Enhancement of signal-to-noise ratio of ultracold polar NaCs molecular spectra by phase locking detection*

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(Received 27 September 2017; revised manuscript received 23 October 2017; published online 20 November 2017)

We report a method of high-sensitively detecting the weak signal in photoassociation (PA) spectra of ultracold NaCs molecules by phase sensitive-demodulated trap-loss spectra of Na atoms from a photomultiplier tube. We find that the signal-to-noise ratio (SNR) of the PA spectra is strongly dependent on the integration time and the sensitivity of the lock-in amplifier, and our results show reasonable agreement with the theoretical analyses of the SNR with the demodulation parameters. Meanwhile, we investigate the effect of the interaction time of the PA laser with the colliding Na–Cs atom pairs on the SNR of the PA spectra. The atom loss rate is dependent on both the PA-induced atom loss and the loading of the MOT. The high-sensitive detection of the excited ultracold NaCs molecules lays a solid foundation for further study of the formation and application of ultracold NaCs molecules.

Keywords: ultracold NaCs molecules, photoassociation, high-resolution spectroscopy, signal-to-noise ratio

PACS: 37.10.Pq, 33.15.Mt, 33.80.Wz **DOI:** 10.1088/1674-1056/26/12/123701

1. Introduction

Ultracold polar molecules have drawn considerable interests over the last decade[1–3] due to their wide applications in precision measurements,[4–8] quantum simulation,[9,10] quantum information,[11,12] and controlled ultracold chemistry.[13,14] Preparation of ultracold polar molecules populating at the ground state has been realized in experiments by photoassociation (PA) and magneto-association (Feshbach resonance, FR) of ultracold atoms, followed by which the absolute rovibrational ground state molecules with $v'' = 0$ and $j'' = 0$ have been firstly obtained by stimulated Raman adiabatic passage (STIRAP) in ^{87}Rb^{40}K.[15] Feshbach resonance combined with STIRAP has been established as a standard method to acquire the ground state molecules. Very recently, ultracold dipolar molecular gases in the absolute rovibrational and hyperfine ground state have been realized in Bosonic ^{87}Rb^{133}Cs [16], ^{23}Na^{87}Rb[17] and Fermionic ^{23}Na^{40}K.[18] The ^{23}Na^{133}Cs molecule is among the most popular diatomic alkali heteronuclear dimmers because of several distinctive merits: the ultracold ground state NaCs molecules have a large electric dipole moment (EDM) of 4.6 Debye (only smaller than 5.5 Debye of LiCs) and the energetically forbidden two-body chemistry reactions.[3] However, the NaCs Feshbach molecules have not yet been produced in experiment although the resonant positions have been theoretically predicted.[19] Ultracold NaCs molecules were formed in a typical magneto–optical trap (MOT) by Bigelow and co-workers for the first time by PA in 2004,[20] since then the PA spectroscopy (PAS) of NaCs at various electronic and vibrational states has been obtained using the ionization spectroscopic detection method.[21,22]

Whenever light is used to prepare ultracold polar molecules in a controlled way or to manipulate their internal state, precise knowledge of the molecular structure is required. In our efforts to create ultracold dipolar Na^{23}Cs^{133} ground state molecules, we have obtained the PAS of near-dissociation levels of ultracold NaCs molecules and studied their hyper-fine structure by the trap-loss spectroscopy.[23] This technique is a unique tool to detect the molecular long-rang states, which has provided a wealth of information about the molecular structures in the near-dissociation region. In particular, the trap-loss spectroscopy of the trapped ultracold atoms has often served as a high resolution PAS for the rovibrational or hyperfine structures of the excited molecules.[24] The trap-loss spectroscopy not only demonstrates the resonant frequency of the PA resonances,[25,26] but also carries information about the relative transition strengths. However, it suffers from the low sensitivity on account of the small Franck–Condon factors (FCFs), which leads to limited understanding of the molecular structure. The spectral signal-to-noise ratio (SNR)[27,28] remains to be improved. Recently, an FR-optimized PA has

*Project supported by the National Key Research and Development Program of China (Grant No. 2017YFA0304203), the ChangJiang Scholars and Innovative Research Team in the University of the Ministry of Education of China (Grant No. IRT13076), the National Natural Science Foundation of China (Grant Nos. 91436108, 61378014, 61675121, 61705123, and 61722507), the Fund for Shanxi "1331 Project" Key Subjects Construction, China, and the Foundation for Outstanding Young Scholars of Shanxi Province, China (Grant No. 201601D021001).

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been proposed theoretically[29] and achieved experimentally for enhancing the PA rate.[30] In addition, the modulation spectroscopic approach has been widely used to improve the SNR of PAS.[31] However, as a critic channel to improve the detection sensitivity of the spectroscopy, the demodulation technique has not yet been investigated in detail.

In this paper, we report our research on the SNR of the PAS by demodulation of the trap-loss spectroscopy of ultracold NaCs molecules, which are produced in a dual-species MOT. The dependence of the SNR of PAS on the demodulation parameters is measured for the first time, and our theoretical analysis shows a good agreement with the experimental results. In addition, we investigate the effect of the interaction time between the PA laser and the colliding Na–Cs atom pairs on the SNR of the PA spectra by changing the scanning time of the PA laser. Theoretical analyses of the relationship between the SNR and the MOT loading time as well as the PA induced trap loss show good accordance with the experimental results.

2. Experimental approach and setup

The experimental setup is schematically depicted in Fig. 1(a). The sample of ultracold Cs atoms and Na atoms is prepared in a standard vapor-loaded magneto-optical trap (MOT).[23,32] Two commercial lasers (Toptica, DL pro, $\sim$ 80 mW with line-width of 0.8 MHz) provide the trapping and repumping lasers for the Cs MOT, and another high-power, tunable, frequency-doubled diode laser (Toptica, TA-SHG pro, $\sim$ 1 W with line-width 0.5 MHz) is employed to obtain the dark-SPOT MOT for the Na atoms. The frequencies of all lasers are stabilized using the standard saturated absorption spectroscopy (SAS), and then are tuned to several particular detunings to cool and trap the Cs and Na atoms in combination with a pair of magnetic field coils in the anti-Helmholtz configuration with a gradient of $\sim$ 12 Gs/cm (1 Gs $= 10^{-4}$ T). A modulation frequency of 3.0 kHz is used in stabilizing the frequencies of the trapping lasers for the Cs and Na atoms. The numbers of Cs and Na atoms trapped in the MOT are measured to be about 6×10^7 and 7×10^7 using the absorption method, with densities of 1.5×10^9 cm^{-3} and 2×10^9 cm^{-3} and temperatures of 120 μK and 150 μK, respectively.

The PA of the ultracold Cs and Na atoms is induced by illuminating a PA laser on the trapped atoms with a $1/e^2$ diameter of 0.78 mm and a maximum available average intensity of $\sim$ 750 W/cm^2. This laser is provided by a widely tunable continuous-wave Ti: sapphire laser system (Coherent MBR 110, linewidth $\sim$ 100 kHz). The long time frequency drift of the laser is suppressed within 500 kHz by locking to its self-reference cavity. The absolute frequency of the PA laser is measured by a wavelength meter (High Finesse-Angstrom WS/U) with an accuracy of 3 MHz. The wavelength meter is repeatedly calibrated against the Cs atomic hyperfine resonance transition, $6S_{1/2}(F = 4) \rightarrow 6P_{3/2}(F' = 5)$, at the beginning of each experimental cycle. When a pair of colliding ultracold Na and Cs atoms absorb a resonant photon provided

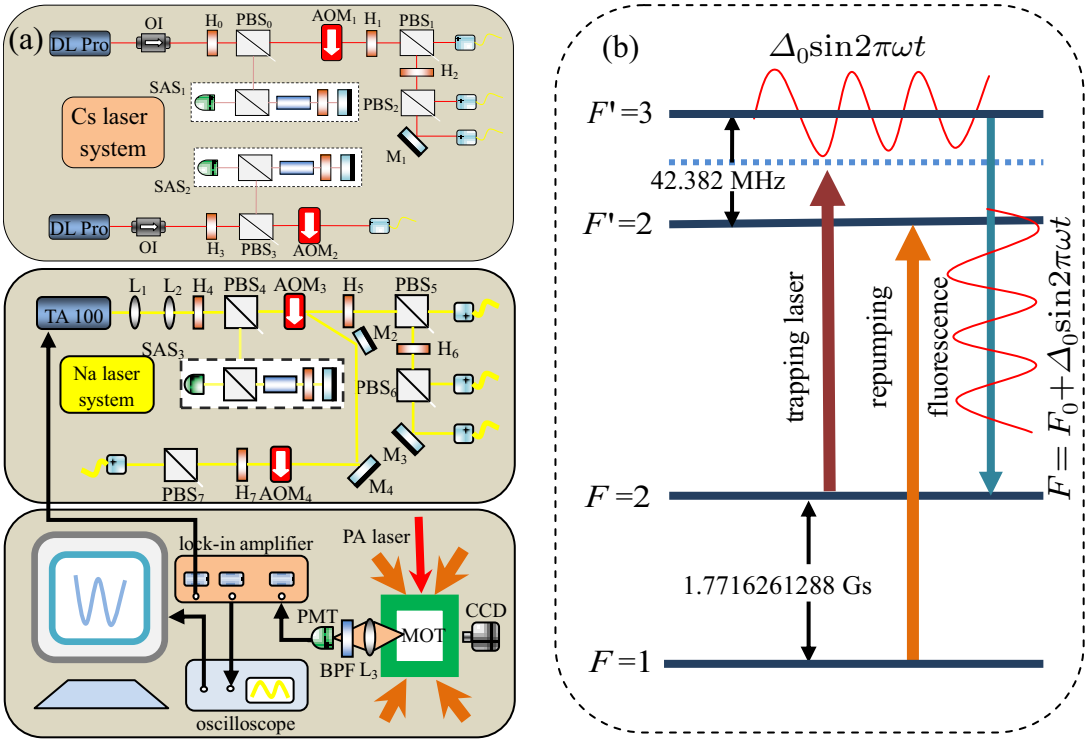


Fig. 1. (color online) (a) Experimental setup. OI: optical isolator; H: half wave plate; PBS: polarization beam splitter; AOM: acousto–optic modulator; SAS: saturated absorption spectroscopy; M: high reflective mirror; L: lens; BPF: band pass filter; PMT: photomultiplier tube; MOT: magneto-optical trap; CCD: charge-coupled device. (b) Modulation principle diagram for the ultracold Na atom.

by the PA laser, they convert into an excited molecule and then predominantly decay into two free atoms escaping from the trap. So we can detect the formation of ultracold molecules in PA by monitoring the fluorescence of atoms in the MOT as a function of the PA laser detuning frequency, which is recorded as the trap-loss spectroscopy of PA.

Due to the weakly spectral signals of ultracold polar NaCs molecules, a high-sensitive photomultiplier (PMT) is used to detect the fluorescence emitted from the Na MOT. In order to improve the detecting efficiency, the fluorescence is collected by using a 2-inch (1 inch = 2.54 cm) convex lens ($f = 50$ mm), and a few 589-nm band pass filters are used to shield the stray light from the Cs MOT. For achieving the optimized PA spectra, we use the method of phase-sensitive detection (PSD), which has been widely applied in the detection of weak and small signals. In the PA experiments, the number of cold atoms is modulated by changing the frequency detuning of the trapping laser with a function generator, so the fluorescence of the cold atoms is also modulated. Figure 1(b) shows the modulation scheme of photoassociation Na atom in the MOT. For the sine wave modulation, the instantaneous frequency of the trapping laser is given as $F(t) = F_0 + \Delta_0 \sin(2\pi\omega t)$, where F_0 is the center frequency about 10 MHz red detuned from the Na atomic transition $3S_{1/2}(F = 2) \rightarrow 3P_{3/2}(F' = 3)$, and the modulation amplitude is 2.23 V. $\Delta_0 \sin(2\pi\omega t)$ is the frequency of the modulation signal, Δ_0 is the modulation depth of frequency. A lock-in amplifier (Stanford Research SR830) is used to demodulate the modulated fluorescence of the ultracold Na and Cs atoms in MOT, where the sine signal used in the demodulation process is from the same frequency synthesizer used in the stabilization of the trapping laser with a different phase.

3. Results and discussion

Figure 2 shows the typical PA spectra of ultracold NaCs molecules in the vibrational level ($v = 61$) of the long-range state $c^3\Sigma^+_{\Omega=1}$ below the threshold ($3S_{1/2} + 6P_{3/2}$) at different sensitivities in the demodulation process. The sensitivity has a great effect on the PA spectroscopy, and an optimized value is needed to obtain high-resolution PA spectra. By analyzing the obtained PA spectra, the SNR of the PA spectroscopy is deduced. The variation of the SNR at different sensitivities is demonstrated in Fig. 3.

For a standard lock-in amplifier, the relationship between the amplitude of the output signal and the sensitivity can be expressed as[28]

$$V_{\text{out}} = (V_{\text{sig}}/S - V_{\text{of}}) \times 10E_{\text{x}}, \tag{1}$$

where V_{sig} is the detected signal from the PMT, S is the sensitivity of the lock-in amplifier, E_{x} is the amplification factor, and V_{of} is the bias voltage of the lock-in amplifier. The SNR is determined by $\text{SNR} = V_{\text{out}}/N$, where N is the noise of the PA spectral signal. The noise of the PA spectra is mainly due to the Johnson noise, $1/f$ noise, and relatively small shot noise of the lock-in amplifier. Here we regard the noise as a constant, therefore the SNR is proportional to the amplitude of the output signal. As demonstrated in Fig. 3, the experimental results for the variation of the SNR with the sensitivity are fitted using Eq. (1). The theoretical curve is in good accordance with the results. In order to obtain a high-resolution PA spectrum, the sensitivity of $S = 200$ mV is chosen as an optimized parameter of the lock-in amplifier.

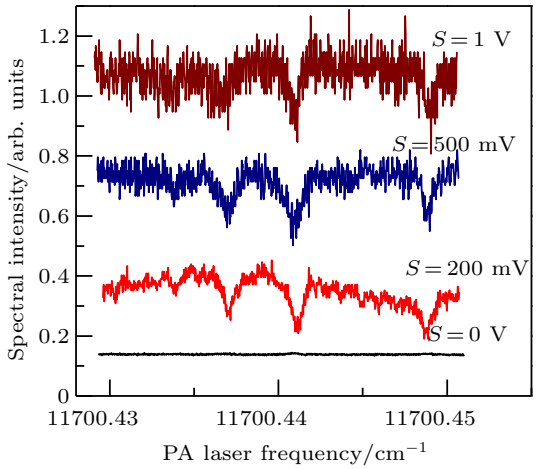


Fig. 2. (color online) PA spectra of ultracold NaCs molecule at different sensitivities of $S = 0$ V, 200 mV, 500 mV, and 1 V with the integration time of $t = 300$ ms.

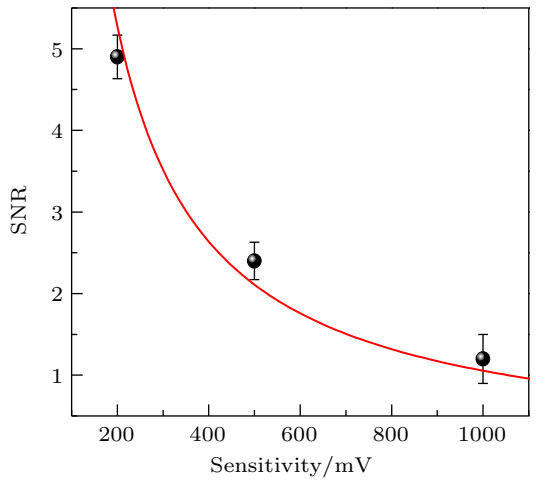


Fig. 3. (color online) The signal-to-noise ratio of ultracold NaCs molecule as a function of sensitivity S with the integration time of $t = 300$ ms. The red line is a fitting curve according to Eq. (1).

In Fig. 4, we study the effect of the integration time on the SNR of PA spectra. The integration time is an important parameter for a lock-in amplifier used in the detection of weak signal. For a sine reference signal and the detected signal $U_{\text{sig}}(t)$, the output signal $U_{\text{out}}(t)$ can be expressed as

$$U_{\text{out}} = \frac{1}{T}\int_{t-T}^{t} \sin(2\pi f_{\text{ref}} \cdot \tau + \phi) U_{\text{sig}}(t)\,\mathrm{d}\tau, \tag{2}$$

where the reference frequency $f_{\rm ref}$ is the modulation frequency of the PA spectra, and ϕ is a phase that is automatically set by the lock-in amplifier to match the detected PA spectral signal. There is a good fitting for the variation of the SNR of PA spectra with the integration time using Eq. (2), as shown in Fig. 4. The observed saturation of SNR with the integral time is also consistent with the general output of a lock-in amplifier for a long integral time, where the output can be expressed as $U_{\rm out} = \frac{1}{2}U_{\rm sig}\cos\theta$ with θ being the phase difference between the fluorescence signal and the reference signal. The integration time is large enough (e.g., much larger than the signal period) to suppress all unwanted parts with the frequency $f \neq f_{\rm ref}$ and the variations at twice the reference frequency.

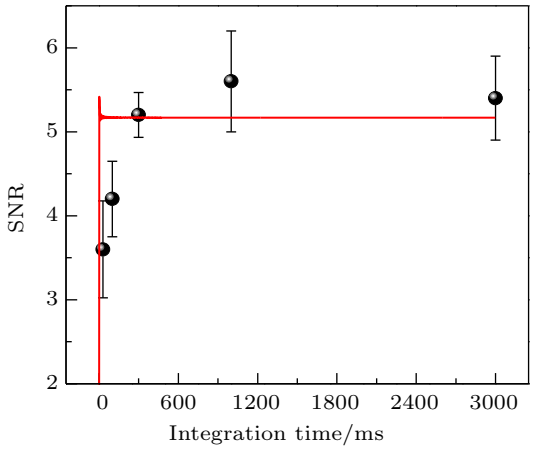


Fig. 4. (color online) The signal-to-noise ratio of ultracold NaCs molecule as a function of integration time t at sensitivity $S = 200$ mV. The red line is a fitting curve according to Eq. (2).

As shown in Fig. 5, the SNR is also strongly dependent on the scanning time of the PA laser, and this can be explained by investigating the effect of the scanning time on the PA-induced atom loss. The scanning time of the PA laser reflects the interaction time between the PA laser and ultracold colliding Na and Cs atoms. For the PA spectrum in a certain frequency detuning range, the atom loss increases with the scanning time and then reaches saturation when the scanning time is beyond a particular value. A model is introduced to describe the depletion of the trapped atoms with scanning time

$$\frac{{\rm d}N}{{\rm d}t} = -\Gamma N - \int K_{\rm PA} n^2 \, {\rm d}V, \tag{3}$$

where Γ is an exponential loss rate, $K_{\rm PA}$ is the PA rate, and n is the density of Na atoms in the MOT. The frequency of the PA laser is resonant with the PA transition, while the PA rate is independent of the local distribution of the atomic density. Both the loading rate of the ultracold Na atoms in the MOT and the corresponding PA rate are averaged on the time. Integration of the atomic density in the right-hand side of Eq. (3) yields a differential equation for the atom number N

$$\frac{{\rm d}N}{{\rm d}t} = -\Gamma N - \beta N^2, \tag{4}$$

where β is proportional to the PA rate $K_{\rm PA}$ at the resonant PA frequency. Thus the model equation to describe the PA-induced loss of atoms in the MOT is given as

$$N(t) = N_0 + \frac{A}{1 + {\rm e}^{-B(t-t_0)}}. \tag{5}$$

Here the parameters A and B are related to the coefficients Γ and β, and N_0 is the effective number of atoms that is used to transfer the number of atoms in the MOT to the number of the lost atoms induced by the PA. Here the maximum loss signal $V_{\rm out}$ in the detected PA spectrum is considered to be proportional to the atom loss, the theoretical fitting with Eq. (5) shows a reasonable agreement with the experimental results.

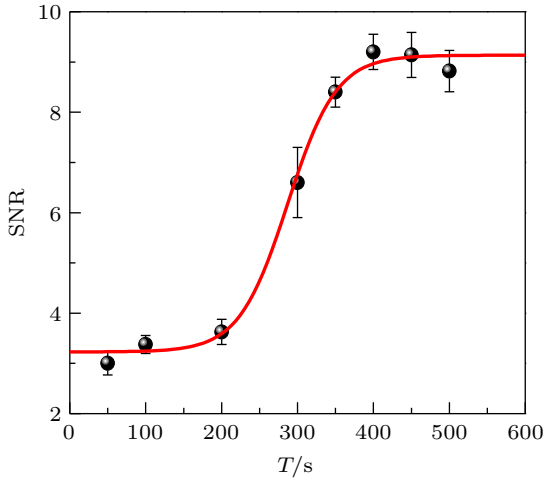


Fig. 5. (color online) The signal-to-noise ratio of ultracold NaCs molecule as a function of scanning time T at integration time $t = 300$ ms and sensitivity $S = 200$ mV. The red line is the fitting curve according to Eq. (5).

4. Conclusion

In summary, we report that a lock-in amplifier can be used to effectively demodulate and enhance the SNR of the PA spectra of ultracold NaCs molecules. High-resolution PA spectra are obtained by the phase sensitive-demodulated trap-loss spectra of Na atoms from a photomultiplier tube. We investigate the dependence of the SNR of the PA spectra on the integration time and sensitivity, which are set on the lock-in amplifier in the detection process. The theoretical variations of the SNR with different demodulation parameters show reasonable agreement with the experimental results for the sensitivity and integration time. We also find that the scanning time of the PA laser has a great effect on the SNR of the PA spectra, and analyze the dependence characteristic of the SNR on the interaction time of the PA laser with both ultracold Na and Cs atoms. We have obtained a good fitting for the variation of the SNR with the scanning time by using the model that characterizes the PA induced atoms loss with the loading time of the MOT. The high-sensitively demodulated PA spectra serve as a useful reference for researchers to obtain precise data of energy levels of excited states of ultracold NaCs or other polar

molecules. The present investigation can also be used to precisely evaluate the optimized level of the excited molecule to reach the ground state ultracold polar molecules by the stimulated Raman adiabatic passage.

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