Cold collision and high-resolution spectroscopy of buffer gas cooled BaF Molecule

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We reported a detailed experimental study of the cold collision of Barium monofluoride (BaF) with buffer gas and the high-resolution spectroscopy relevant with direct laser cooling. BaF molecules are efficiently produced with laser ablation and buffer-gas cooled in a cryogenic apparatus. The laser cooling relevant transition \( |X^2\Sigma, v = 0, N = 1\rangle \to |A^2\Pi, v' = 0, J' = 1/2\rangle \) is identified. The collision cross section with buffer gas is measured to be \( 1.4(7) \times 10^{-14} \text{cm}^{-2} \), which is very suitable for buffer gas cooling. Both rotational and vibrational temperatures are effectively cooled, and a large amount of molecules are quenched into the desired states. Our study provides an important benchmark for further laser cooling of BaF molecule.

I. INTRODUCTION

Cold molecules have plenty of important applications in fields ranging from quantum chemistry [1–4], quantum simulation [5–7], to precise measurement [8, 9], and so on. Over the last 20 years, the development of cold molecule physics has been pushed one after another, from traditional method by taming molecules with electric and magnetic field, such as Stark decelerator [10–12] and Zeeman decelerator [13, 14], to the laser cooling technique. Direct laser cooling, which is believed to be able to bridge the "μK" gap [15], is relatively a new direction and has achieved great progress in recent years. Lots of diatomic molecules have been laser cooled[16–21] or under explored [22–24], and even polyatomic ones have received significant attentions[25–27]. Among these molecules, BaF is a good candidate for laser cooling [28], the Franck-Condon factors are feasible for quasi-cycling transition. The cooling and repumping transitions are around 900nm, in the good regime of the diode laser, and the rotational constant is about 6.5 GHz [29], which can be easily dressed with microwaves.

Most of the molecules which can be laser cooled are highly reactive, and they are nonexistent in nature, thus must be produced in laboratory from gases or solid precursors. Molecules created in this way are usually very hot, the temperature is as high as \( \sim 1000 \text{ K} \). For laser cooling considerations, pre-cooling such hot molecules is of great importance to obtain a stable molecular source. Buffer-gas cooling is a very good method for such purpose, and has been developed as an effective process to produce a molecular beam [30]. For buffer gas cooling, both the elastic and inelastic collisions with cold buffer gas dissipate the energy of molecules, making all the degrees of freedom of molecules be cooled [26, 31–34]. Not only the translational velocity can be efficiently slowed down, but also the molecules are largely enriched in the lowest vibrational level and a few lower-lying rotational levels, making the science states used in laser cooling scheme largely populated.

Another concerned issue for further BaF laser cooling experiments is the identification of the cooling and repumping laser transitions. For the current in-cell stage experiment, direct measurement on the absorption spectroscopy is a simple and practical means to acquire the collision properties and the required transition information. Meanwhile, the temperature of the BaF molecule can be resolved to check the efficiency of the buffer gas cooling.

In this paper, we demonstrate the effective creation of BaF molecules from laser ablation, and buffer gas cooling with the 4 K helium gas. The cold collision cross section and the high-resolution spectroscopy relevant with laser cooling are measured. In Sec.II, we describe the experimental setup, including the 4 K cryogenic apparatus, the BaF \(_2\) target making and the absorption measurement. Section III gives the analyses on the absorption signal. We report the effective production of BaF molecules by laser ablation, and the measurement of the high-resolution spectroscopy helps to identify the main laser cooling transition. Furthermore, the collisional cross section between BaF and helium is measured and the efficient rotational and vibrational cooling are observed. In the final section, we conclude and summarize our results.

II. EXPERIMENT

The BaF molecules are created with laser ablation followed by buffer gas cooling. Figure 1(a) shows the schematic cryogenic apparatus, including a pulse tube refrigerator (Sumitomo, SRP-082B-F70H), which can achieve a temperature of 30 K at the first stage, and 4 K at the second stage. The vacuum chamber contains three layers of shielding. The outer one is an aluminum vacuum chamber, which provides an vacuum of better than \( 10^{-6} \text{ Pa} \). The vacuum is maintained by a 300 L/s turbo pump. The second layer is the 30 K shielding, which is attached to the first stage cryogenic head of the pulse tube refrigerator. The center one is the 4 K shielding, and attached to the second stage cryogenic head. In order to minimize the black-body radiation effect from the
FIG. 1. (Color online) Schematic experimental setup. (a) The cryogenic apparatus. The whole system is a aluminum vacuum chamber, the 30 K and 4 K shielding layers are attached to the first stage and the second stage coolers of a pulse tube refrigerator respectively. The buffer gas is pre-cooled by the 30 K and 4 K heat sinks before sending into the cell. Charcoal is used to help pump helium gas at 4K. (b) The cooling process from room temperate to 4 K. It takes roughly 8 hours. (c) The warming process from 4 K to room temperature.

Due to the naturally unstable property of BaF, we have to produce it artificially in the laboratory. Generally, two methods are used to create such diatomic molecules. One is laser ablation, and another is chemical reaction [20, 35]. The later method can produce molecules with higher flux, while the first one is much simpler to operate. We use laser ablation in our experiment. The ablation laser is a 532 nm pulse laser (Lapa-80, Beamtech Optronics), with a pulse duration 12 ns, the repetition rate can be tuned from 1−20Hz, and the highest output power is 50 mJ.

The laser ablation and absorption spectroscopy are performed in the science cell, as schematically illustrated in Fig. 2(a). The ablation 532 nm pulse laser is focused to a 1/e² diameter of 0.5 mm on the BaF₂ target by a f = 400 mm lens. An absorption spectroscopy scheme is setup to detect and monitor the molecule creation and collision dynamics. The probe beam passes through the cell and is detected by a photodetector (PD). We use a transfer cavity locked diode laser to ensure the frequency stability [36]. Once the 860 nm laser hits the molecular transitions, absorption happens and a dip appears at the time trace of the PD signal.

The laser ablation and absorption spectroscopy measurement. The BaF molecules are produced by laser ablation in the buffer gas cell. (b) The typical absorption signal of molecules with laser ablation. The pulse laser fires at t = 0 ms, the absorption signal shows up and then decays. (c) The normalized signal. The red line is the fitting function with formula (3). (d) The ablation laser power dependence of molecule production. The pulse laser is 532 nm, 2 Hz, 12 ns, 1/e² diameter is 0.5 mm. (e) The decay of laser ablation at one point. When the target are hit by laser more and more at one point, the produced molecule number decay. The decay constant is about few hundreds shots. All the data are measured with the He buffer gas rate of 5 sccm.
III. RESULTS AND DISCUSSIONS

A. Absorption signal from single shot

The typical absorption signal from single shot of the ablation laser is shown in Fig. 2(b). In order to extract the information behind the signal, we first normalized it to the absorption fraction

\[ c = 1 - \frac{I_{\text{abs}}}{\text{Max}(I_{\text{abs}})}, \]

as shown in Fig. 2(c). Then we construct a two-step model. Firstly, the molecules creation process is modeled by a logistic function

\[ y = \frac{A}{1 + e^{-(t-t_0)/\tau_1}}, \]

where \( t_0 \) means the delay after the pulse laser fired, and it depends on the distance between the target and the probe beam. \( \tau_1 \) evaluates how fast the creation process happens.

The second step is the decay process caused by the diffusion of molecules in the buffer gas cell, including the colliding with the cell walls and inelastic collisions with the buffer gas. Over all, we can model it with an exponential decay, so the two-step model gives a fitting function

\[ y = \frac{A}{1 + e^{-(t-t_0)/\tau_1}} \cdot e^{-(t-t_0)/\tau_2}. \]

Here \( \tau_2 \) describes how fast the signal decays. This two-step model fits quite well with the experimental data as shown in Fig. 2(c).

From the fitting function (3), we define the contrast, \( \sigma \), i.e., the maximum absorption fraction, to reflect the molecule number from laser ablation. Figure 2(d) shows the ablation laser power dependence of the molecule creation. The contrast starts to saturate around 40 mJ, and this point is set for the daily operation. Another issue is the decay of signal when we continually hit one point on the BaF\(_2\) target with the ablation laser. Figure 2(e) shows a typical data for such measurement. The lifetime for laser ablation at one position is about a few hundreds shots. In order to maintain good molecule creation condition, we need to change the fire point by adjusting the direction of the ablation laser from time to time.

B. Collisional cross section with buffer gas

As shown in Fig. 2(b), once the molecules are created by the ablation laser, it decays. The relaxations are mainly caused by the diffusion to the cell walls and the leakage via the aperture to form a molecular beam. Figure 3(a) plots the absorption fraction signal on logarithmic scale, where the helium flow rate is 5 sccm. Under this condition, the flow rate is relatively low, the diffusion to the cell walls dominates in molecule loss, and consequently the in-cell lifetime of the BaF molecule is proportional to the helium density in the cell [37]. Within the first 2 ms, the high-order diffusion mode rapidly decays. However, from 2 ms to 6 ms, the logarithmic signal can be fitted well with a line, which in fact reflects the in-cell colliding dynamics. The decay time constant \( \tau_d \) depends on the collision cross section with a simple formula [26],

\[ \tau_d = \frac{n_{\text{He}} \sigma_d}{\bar{v} G}, \]

where \( n_{\text{He}} \) is the helium density, \( \sigma_d \) is the collision cross section between molecule and the buffer gas, \( \bar{v} = (8k_B T/\pi \mu)^{1/2} \) is the average velocity between molecules and the helium gas at temperature \( T \), and \( \mu \) is the reduced mass of the BaF-He system.

For a cylindrical tube,

\[ G = \frac{3\pi}{32} \left( \frac{2.405^2}{r^2} + \frac{\pi^2}{L^2} \right), \]
where $r$ and $L$ are the radius and length of the cylindrical tube respectively. In our experiment, we control the flow rate of helium gas into the cell from 2 to 20 sccm. The helium density is determined by \[ n_{\text{He}} = \frac{\kappa f}{A_{\text{aperture}} v_b}, \] (6)

where $A_{\text{aperture}}$ is the area of the aperture and it is 78 mm$^2$ in our case, $v_b$ is the mean velocity of the buffer gas. At the full effusive limit (low helium flow rate regime), $\kappa = 2\sqrt{\pi}$, and at the supersonic regime, $\kappa$ could be a factor of 2 lower [33].

In Fig. 3(b), we plot the decay time constant $\tau_d$ versus the helium flow rate. By fitting it with a linear function at low flow rate regime, the collision cross section between BaF and helium is estimated from formula (6) at the effusive limit. It is about $1.4(7) \times 10^{-14}$ cm$^{-2}$. The error mainly comes from uncertainties of the determination on the helium density $n_{\text{He}}$ and the geometry factor $G$ of the cell.

We also measured the decay time constants for different rotational states ($|X^2\Sigma, v = 0\rangle$). The helium flow rate is fixed to be 5 sccm, and the probe laser is scanned to find the transitions for different rotational states. Then we picked the resonance frequencies and measured the absorption spectroscopies, and extract $\tau_d$. As shown in Fig. 4, $\tau_d$ decreases when the rotational number increases. It might be due to the collision cross section of BaF with helium is different for different rotational number at this temperature, as theoretic suggested for other molecules [39].

C. High-resolution spectroscopy

Laser cooling technique requires the lasers to be stabilized to 1 $\sim$ 2 MHz and the transition should be identified to be within a few MHz. High-resolution spectroscopy should be performed to find those transitions. Fortunately, the spectroscopy of BaF free radical has been studied for a long time, and the molecular constants have been determined very accurately [28, 29, 40–43]. Figure 5(a) shows the main cooling transitions from $A^2\Pi, v = 0, J' = 1/2, +$ to $|A^2\Pi, v' = 0, J' = 1/2, +\rangle$. The hyperfine structure of lower $N = 1$ level has splittings round 30 MHz and 120 MHz, while the hyperfine splitting of the exciting state $A^2\Pi$ is about a few MHz, and is hard to resolve.

In spectroscopy experiments, the transition $|N, J \rangle \rightarrow |N', J'\rangle$ is labeled by $\Delta N \Delta J(N)$, where $\Delta N = N' - N$, $\Delta J = J' - J$, $\Delta J(\text{or } \Delta N) = -1, 0, +1$ are corresponding to the $P, Q, R$ branches. Figure 5(b) shows the typical in-cell spectrum of the main cooling transition $|X^2\Sigma, v = 0, N = 1\rangle \rightarrow |A^2\Pi, v' = 0, J' = 1/2, +\rangle$. This transition involves the $P(1)$ and $R(1)$ branches, corresponding to the left and right peaks of the spectrum. The separation between the two peaks is 135(5) MHz, and it is consistent with the calculated energy difference between the $J = 1/2$ and $J = 3/2$ fine structure levels [28]. The full width half maximum is about 80(8) MHz, much bigger than the Doppler broadening (40 MHz) for BaF at $T = 4$ K, which might be caused by collision with the buffer gas. For the current-state experiment, we can not resolve $F = 0, 1$ and $F = 1, 2$ states (the hyperfine structure) with such large broadening.

We have also measured the $Q(0)$ transition from $|X^2\Sigma, v = 0, N = 0\rangle$ to $|A^2\Pi, v' = 0, J' = 1/2, -\rangle$, as shown in Fig. 5(c). The separation of the two peaks is
(Color online) The rotational relaxation for buffer gas cooling. (a) The theoretic calculation of the rotational distribution for different temperatures. Inset is the log scale plot. At room temperature, less than 1% molecules are in $N = 1$ state, while for 4K, it is enhanced to more than 10%. (b) The experimental data for different rotational populations. All data is normalized with $N = 0$ population. The lines are the theoretic curve for different temperature, and the best fitting shows the rotational temperature is $4.0(7)$ K.

77(5) MHz, which is consistent with the energy gap between $F = 0$ and $F = 1$ states for $|v = 0, N = 0\rangle$. The hyperfine structure of the $N=0$ rotational state is resolved. The absolute value of the wavelength in Fig.5 comes from a ultra-high resolution wavemeter (High-Finesse WS-7), with an uncertainty of 60 MHz, which is the limit of our measurement.

D. Rotational and vibrational cooling

Another concern is the rotational and vibrational cooling via the buffer gas collisions. Generally, the energy difference between different vibrational states is about a few 1000 K. Even at room temperature, only the ground vibrational state are largely populated, so it is not a big problem. However, for rotational states, the energy gaps are on the order of 1 K. At room temperature, molecules can populate to lots of different higher-lying rotational levels, as shown in Fig.6(a). For the laser cooling scheme [28], only a few low rotational states are involved. The small populated fractions lead to inefficient laser cooling.

According to the theoretic calculation of the rotational population at different temperatures in Fig.6(a), by cooling the molecules from room temperature down to 4 K, the populated fractions for the lower-lying rotational levels can be enhanced by two orders of magnitude. And indeed, experimentally, after buffer-gas cooling of BaF, the populations at low rotational states get largely enhanced, as shown in Fig.6(b). To measure the rotational fraction, we choose the transitions in the $P(N)$ branch, i.e., $|N, J\rangle \rightarrow |N' = N - 1, J' = J - 1\rangle$. The absorption strength reflects the population for the $N = 1 - 8$ rotational states respectively. For $N = 0$ state, we use the data from the $Q(0)$ measurement in Sec.III.C. We plot the measured the populations at different rotational lev-
els versus the rotational numbers, the best fits tell us the rotational temperature is $4.0 \pm 0.7$ K, which is consistent with the buffer gas temperature.

For the vibrational states, usually the buffer gas collision-induced quenching is far less efficient than that for the rotational states [44–46]. Theoretic calculation indicates (Fig. 7(a)) the population at $v = 1$ for $T = 4$ K is almost zero. But experimentally, we still observed the absorption signal from $v = 1$ to $v' = 0$. The signal is much weaker than the $v = 0 \rightarrow v' = 0$ transition, as shown in Fig. 7(b), and it takes long time to average down the noise. We estimated the population of $v = 1$ is about $10 - 100$ times smaller than that of $v = 0$ state.

IV. CONCLUSION

To conclude, we have created BaF molecules by laser ablation with a 532 nm pulse laser, and buffer-gas cooled them with the 4 K helium gas. The vibrational and rotational degree of freedom has been effectively quenched. Almost all the molecules populate the $v = 0$ level, and the rotational temperature is about 4 K. Large amount of molecules have been accumulated into the lower-lying rotational states, making further laser cooling experiments feasible. The collision cross section between BaF and helium gas is measured to be $1.4(7) \times 10^{-14}$ cm$^{-2}$, and it demonstrates that the BaF molecule is very suitable for buffer gas cooling. Meanwhile, the laser cooling relevant transition $|X^2\Sigma, v = 0, N = 1\rangle$ to $|A^2\Pi, v' = 0, J' = 1/2, +\rangle$ is observed with a high resolution. Our study paves the way for the laser cooling of BaF molecule.

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