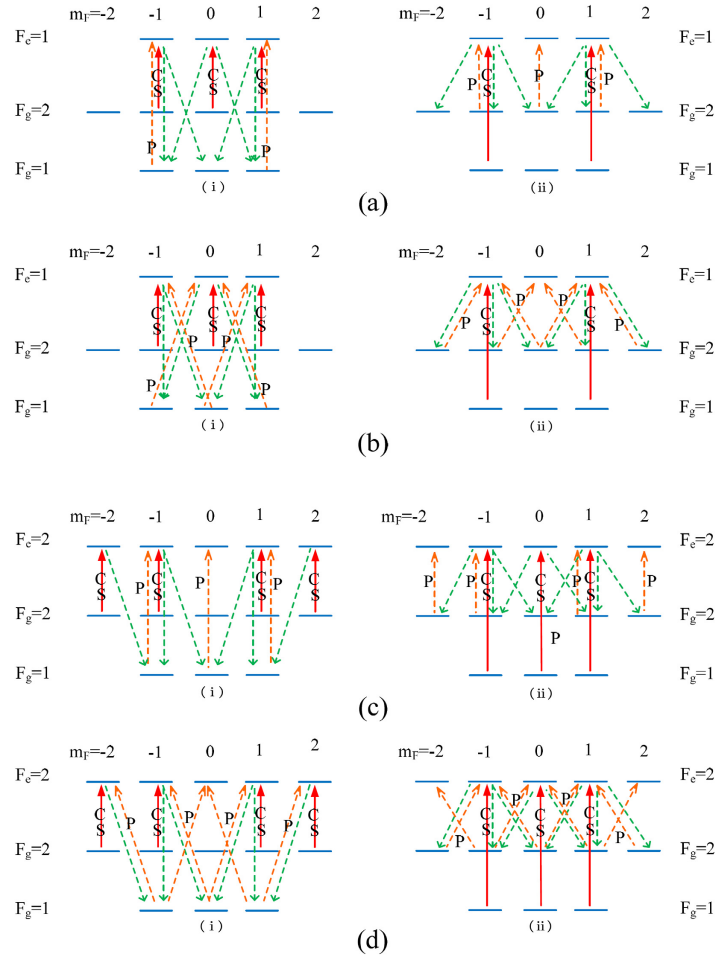


Fig. 7. The energy level diagrams for the crossover signal under the parallel (a, c) and perpendicular (b, d) linear polarization configurations. The transitions and spontaneous emissions of the two components, $F_g = 1, 2 \rightarrow F_e = 1$ (a, b) and $F_g = 1, 2 \rightarrow F_e = 2$ (c, d), are displayed in the diagrams.

the crossover signal in the lin $\perp$ lin configuration is higher than that in the lin||lin configuration. When the pump beam is tuned to $F_g = 1 \rightarrow F_e = 1$, the amplitude of the crossover signal in the lin||lin configuration is higher.

The component of $F_g = 1, 2 \rightarrow F_e = 2$ (c, d) results from the situation when the pump beam is tuned to $F_g = 2 \rightarrow F_e = 2$ and the probe beam is tuned to $F_g = 1 \rightarrow F_e = 2$, there is no forbidden transition in either the lin||lin configuration or the lin $\perp$ lin configuration. According to Eq. (4), the crossover signal can be described as

$$S(\omega_m) \propto \eta \cdot \rho_{C,S} \cdot \gamma_1 \cdot \rho_P \cdot \gamma' \quad (7)$$

where γ' is the spontaneous emission probability, $\rho_{C,S}$ are the relative transition probabilities under the action of pump beam containing carrier and sideband components. According to Eq. (5) and Eq. (6), the amplitude of the crossover signal in the lin $\perp$ lin configuration is higher than that in the lin||lin.

4.4. The Residual Amplitude Modulation (RAM)

The asymmetric resonance peak in MTS of lithium is caused by RAM. When the RAM is superimposed on symmetric peak in MTS, the zero level shifts, thus results in an asymmetric profile. The RAM is generated by EOM [23], or beam overlap [24], or absorption medium [25, 26]. The absorption coefficient of a laser beam passing through the absorption medium depends on its frequency. The pump beam includes two sidebands, which are separated in frequency by 2Ω (Ω is the modulation frequency). Due to the nonlinear interaction, the different absorption coefficients of two pump sidebands cause RAM in the probe beam. The RAM arising from EOM and beam overlap affects all spectral features equally, while the RAM arising from absorption medium affects spectrum differently. According to experimental results, the RAM in MTS of lithium is caused mainly by absorbing medium. The magnitude of the RAM sidebands generated by this process [26] is given by

$$r_{medium} = \frac{1}{2} r_{exit} = \frac{\exp[-\alpha(\omega + \Omega)\frac{L}{2}] - \exp[-\alpha(\omega - \Omega)\frac{L}{2}]}{4}, \quad (8)$$

where

$$\alpha(\omega) = \sum_{i=1}^N \alpha_i \exp\left[-\frac{(\omega - \omega_i)^2}{\sigma_i^2}\right], \quad (9)$$

and L is the length of the absorption cell, σ_i is the Doppler broadened width of hyperfine transitions with frequency of ω_i , $\alpha(\omega)$ is absorption coefficient which is the linear combination of the N hyperfine components. If all of the hyperfine transitions have equal absorption coefficient (α_0) and Doppler broadened width (σ_0), then

$$\alpha(\omega) = \alpha_0 \sum_{i=1}^N \exp\left[-\frac{(\omega - \omega_i)^2}{\sigma_0^2}\right]. \quad (10)$$

For the D_2 transitions in ^7Li ,

$$\alpha(\omega) = \alpha_0 \left\{ \exp\left[-\frac{(\omega - \omega_1)^2}{\sigma_0^2}\right] + \exp\left[-\frac{(\omega - \omega_2)^2}{\sigma_0^2}\right] \right\}. \quad (11)$$

Figure 8 shows the dependence of relative amplitude of RAM on laser frequency detuning, as depicted by Eq. (8). Here, we assume that all of the hyperfine transitions have equal absorption coefficient (α_0) and Doppler broadened width (σ_0), and for convenience the value of α_0 is

assumed to be 1, and the detuning of resonance peak $[2, F']$ is defined as zero. The inset of Fig. 8 shows the calculated result of RAM. The relative amplitudes of RAM at two resonance peaks are non zero and their signs are opposite, while the relative amplitude at the crossover is close to zero. When this kind of RAM signal is superimposed on the original symmetric MTS signal, the profile of crossover peak does not change obviously, but the profiles of resonance peaks do change and their zero level lines shift oppositely. We also consider the situation that the absorption coefficients for the two hyperfine transitions may be different. In this case, the zero level line in Fig.8 does shift, which means that the RAM at the crossover is not negligible.

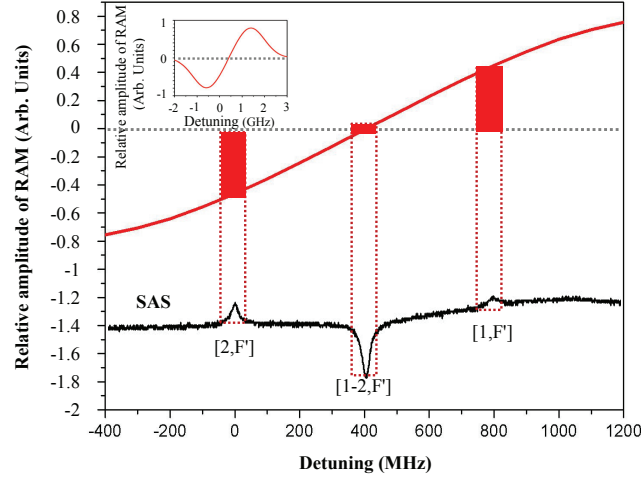


Fig. 8. The relative amplitude of RAM. The inset at the top left shows the calculated result in a large frequency range. The gray dashed line means the zero level of RAM, and the red shades are the amplitude of the RAM.

According to [27], the spectral line disturbed by the RAM can be described as the superposition of an ideal In-phase part of MTS and a perturbation part,

$$\text{Sig}(f) = C \left[\begin{array}{l} \{L(-\omega_m, f) - L(-\omega_m/2, f) + L(\omega_m/2, f) - L(\omega_m, f)\} + \\ r\{L(-\omega_m, f) + L(-\omega_m/2, f) + L(\omega_m/2, f) + L(\omega_m, f)\} \end{array} \right], \quad (12)$$

where

$$L(\omega_m, f) = \frac{\Gamma^2}{\Gamma^2 + (f_0 - f - \omega_m)^2}. \quad (13)$$

Here, Γ is the half line-width at half maximum, ω_m is the modulation frequency, f_0 is the centre frequency of the transition, f is the frequency of laser, r is the magnitude of RAM relative to MTS signal.

Figure 9 shows the fitting of the data from the in-phase part in MTS of the resonance peak $[F=2, F']$ under $\text{lin} \perp \text{lin}$ polarization configuration. According to the fitting result, we can calculate the amount of RAM in the signal, $r = -0.024(1)$. It can be seen that even a small value of r , can cause great influence to the line. The vertical dashed line shows the centre frequency of the resonance transition and the horizontal dashed line represents the zero line. From Fig. 9 we see there is an offset in the zero-crossing of the signal because of the existence of the RAM. As already mentioned before, for phase to 50° , we can get a symmetrical signal of MTS

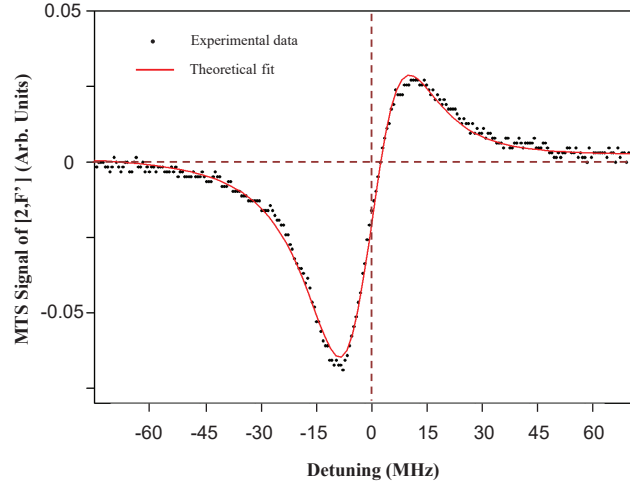


Fig. 9. A theoretical fit to the data from the in-phase part in MTS of the resonance peak $[F=2,F']$ under $\text{lin} \perp \text{lin}$ polarization configuration with $r = -0.024(1)$.

for the crossover resonance peak. After the data fitting, we got that $r = 0.002(2)$. In this case, the proportion of RAM decreases greatly and the disturbance of the spectral line is greatly suppressed.

5. Conclusions

In conclusion, we have investigated the modulation transfer spectroscopy of D_2 transitions of ^7Li in a lithium atomic vapor cell. Firstly we have researched the role of the intensity of the probe beam in the spectrum. With using the high intensity ($4I_{\text{sat}}$) of probe beam, the MTS spectra of lithium shows unique features because of the special energy level structure. The dependence of the MTS on laser polarization is studied. From the spectrum of MTS we see, the signal to noise ratio (SNR) of signals corresponding to the resonance peak $[2,F']$ and crossover peak $[1-2,F']$ are both high, about 23:1, and suitable for frequency locking. To get a higher value of SNR the $\sigma^+ - \sigma^+$ ($\sigma^- - \sigma^-$) configuration and $\text{lin} \perp \text{lin}$ configuration should be considered. The distortion of MTS due to RAM is also analyzed, the fitting results show that the RAM in the signal of crossover peak can be compensated by tuning the phase. However, from the slope and offset in the zero-crossing of the signal to see, coupled with the less RAM, the MTS signal of the crossover peak for the phase of 50° is more suitable for laser locking. Of course, in order to optimize the spectrum of MTS, parameters such as the modulation index [28] and modulation frequency should be carefully improved further. All of these results are useful for optimizing the MTS signal and for laser stabilization with the spectrum.

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