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Li_2 Autler-Townes 分裂的实验观察*

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摘 要： 用光学-光学双共振激光光谱研究了 $^7Li_2$ A $^1\Sigma_u^+$ 态的 Autler-Townes（A-T）分裂. 一个强的耦合场（泵浦激光）激发 $^7Li_2$ A $^1\Sigma_u^+$ v′，J′← X $^1\Sigma_g^+$ v″，J″跃迁，诱发 A $^1\Sigma_u^+$ v′，J′能级和 X $^1\Sigma_g^+$ v″，J″能级的 A-T 分裂. 另一个探测激光从 A $^1\Sigma_u^+$ v′，J′能级进一步激发到 4 $^1\Sigma_g^+$ 态. 扫描探测激光，监测 4 $^1\Sigma_g^+$ 态碰撞诱导紫色荧光，从而探测 A $^1\Sigma_u^+$ v′，J′能级的 A-T 分裂. 当耦合场频率偏离共振时，激发光谱线出现双重分裂. 在该实验条件下，分裂大小和泵浦激光频率偏离共振频率的失谐量成正比. 研究了 A-T 分裂的两条线的相对强度与泵浦、探测光的强度及缓冲气体压力的关系.

关键词： Li_2；Autler-Townes 分裂；ac Stark 效应；Li_2 A $^1\Sigma_u^+$ 态；OODR
中图分类号： O643.2 **文献标识码：** A

Experimental Observation of Autler-Townes Splitting of Li_2 *

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Abstract Autler-Townes (A-T) splitting of the A $^1\Sigma_u^+$ state of $^7Li_2$ has been experimentally studied by pulsed optical-optical double resonance (OODR) spectroscopy. A stronger coupling field (the pump laser) excited an A $^1\Sigma_u^+$ v′, J′← X $^1\Sigma_g^+$ v″,J″ transition and induced the A-T splitting of the A $^1\Sigma_u^+$ v′, J′ and X $^1\Sigma_g^+$ v″,J″ levels, while a detecting laser (the probe laser) detected the A-T splitting of the intermediate A $^1\Sigma_u^+$ v′, J′ level by the further exciting to the 4 $^1\Sigma_g^+$ state and monitored the collision-induced violet fluorescence from the upper 4 $^1\Sigma_g^+$ level. When the coupling laser frequency was tuned off resonance, the excitation signals displayed doublet splitting which was proportional to the frequency detuning of the coupling laser under the experimental conditions. The dependence of relative intensities of the two lines on coupling and detecting laser intensities and buffer gas pressure was studied.

Key words Li_2, Autler-Townes splitting, ac Stark effect, A $^1\Sigma_u^+$ state of Li_2, OODR

1 Introduction

One of the most important aspects of nonlinear interactions is that a strong field alters the properties of a physical system. One of these is ac Stark effect, which includes the Autler-Townes (A-T) splitting and ac Stark shift[1], gain without inversion[2,3], and electromagnetically induced transparency[4,5], etc.

The A-T splitting, which is well studied in atomic systems, has only been investigated in gas-phase molecules in recent years[6-11]. Quesada *et al*. observed A-T splitting in the multiphoton ionization of H_2 and

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proposed a new method to measure the transition moment between excited electronic states[8]. Girard *et al.* examined the polarization dependence of the ac Stark effect on the 2 + 2 REMPI spectrum of N_2[9]. Xu *et al.* studied A-T splitting and ac Stark shift of CO by using REMPI[10]. Recently Qi *et al.* demonstrated that the A-T splitting could be used to facilitate the all-optical control of molecular angular momentum alignment[11].

In this paper, we report experimental observation of the A-T splitting of the $A^1\Sigma_u^+$ state of $^7Li_2$ by optical-optical double resonance (OODR) spectroscopy.

The ac Stark effect can be described with the dressed state theory[12]. When a coupling laser beam interacts with a two level system, it will induce the A-T splitting for each level. One dressed state is called a real state, and another is called a virtual state with one photon added to or subtracted from a real state. The separation S for the two dressed levels of the "molecule + field" system is given by

$$S = (\triangle^2 + \Omega^2)^{1/2} \quad (1)$$

Here $\Omega \equiv D(R)E/\hbar$ is the Rabi frequency and $D(R)$ is the electric dipole transition moment induced by the coupling laser with electric-field amplitude E and frequency detuning $\triangle$. In the OODR experiment, another detecting laser is used to probe these two new eigenlevels by further transition to a third level. One can see from Eq. (1) that when $\Omega << \triangle$, $S \approx \triangle$, i. e. S is proportional to laser frequency detuning $\triangle$.

2 Experiment

The experimental setup is similar to that described in Refs. [13,14]. In brief, lithium vapor was generated in a crossed heatpipe oven with about 133 Pa argon buffer gas. The lithium vapor temperature was about 1000 K. An excimer laser (Lambda Physik EMG 202MSC) pumped two dye lasers (Lambda Physik FL3002E), which served as the coupling and detecting lasers, respectively. The two dye lasers, whose line widths were about 0. 04 cm^{-1} with an intracavity étalon, counter-propagated and overlapped at the center of the heat pipe oven. Dye laser frequencies were calibrated using the standard iodine calibration[15]. The coupling laser excited the A $^1\Sigma_u^+$ v′ = 5, J′ = 20 ← X $^1\Sigma_g^+$ v″ = 0, J″ = 19 or 21 transition[16,17], the detecting laser further excited transition to the 4 $^1\Sigma_g^+$ v = 4, J = 21 upper level[18]. Fig. 1 gives the energy level diagram. The OODR excitation signals were detected by monitoring the collision-induced violet fluorescence with a filtered photomultiplier (PMT). The output of the PMT was integrated by a SR250 Boxcar and recorded in a computer.

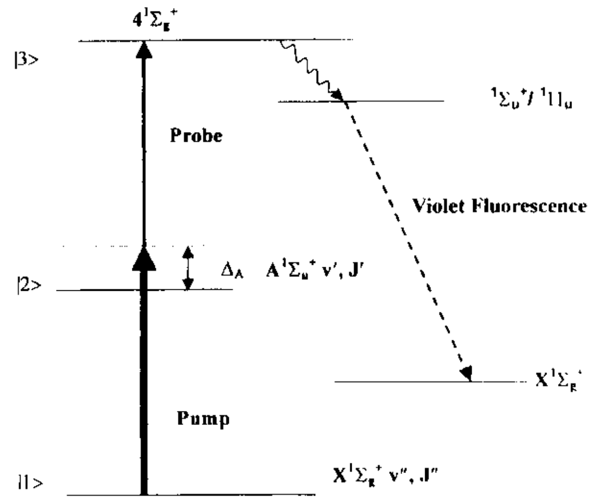


Fig. 1 Energy level diagram of the Li_2 OODR excitation scheme

3 Results

3.1 Splitting vs. coupling laser detuning

When the coupling laser was held fixed at the center of the Doppler profile of the A $^1\Sigma_u^+$ v′ = 5, J′ = 20 ← X $^1\Sigma_g^+$ v″ = 0, J″ = 19 transition, the detecting laser was scanned through the predicted frequency of the 4 $^1\Sigma_g^+$ v = 4, J = 21 ← A $^1\Sigma_u^+$ v′ = 5, J′ = 20 transition and collision-induced violet fluorescence from the 4 $^1\Sigma_g^+$ v, J upper level was monitored. An OODR excitation line appeared (Fig. 2(a)) at the predicted position. With our laser linewidth and pump and probe intensities (4.2 and 1.8 W/mm^2, respectively), no A-T splitting was resolved on this spectral line.

When the coupling laser frequency was detuned by $\triangle$ away from the center of the Doppler profile of the A $^1\Sigma_u^+$ v′ = 5, J′ = 20 ← X $^1\Sigma_g^+$ v″ = 0, J″ = 19 transition ($\triangle = \omega_{pump} - \omega_{12}$), in addition to the peak via the A $^1\Sigma_u^+$ v′ = 5, J′ = 20 level, a second peak appeard when the detecting laser frequency was scanned. The detecting laser frequency of the OODR excitation line via the A $^1\Sigma_u^+$ v′ = 5, J′ = 20 level was not changed when the coupling laser frequency was off resonance with our measurement accuracy, but the detecting laser frequency of the additional peak changed with the detuning: $\omega_{detecting} = \omega_{23} + \triangle$. The peak via the A $^1\Sigma_u^+$ level, whose frequency was not changed with detuning, was called real line and the additional line whose frequency changes with detuning was called a creeper line. Fig. 2(b) ~ (f) show the real and creeper lines of the 4 $^1\Sigma_g^+$ v = 4, J = 21 ← A $^1\Sigma_u^+$ v′ = 5, J′ = 20 transition recorded at different pump coupling laser detuning.

When the coupling laser frequency was tuned at the center of the Doppler profile of the A $^1\Sigma_u^+$ v′, J′ ← X $^1\Sigma_g^+$ v″, J″ transition, $\triangle = 0$ and $S = \Omega$ according to Eq. (1). We did not resolve the splitting (Fig. 2 (a)), indicating that the Rabi frequency Ω under our experimental conditions was smaller or comparable with our laser linewidth and Doppler width (0.04 and 0.1 cm^{-1}, respectively). For a linearly polarized laser light[5],

$$\Omega = D(R)\, E/\hbar = (\mu_{\parallel} E/\hbar)\, |\langle \nu_1 | \nu_2 \rangle|\, F^0_{\Sigma-\Sigma} \quad (2)$$

where $F^0_{\Sigma-\Sigma}$ is the transition dipole moment orientation factor for $^1\Sigma \rightarrow {}^1\Sigma$ electronic transition and equals to $[(2J+1)(2J+3)]^{-1/2}[(J+1)^2 - M^2]^{1/2}$ for R-branch when linearly polarized light is used, $D(R)$ is the M-dependent transition dipole moment, $\mu_{\parallel}$ is the $^1\Sigma \rightarrow {}^1\Sigma$ electronic transition dipole moment, and $|\langle \nu_1 | \nu_2 \rangle|^2$ is the Franck-Condon factor. For A $^1\Sigma_u^+$ v′ = 5, J′ = 20 ← X $^1\Sigma_g^+$ v″ = 0, J″ = 19 transition, $|\langle \nu_1 | \nu_2 \rangle|^2 = 0.113$ and $\mu = 2.4$ a. u. [19], an estimated Rabi frequency for $M = 12$ was $\Omega \approx 0.02$ cm^{-1} [5] under our experimental conditions, which could not be resolved with our resolution.

Fig. 3 shows the splitting S vs. coupling laser detuning △ for the A $^1\Sigma_u^+$ v′ = 5, J′ = 20 ← X $^1\Sigma_g^+$ v″ = 0, J″ = 19 transition under our experimental conditions. That $S \approx \triangle$ was consistent with our discussion when $\triangle \gg \Omega$.

Similarly, when a stronger probe laser was used as the coupling field with detuning $\triangle = \omega_{probe} - \omega_{23}$ for

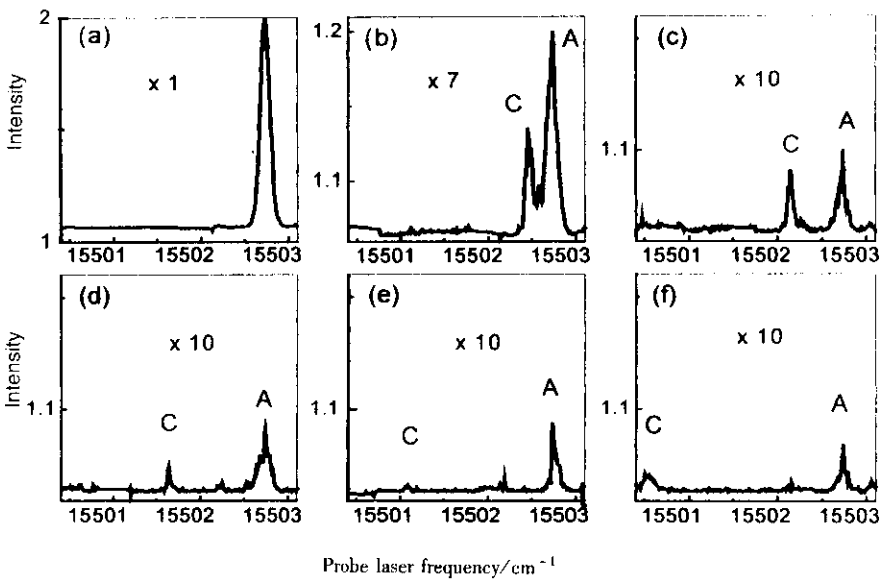
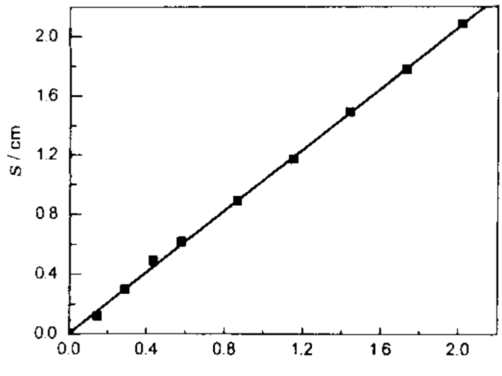


Fig. 2　Real and creeper lines of the 4 $^1\Sigma_g^+$ v = 4, J = 21 (A $^1\Sigma_u^+$ v′ = 5, J′ = 20 ← X $^1\Sigma_g^+$ v″ = 0, J″ = 19 transition when the pump laser frequency had different detunings (a) △ = 0; (b) △ = 0.28 cm^{-1}; (c) △ = 0.55 cm^{-1}; (d) △ = 1.11 cm^{-1}; (e) △ = 1.66 cm^{-1}; (f) △ = 2.22 cm^{-1}. The pump and probe laser intensities were 4.2 and 1.8 W/mm^2, respectively. The real line is marked on top with A, and the creeper line is marked with C. The sensitivities of (b) and (c) ~ (f) are higher than (a) by 7 and 10 times, respectively.



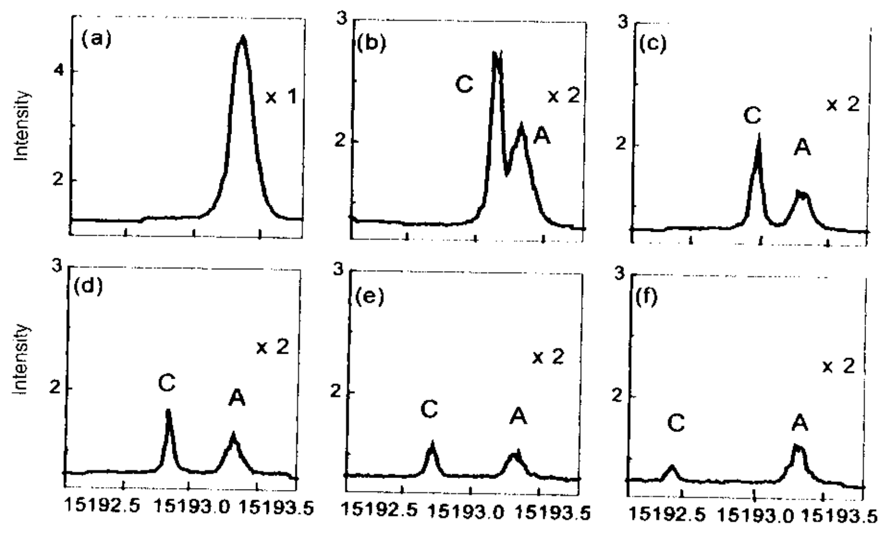
$\triangle = \omega_{pumbe} - \omega_{12}/cm^{-1}$

Fig. 3 A-T splitting separations S versus the pump lasers detuning Δ The pump and probe lasers excited the 4 $^1\Sigma_g^+$ v = 4, J = 21 ← A $^1\Sigma_u^+$ v′ = 5, J′ = 20 ← X $^1\Sigma_g^+$ v″ = 0, J″ = 19 transition, and their intensities were 4.2 and 1.8 W/mm², respectively.

the 4 $^1\Sigma_g^+$ v, J ← A $^1\Sigma_u^+$ v′, J′ transition, A-T splitting of the A $^1\Sigma_u^+$ v′, J′ level was also observed when a weak pump laser (now it served as a detecting laser) was scanned and collition-induced violet fluorescence from the upper 4 $^1\Sigma_g^+$ v = 4, J = 21 level was monitored. Fig. 4(b) ~ (f) show the real and creeper lines when the probe laser frequency had different detunings. The intensities of the pump (detecting) and probe (coupling) lasers were 1.6 and 21 W/mm², respectively.

3.2 Intensities of the real and creeper lines vs. laser intensities

A-T splitting is a nonlinear effect. Not only the splitting S is laser intensity dependent, but also the intensities of the real and creeper lines (both absolute as well as relative intensities) are laser intensity dependent. Under our experimental conditions, the laser intensity dependence of the splitting S was not observed, but the laser intensity dependence of the intensities of the real and creeper lines has been observed. Fig. 5 (a1) ~ (a3) show the intensities of the real and the creeper lines of the 4 $^1\Sigma_g^+$ v = 4, J = 21 ← A $^1\Sigma_u^+$ v′ = 5, J′ = 20 ← X $^1\Sigma_g^+$ v″ = 0, J″ = 21 transition at different probe (detecting) laser intensities when pump (coupling) laser intensity was 130 W/mm². We can see that not only the intensities of both lines increased when the probe (detecting) laser intensity increased, but the relative intensity of the creeper line also increased. Fig. 5(b1) ~ (b3) show the intensities of the two lines at different pump intensities when the probe intensity was 7.2 W/mm². In this case, the intensities of both lines increased when the pump laser intensities



Pump laser frequency/cm⁻¹

Fig. 4 Real and creeper lines of the 4 $^1\Sigma_g^+$ v = 4, J = 21 ← A $^1\Sigma_u^+$ v′ = 5, J′ = 20 ← X $^1\Sigma_g^+$ v″ = 0, J″ = 19 transitions when the probe laser frequency had different detunings and pump laser was scanned (a) △ = 0; (b) △ = 0.14 cm⁻¹; (c) △ = 0.29 cm⁻¹; (d) △ = 0.43 cm⁻¹; (e) △ = 0.58 cm⁻¹; (f) △ = 0.72 cm⁻¹. The pump and probe laser intensities were 1.6 and 21 W/mm², respectively. The real line is marked on top with A, and the creeper line is marked with C. (b) ~ (f) were recorded at higher sensitivity than (a) by a factor of 2.

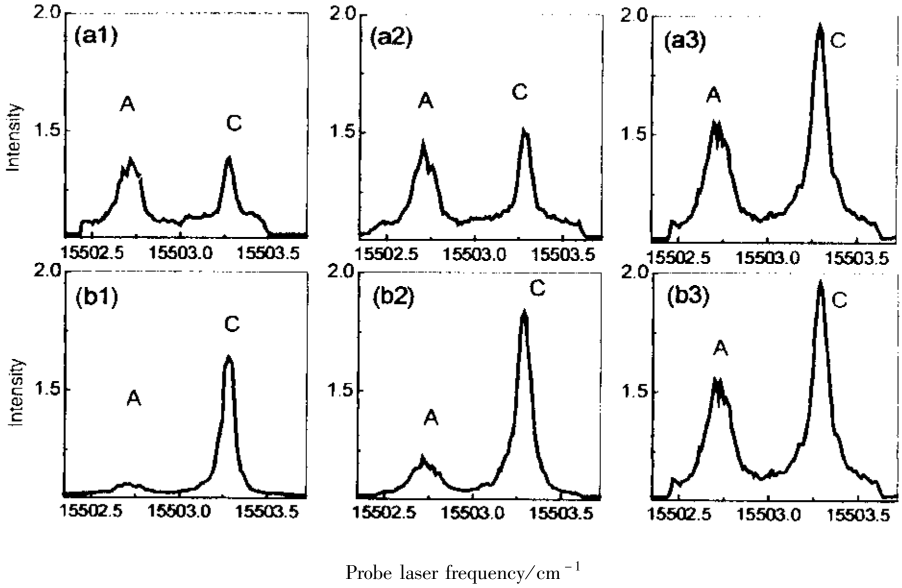


Fig. 5　Laser intensity dependence of the real and the creeper lines of the 4 $^1\Sigma_g^+$ v = 4, J = 21←A $^1\Sigma_u^+$ v′ = 5, J′ = 20 ←X $^1\Sigma_g^+$ v″ = 0, J″ = 21 transitions when the pump laser detuning △ = 0.55 cm^{-1}
(a1) ~ (a3): $I_{pump} = 1.3\times10^2$ W/mm^2, I_{probe} = 1.2, 1.8 and 7.2 W/mm^2, respectively;
(b1) ~ (b3): $I_{probe} = 7.2$ W/mm^2, I_{pump} = 34, 68 and 1.3×10^2 W/mm^2, respectively.

increased. In Fig. 5(a) and (b), the creeper line looked narrower than the real line and its relative intensity increased while the relative intensity of the probe laser increased under our experimental conditions.

4　Discussion

In our OODR spectra, the creeper lines could also come from two-photon transitions (one photon from the pump laser and one photon from the probe laser) since the sum of energies of the two photons equaled to the energy difference between the upper level and the ground level. This kind of two-photon transition had been observed in Na atom by Liao *et al.* [20]. In their experiment, two-photon transitions from the virtual and real levels were observed with the sub-Doppler resolution when the buffer gas pressure was not zero. However, no resonance transition (the real line) was observed when the buffer gas pressure was zero. When the pressure increased, the broader collision induced resonance line appeared.

In our experiment, the real line always appeared under our experimental condition (106 ~ 293 Pa). Fig. 6(a) ~ (c) show the pressure dependence of the intensities of the real and creeper lines when the probe laser detuning △ = 0.55 cm^{-1}. In this case, the relative intensities of the two lines had no big changes with our accuracy. The relative intensities between the real and creeper lines were not strongly pressure dependent, indicating that the real line in our spectra was not collision induced, but one of the components of the A-T splitting.

5　Conclusions

Autler-Townes splitting of the A $^1\Sigma_u^+$ mixed levels of $^7Li_2$ has been observed by the OODR excitation spectroscopy. When the pump (coupling) laser frequency was detuned in addition to the OODR excitation lines via the A $^1\Sigma_u^+$ level, a creeper line was also observed. The real and creeper line intensities increased as the laser intensities increased. The relative intensities of the real and the creeper lines strongly depend on the intensity ratio of pump to probe laser. Collision did not play an important role in our experiment. Rabi frequency was not resolved and the splitting was equal to the detuning under our experimental conditions.

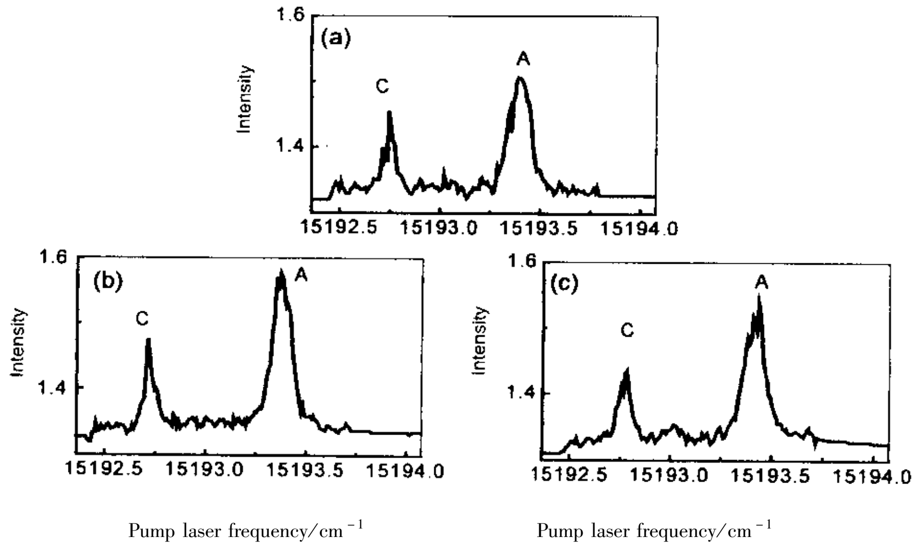


Fig. 6 Pressure dependence of the intensities of the real and the creeper lines of the $4\ ^1\Sigma_g^+\ v=4$, $J=21 \leftarrow A\ ^1\Sigma_u^+\ v'=5$, $J'=20 \leftarrow X\ ^1\Sigma_g^+\ v''=0$, $J''=19$ at three different pressures (a) 106 Pa, (b) 186 Pa, (c) 293 Pa. The probe laser frequency was held fixed at detuning $\triangle = 0.55\text{cm}^{-1}$ while the pump laser was scanned. The pump and probe laser intensities were 1.6 and 21 W/mm^2, respectively.

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