Radiative deflection of a BaF molecular beam from the optical cycling

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We demonstrate a quasi optical cycling for the $X(v = 0) \rightarrow A(v' = 0)$ transition and a radiative force induced deflection on the buffer-gas cooled BaF molecular beam. The laser induced fluorescence enhancement with additional sidebands and a polarization modulation scheme indicates that the hyperfine states and the Zeeman sublevels are closed. The quasi optical cycling by repumping the $X(v = 1) \rightarrow A(v' = 0)$ leads to a $\sim 0.8$ mm deflection of the beam via scattering $\sim 150$ photons per molecule, in good agreement with the predictions from our multi-level rate equation model. Further improvement by closing the leakage $X(v = 2)$ and $\Delta$ state allows scattering thousands of photons, and laser cooling and slowing of BaF.

I. INTRODUCTION

Laser cooling and trapping [1] using the light scattering force have led to lots of fundamental breakthroughs in atomic and quantum physics, especially the frequency standard for precision measurement [2] and the applications of the degenerate quantum gases [3, 4]. Over the last decade, great efforts have been spared into extending the techniques for control and cooling neutral atoms to polar molecules [5, 6] due to the additional vibrational, rotational degrees of freedom, which provide potential novel applications in many-body physics [7, 8], cold controlled chemistry [9, 10], and quantum simulation and computation [11–13]. While high phase space density has been achieved in closed-shell bi-alkali molecules by external association and adiabatic transferring techniques [14, 15], producing a degenerate open-shell molecular sample, such as alkali-alkaline-earth system, is still under exploration [16, 17]. Besides, another type of open-shell molecule, alkaline-earth-metal monohydride and monofluoride, first proposed by Di Rosa [18], can be directly laser-cooled [19], which has received quite great interests in recent years.

In fact, for molecules, it is difficult to find a perfect closed optical cycling channel to provide successive photon-molecule interactions required by laser-cooling because of the additional complexities. Fortunately, molecules like alkaline-earth-metal monohydride and monofluoride have special internal level structures, leading to nearly diagonal distribution of the Franck-Condon factors (FCFs), to close the vibrational branching. The cycling enhancement with additional sidebands and a polarization modulation scheme to generate four frequency branches required by laser cooling has already been demonstrated [20]. Recently, a rovibrational cooling of a supersonic BaF beam to a rotational temperature of $\sim 6K$ with broadband laser sources has also been reported [21], which provides another possible approach for preparing the cold molecular source.

In this paper, we experimentally demonstrate the quasi-cycling transition and further observe the light scattering force induced deflection on the buffer-gas cooled BaF molecular beam. We use $X^2 \Sigma_{1/2} \rightarrow A^2 \Pi_{1/2}$ electronic transition (with the linewidth of $\Gamma = 2\pi \times 2.84$ MHz [22]), which has required highly diagonalized FCFs, to close the vibrational branching. The cycling scheme has been described in detail in Ref. [23]. The $N = 1 \rightarrow J' = 1/2$ rotational transition is employed to eliminate the rovibrational branching, and a sideband modulation scheme to generate four frequency branches to cover the hyperfine levels. In current experiment, we have not taken the leakage channel from the $A^2 \Delta$ state into account yet. The contents are organized as following. Section [11] describes the experimental details. In Sec[11] we present the enhancement of the laser induced fluorescence (LIF) by introducing the $X(v = 1) \rightarrow A(v' = 0)$ repump laser, the sideband modulation and the polariza-
tion modulation of the light. Furthermore, we show the deflection of the molecular beam induced by the quasi-cycling photon scattering. The last section gives a brief conclusion and outlook.

II. EXPERIMENT

Figure 1(a) shows the diagram of the deflection experiment. We demonstrate the cycling scheme based on the buffer-gas cooled molecular beam of BaF produced with the laser ablation. Different from our previous study of the cold collisions between BaF and He [22], the He buffer gas here flows into the cell at a rate of 2 sccm (standard cubic centimeters per minute). The effectively thermalized (∼4K) mixture of He and BaF forms a beam via a 3 mm exit aperture of the cell. Another 3 mm aperture lying at 20 cm downstream from the cell filters out the molecules with higher transverse velocity, and collimate the beam. To deflect the molecules, we apply several laser beams along the $\hat{z}$ direction, perpendicular to the beam propagation. The molecule-light interaction time is controlled just by varying the pass number of the beams, and the maximum pass number can be tuned to 8 in our experiment. The pump (860 nm) and repump (896 nm) lasers, see Fig.1(b), are spatially overlapped with a diameter of $d = 2$ mm and powers of 160 mW and 100 mW respectively. To make all passes along the same direction, the laser beams are circularly reflected around the vacuum chamber [20]. The LIF from the $A(v' = 0) \rightarrow X(v = 0)$ transition is collected by an avalanche photo diode (APD), which focuses on the first laser beam in the 10 cm long interaction region. The deflection probe region locates $D = 35$ cm away from the interaction region, and between them a clean-up laser (896 nm) with a diameter of 8 mm and power of 50 mW hits the molecular beam to pump the molecules from the $X(v = 1)$ state back to the $X(v = 0)$ state. The BaF molecular beam profiles, including the width and position, are recorded by imaging LIF from a retroreflected laser beam (only 860 nm) on a CCD camera in probe region. The zoom ratio of the image system is 3 : 1. A band-pass filter of 860 ± 10 nm is used to decrease the background noise from the ablation laser and other stray lights.

To eliminate the hyperfine dark state, both the pump and repump lasers should cover all four hyperfine levels of the $X(N=1)$ states; see Fig1(c). Recalling the analysis in Ref [35], a resonant-type electro-optic modulator (EOM) with a modulation frequency of 38 MHz is employed in our experiment, and a modulation depth of 2.6 results in the first and second sidebands with equal amplitude, nearly matching the four hyperfine transitions in $X(N=1, -) \rightarrow A(J' = 1/2, +)$. On the other hand, the Zeeman dark state could be remixed by applying either an angled magnetic field or time-dependent polarization modulation [36]. Here we use a pockel cell to implement the polarization switching scheme and the modulation frequency is set as 1 MHz. Additionally, both the clean-up and the deflection probe beams are sideband modulated and polarization modulated as well.

III. RESULTS AND DISCUSSION

A. Quasi optical cycling

Figure 2 shows the time of flight (ToF) LIF signals from the main pump transition monitored by the APD with the toggle technique applied. From Fig2(a), by introducing the 38 MHz sideband modulation to the pump laser to address the hyperfine sublevels, the LIF signal is $\sim 2.5 \times$ enhanced in comparison with that when only one single-frequency pump laser resonant with the $F = 2$ sublevel applied. This can be easily understood since much more sublevels are excited by the additional sidebands, leading to more scattering photons before the molecules
FIG. 2. (Color online) LIF enhancement to demonstrate the quasi optical cycling. (a) Applying sidebands to the pump laser leads to the LIF enhancement by a factor of $\sim 2.5$. The red line indicates the APD signal when the frequency of the pump laser hits the $F = 2$ sublevel. (b) The addition of polarization switching to both pump and repump laser to remix Zeeman sublevels leads to $\sim 1.5 \times$ enhancement. (c) The addition of $X(v = 1) \rightarrow A(v' = 0)$ repump laser results in another $\sim 1.5 \times$ enhancement of the LIF signal, indicating the cycling of the vibrational levels. All the three group signals are normalized with the peak values of the lower signals respectively, and are averaged for hundreds of times to improve the signal-noise ratio.

FIG. 3. (Color online) The dependence of the LIF intensity on the laser frequency: (a) for 860 nm pump laser; (b) for 896 nm repump laser. The frequency scan was performed with a 38 MHz sideband modulation. For pump laser scan, no repump laser was introduced; while for repump scan, the pump laser was locked at the optimal point: +270 MHz. The solid lines are gaussian fits to the data points respectively, and we lock the lasers at the frequency for the peak: 11630.0848 cm$^{-1}$ for the 860 nm laser and 11164.3414 cm$^{-1}$ for the 896 nm laser.

populates the Zeeman dark states and $X(v = 1)$ state. On the other hand, the ToF signal tells us the time window of the detection. The peak LIF signal appears at $\sim 1.7$ ms, while the ablation laser fires at 0 ms and the distance between the cell and the interaction region is 35 cm, indicating the most probable velocity is $u_0 \approx 200$ m/s. This means that the time window of the APD is about $\tau = d/u_0 = 10$ $\mu$s.

The addition of the time-dependent 1 MHz polarization modulation to the pump laser increases the LIF signal by a factor of $\sim 1.5$; see Fig. 2(b). We find that the enhancement seems insensitive to the modulation frequency, and a 5 MHz modulation also leads to a similar result. However, our 4+13 multi-level rate equation (MLRE) model with the experimental parameters in Sec. II indicates about $3 \times$ enhancement of the scattering photon number per molecule within $\tau = 10 \mu s$ interaction time; see Appendix A for details. Due to the strong pump laser intensity (the saturation factor $s$ for each sideband is $\sim 300$), interaction time of 10 $\mu$s is enough to pump the molecule to dark $X(v = 1)$ state, and the model shows that each molecule scatters about 18 photons, which is close to the predicted value of $N_{00} \sim 1/(1 - q_{00}) \approx 20$ and $q_{00} = 0.9508$ [35] is the FCF for the $X(v = 0) \rightarrow A(v' = 0)$ transition. Consequently, the LIF enhancement with polarization switching indicates that about $18/1.5 = 12$ photons are scattered when no switching scheme applied. This might resort to the earth’s magnetic field which can also remix the Zeeman sublevels, since from the 4+13 model we expect only 6 photons are scattered before the molecule populates the Zeeman dark states or $X(v = 1)$ state without any remixing technique involved.

As shown in Fig. 2(c), the addition of the $X(v = 1) \rightarrow A(v' = 0)$ repump laser further makes the LIF signal $\sim 1.5 \times$ enhanced. This indicates that the scattering photon number within $\tau = 10 \mu s$ increases to $18 \times 1.5 = 27$, which is consistent with the predicted value from the 4+25 MLRE model with polarization switching scheme (see Fig. 6 in Appendix). Till now, the quasi optical cycling has been implemented by applying the 38 MHz sideband modulation, the 1 MHz polarization switching scheme and the $v = 1$ repump laser to close the hyperfine, the Zeeman and the first vibrational dark states respectively. The observed LIF enhancement agrees well with the predictions from our theoretical models.

Another important issue for the deflection experiment is the frequency of the pump and repump lasers. Because of the different excitation rates for each hyperfine sublevel in $X(N = 1)$, we scan the frequency within several hundreds of MHz to find an optimal position to lock
the frequency of the two lasers respectively. Figure 3 illustrates the dependence of the LIF signal intensity (the peak value of the ToF signal) on the laser frequency with the 38 MHz sideband modulation. For the pump laser, the fit tells us that the lock point should be +270 MHz, corresponding to 11630.0848 cm\(^{-1}\) (identical to the value resolved from the in-cell spectroscopy [32]); while for the repump laser, the best point is +30 MHz, corresponding to 11164.3414 cm\(^{-1}\). The clean-up laser and probe laser in Fig.1 are also locked at these two frequency points respectively [31].

B. Radiative deflection

The LIF enhancement for a single pass of the deflection beam in the interaction region indicates a significant radiative force on the molecules once the pass number \(n\) increases. Figure 4 shows the resolved molecular beam deflection along the \(\hat{z}\) direction monitored by the CCD camera for the pass number \(n = 8\). The shapes of the deflected and the unperturbed beams in the probe region are illustrated as Fig.4(a) and (b) respectively. An integration of the unperturbed image along the \(\hat{x}\) axis resolves the transverse width of the BaF molecular beam, about 3 cm, as shown in Fig.4(c). The addition of the deflection beam and clean-up beam leads to a \(\sim 0.8 \text{ mm}\) shift in the \(+\hat{z}\) direction while the beam width remains about 3 cm; see the normalized signals in Fig.4(d). We have also tested the effect of the \(X(v = 1) \rightarrow A(v' = 0)\) repump laser and clean-up laser, without which only \(\sim 10\%\) molecules remain in \(X(v = 0)\) state after suffering the \(X(v = 0) \rightarrow A(v' = 0)\) pump in the interaction region. Putting the repump and clean-up laser into the system again recovers the molecular signal to \(\sim 80\%\), which indicates effective optical pumping and repumping. The 20\% loss is due to the leakage \(X(v \geq 2)\) and the \(A^2\Delta\) channels [35].

Let us make an estimation of the scattering photon number \(N_{sc}\) from the deflection length \(l\). The time required for the molecular beam propagating from the interaction region to the probe region is \(\sim D/u_0\), then the transverse velocity changes by \(\delta u = u_0l/D\). The photon recoil momentum is given by \(p = h/\lambda\), where \(h\) is Planck constant and \(\lambda = 860 \text{ nm}\) is the wavelength of the main pump transition. The observed deflection length \(l \approx 0.8 \text{ mm}\) corresponds to a scattering photon number \(N_{sc} = m\delta u/p \approx 150\), here \(m\) is the mass of the BaF molecule.

We have also measured the dependence of the scattered photon number \(N_{sc}\) on the interaction time \(t = nt\), simply derived from deflection length \(l\) versus the pass number \(n\) of the deflection beam, as shown in Fig.5. Decrease of the pass number results in a linear decrease of the scattering photon number. The fit tells us the average scattering rate \(\Gamma_{sc} = 2 \text{ MHz}\), which is only a little different from the numerical result (also plotted in Fig.5) predicted by the 4+25 MLRE model with switch-
ing scheme. On the other hand, the theoretical maximum scattering rate for a multi-level 4+24 system is given as $\Gamma_{\text{max}} = \Gamma / 7 \approx 2.5$ MHz. The unsaturated average scattering rate in our experiment might result from the detunings of the sidebands for the hyperfine transitions.

IV. CONCLUSION

To summarize, we have clearly shown the evidence of the quasi optical cycling and further the radiative force from the $\sim 156$ scattering photons with only one additional $X(v = 1) \rightarrow A(v' = 0)$ repump. By applying the 38 MHz sideband modulation to the pump and repump lasers, the hyperfine dark states are eliminated. For Zeeman dark states, we have employed the 1 MHz polarization switching scheme to mix them to the cycling. Putting all these techniques together and increasing the pass number of the beam to achieve longer interaction time, we have observed a significant transverse deflection ($\sim 0.8$ mm) of the BaF molecular beam, indicating a scattering rate of $\sim 2$ MHz, which agrees well with the theoretical prediction from our MLRE model.

By adjusting the detunings of the pump and repump lasers, retroreflecting the both laser beams and providing sufficient interaction length, the molecular beam should be transversely cooled. Furthermore, the scattering photon number required for loading the beam to a trap is about $m_{u0}/p \approx 6.5 \times 10^4$ with a frequency-chirped or white light to longitudinal slow the beam, as a consequence, another transition, for example, $X(v = 0) \rightarrow B(v'' = 0)$, might be employed to improve the scattering rate. To build a magneto-optical trapping (MOT) of BaF, the addition of $X(v = 2) \rightarrow A(v' = 1)$ repump laser should be required due to the calculated larger branching ratio of $\sim 1.5 \times 10^{-3}$ for $A(v' = 0) \rightarrow X(v = 2)$ than those of CaF and SrF. Besides the RF-MOT with polarization switching, our previously proposed microwave mediated MOT ($\mu$-MOT) might be another candidate for our future laser cooling and trapping experiment.

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Appendix A: Multi-level rate equation model

The rate equations to describe the time evolution of the populated fraction in each sublevel for a multi-level system is given as

$$\frac{dN_l}{dt} = \Gamma \sum_u r_{l,u} N_u + \sum_{l,p} R_{l,u,p} (N_l - N_l),$$

$$\frac{dN_u}{dt} = -\Gamma N_u + \sum_{l,p} R_{l,u,p} (N_l - N_u),$$

where $N_l$ and $N_u$ are the populated fractions for the $l$-th sublevel in the ground state and the $u$-th sublevel in the excited state, $\Gamma$ is the spontaneous decay rate of the excited state, $r_{l,u}$ is the branching ratio for $u \rightarrow l$ transitions (see the values in Ref. [35]). $R_{l,u,p}$ is the excitation rate for $l \rightarrow u$ transition from the $p$-th laser beam, and $s_p$ is the saturation factor and $\Delta_p$ is the detuning. For the evaluation of $R_{l,u,p}$ with polarization switching scheme, we should take the selection rules into account, i.e., $R_{l,u,p} = 0$ for $m_u = m_l + 1$ when $\sigma_p = \sigma_-$ and $m_u = m_l - 1$ when $\sigma_p = \sigma_+$. The scattered photon number at time $t_0$ is evaluated from $N_{sc}(t_0) = \sum_u \int_{t_0}^t q_{00} \Gamma N_u dt$.

We firstly build a 4+13 model with linearly polarized laser applied, considering 4 excited states in $A(v' = 0, J' = 1/2, +)$ and 12 sublevels in $X(v = 0, N = 1)$. The 13rd level is the assumed loss channel with a branching ratio of $q_{loss} = 1 - q_{00} = 0.05$. For linearly polarized excitation, the $X(v = 0, N = 1, F = 2, m_F = \pm 2)$ sublevels are dark states. Our numerical calculation indicates that the molecule will loss to the 13rd level or populate the Zeeman dark states just after scattering $\sim 6$ photons; see Fig. 6. By introducing the 1 MHz polarization modu-

![FIG. 6. (Color online) Predicted scattering photon number as a function of the interaction time within 20 $\mu$s. The values are rapidly saturated for 4+13 model, even with polarization switching scheme to eliminate the Zeeman dark state. With both switching scheme and $X(v = 1)$ repump, the 4+25 model shows a linear relation between scattering photon number and interaction time, but the number saturates to $\sim 600$ with a rather longer interaction time (not plotted in the figure).]
luation, the model shows that scattered photon number increases to $\sim 20$, three times larger than that without switching, before the molecule entirely populates the dark states.

To close the loss channel, we add the $X(v = 1) \rightarrow A(v' = 0)$ repump laser to our model, i.e., $4+25$ model. Besides the 4 excited states and 12 sublevels for $X(v = 0, N = 1)$ and $X(v = 1, N = 1)$ respectively, the other loss channels, for example, $X(v \geq 2)$ and $A^{\text{se}}\Delta$ states, are all labeled as the 25-th level with a total branching ratio $q_{\text{loss}} = 1 - q_{\text{off}} - q_{\text{on}} = 1.6 \times 10^{-3}$. This model indicates that the molecule suffers from nearly successive photon scattering within 100 $\mu$s (larger than the interaction time in our deflection experiment), and finally the scattering process terminates after $\sim 600$ photons (close to the value of $1/q_{\text{loss}}$) are scattered for an interaction time of about 1 ms. Figure[3] shows the scattering photon number as a function of the interaction time within 20 $\mu$s. For 10 $\mu$s interaction time, the addition of the $X(v = 1)$ repump laser only increases the scattering number by a factor of $\sim 1.5$, which is consistent with our experimental observation. Finally, to achieve laser cooling of BaF, additions of $X(v = 2)$ repump laser and microwave remixing of $\Delta \rightarrow X(N = 0, 2)$ channels [35] are necessary to scatter thousands of photons.

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