Focusing of a cold PbO molecular beam with a superconducting microwave resonator

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Abstract
Focusing of a molecular beam in a high-field seeking state by an intense microwave standing wave is demonstrated. An 18 GHz microwave field that is near resonant to a rotational transition deflects a molecular beam due to the first-order ac Stark shift. The intense standing wave is obtained with a 47 cm long superconducting microwave resonator with a high unloaded quality factor of about 10⁶. A cold molecular beam of PbO in the rotational ground state with a center velocity of 145 ms⁻¹ is efficiently focused to have about a factor 10 enhancement of the flux density by the resonator with a microwave input power of about 1 W, despite the heaviness of the PbO molecule.

Keywords: cold molecule, focuser, superconducting resonator

(Some figures may appear in colour only in the online journal)

1. Introduction

Cold and ultracold molecules have attracted increasing attention in the fields of physics and chemistry. It is now well established that cold molecules are significantly useful to explore physics beyond the standard model via precision measurements [1], such as measurements of the electric dipole moment of the electron [2–5]. Cold buffer gas environments have offered novel molecular spectroscopy experiments [6–8]. Trapped cold molecules allow us to study cold collisions for well-defined states and with long interaction times [9–13]. Furthermore, chemical reactions can be highly controlled by external fields in the ultracold regime [14]. Recent progress of laser cooling [15–17] and optoelectrical cooling [18] of molecules has reached the ultracold regime. Many of these applications have been established on combinations of methods to control the translational motion of molecules, such as focusers, guides, decelerators, and traps [19].

Focusing is important to increase the molecular beam flux density. Hexapole (or quadrupole) electrodes have been conventionally used as the focuser, for example, to introduce molecular beams into a decelerator [20] and to select a state for a precision measurement [21]. They can focus molecular beams in low-field-seeking (LFS) states, which have lower energies at a smaller electric field. On the other hand, high-field-seeking (HFS) states are difficult to handle. For example, Earnshaw’s theorem claims that the HFS states cannot be trapped by a static field. The HFS states are of more importance than the LFS states, since the lowest energy state is always the HFS state for static fields. Even in rotationally excited states, polar molecules of small rotational constants are effectively HFS states, since they can only be LFS states for small electric fields.

So far, there have been several works to confine HFS state molecules 2-dimensionally. A thin wire with a high voltage can trap molecules in Kepler orbits [22]. Guides of HFS state molecules have been demonstrated by using alternate-gradient electric fields [23–25]. Decelerators...
The lowest-order transverse magnetic ground state is described in the present work, a cold and slow PbO beam in the rotational stage is used for confirming the molecular beam alignment.

[26–28] and traps [29] with this confinement have also been performed. Since the alternate-gradient method is dynamical, the effective confinement potential is not so deep.

A standing wave in a microwave resonator has also been used to focus a molecular beam in a HFS state [30–32]. These experiments have been performed with cavities made from copper which have quality factors \( Q \) on the order of \( 10^4 \). The electric field amplitude causing the ac Stark shift is proportional to \( \sqrt{PQ} \), where \( P \) is the input microwave power, and easily available amplifiers of \( P \approx 10 \) W cannot provide deep potentials. An intense microwave field is demanded not only for the focuser but also for decelerators [33–35] and traps [36, 37].

Here, we report the focusing of a molecular beam in a HFS state by using a superconducting microwave resonator. Our superconducting resonator has a much higher quality factor of \( Q \approx 10^6 \) than that of a normal conducting resonator. This fact reduces the requirement of \( P \) to \( \approx 1 \) W. In the previous paper [38], we have tested such a superconducting resonator and have confirmed that it tolerates input powers of several watts while maintaining a high quality factor. In the present work, a cold and slow PbO beam in the rotational ground state is deflected by an intense standing wave of the lowest-order transverse magnetic (TM_{010}) mode of a cylindrical cavity. The ac Stark potential is axially symmetric and nearly harmonic for the ground state of PbO with a potential depth of 0.11 K, which is much deeper than those of the other works of the dynamical confinement and the normal conducting resonators.

### 2. Focuser

The experimental setup is shown in figure 1. Our focuser is a superconducting microwave resonator of a Pb/Sn-coated cylindrical copper cavity. The structure is similar to the prototype resonator of our previous work [38]. The molecular beam of PbO passes through this cavity along the center axis. The cavity inner length is \( d = 470 \) mm and the inner radius is \( a = 6.3 \) mm. The inner surface is electroplated with an alloy of Pb (90%) and Sn (10%), which has a critical temperature of 7–8 K. About 2/3 of the resonator is immersed in liquid helium at 4.2 K, and the rest of the resonator is wrapped with superinsulation foil. The resonator has two end caps with center holes. The diameter and length of the holes are 4 mm and 8 mm for the molecular beam entrance and 5.5 mm and 8 mm for the exit, respectively. There are two additional holes of 2.4 mm diameter on the side wall of the main body to introduce loop antennas into the cavity. One antenna is used to couple the microwave into the resonator, while the other is used to monitor the resonator power. The antennas are made at the tip of semi-rigid cables of cupro-nickel, which has a low thermal conductivity. The insertion depth of the antenna for the input is optimized to obtain the strongest standing wave, while the insertion depth for the monitor is adjusted to couple the resonator very weakly. In other words, the input antenna is at the critical coupling condition, and the monitor antenna is highly undercoupled. The microwave is provided by a signal generator. When high power is required, the microwave is amplified by a traveling wave tube (TWT) amplifier. The maximum output power of the TWT is about 15 W, and the attenuation of the cables provides a loss of 7 dB.

The cavity supports the TM_{010} mode at a microwave frequency of \( \nu = 18.290 \) GHz. This frequency is slightly red-detuned from the \( J = 0 \) rotational transition frequency of PbO (\( \nu_{eg} = 18.37 \) GHz). Since the spontaneous emission rate in the microwave range is negligibly small, the microwave frequency can be tuned very close to the resonance. The TM_{010} mode has an electric field amplitude of

\[
E_z = E_0 j_0(x_0, \mu/\alpha a), \quad E_\rho = E_\phi = 0, \tag{1}
\]

where \( (\rho, \phi, z) \) are the cylindrical coordinates, \( E_0 \) is the maximum amplitude, \( J_n \) is the \( n \)th order Bessel function, and \( x_0 \approx 2.4048 \) is the first root of \( J_0 \). The TM_{010} mode is the simplest and ideal mode for focusing HFS states, since it can provide a cylindrically symmetric harmonic potential. The electric field amplitude of this mode is maximum on the center axis, and thus HFS state molecules experience a force towards the center axis. The ac Stark shift potential of \( U \) of the \( J = 0 \) ground state is given by \( U \approx -E \mu_{eg}/2 \) for small \( |\mu_{eg}| - \nu_f \), where \( E \) is the electric field amplitude and \( \mu_{eg} = 2.7(2) \) D [39] is the transition dipole moment of the \( J = 0 \) \( \rightarrow \) 1 transition. A molecular beam parallel to the cavity axis at the entrance of the cavity is focused at

\[
f = \frac{v_z}{\alpha \tan \frac{ad}{v_z}}, \tag{2}
\]

from the exit, where \( v_z \) is the axial velocity and \( \alpha = \sqrt{E_0 \mu_{eg} x_0^3 / 4a^2} m \) with \( m \) as the mass of the molecule (see appendix A).
The loaded quality factor \( Q_L \) is experimentally determined from the resonance linewidth \( \delta \nu \) as \( Q_L = \nu / \delta \nu \). It is also measured from the transient decay time of the stored energy \( W \) as \( W(t) = W(0) \exp(-2\pi t / Q_L) \), where \( t \) is the time after the microwave input is suddenly turned off. From the linewidth measurement shown in figure 2(a), \( Q_L = 5 \times 10^5 \) is obtained with a low input power of \( P = 3 \) mW. From the decay measurement after a pulse of a high input power of a few watts \( Q_L = 2.6 \times 10^5 \) is obtained, and it is almost independent of the pulse duration from 1 to 100 ms. These results indicate that the intrinsic unloaded quality factor, which is \( 2Q_L \) in our case, is about \( 10^6 \). The difference in \( Q_L \) between the two measurements may be attributed to the heating of the cavity for the high power input case. From equations (3) and (4) a parameter set of \( P = 1.9 \) W and \( Q_L = 2.6 \times 10^5 \) gives \( E_0 = 3.5 \) kV cm\(^{-1} \), which corresponds to the potential of \( U/k_B = 0.11 \) K with \( k_B \) as the Boltzmann constant. This deep potential enables us to successfully focus a heavy PbO molecular beam. The evaluation of \( P \), as well as the skew reflection spectrum in figure 2(a), is discussed in appendix B.

3. Experiment

The molecular beam is a cryogenic helium buffer-gas-cooled beam [40]. This beam source is similar to our previous works on the precise spectroscopy of atoms [41] and PbO [42]. A solid PbO target is ablated in a metallic cell at 4 K by a 532 nm pulse laser with a 10 Hz repetition rate. The laser fluence is around 10 mJ mm\(^{-2} \), and the pulse width is about 4 ns. The inner volume of the cell is about 30 cm\(^3 \), and the diameter of the cell exit aperture is 4 mm. The helium gas flow is continuous with a rate of 20 standard cubic centimeters per minute. The entrance and exit of the microwave cavity are at 64 cm and 111 cm from the cell aperture, respectively. The molecular beam is monitored via fluorescence at positions located 5, 45, and 135 cm (or 156 cm when a translation stage is incorporated) from the cell aperture. The excitation laser is tuned to the B(1)(\( \nu' = 5 \), \( J' = 1 \)) – X(0') (\( \nu'' = 0 \), \( J'' = 0 \)) transition of \(^{208}\)PbO at 406 nm [42] by locking to a temperature-stabilized ultralow expansion etalon [41]. The fluorescence is detected by photomultipliers with spectral filters.

The center of the cell aperture, the axis of the cavity, and the last detection points at 135 and 156 cm are all aligned on a straight line. This alignment is confirmed by a telescope and also by monitoring the spatial distribution of an indium atomic beam at 156 cm. The indium atomic beam is produced by laser ablation and buffer-gas cooling similarly to the PbO beam. The detection system is scanned 2-dimensionally by the translation stage to see the spatial distribution. The flux density is confirmed to be maximum on the cavity axis. This supports that the enhancement of the flux density of the PbO beam on the axis is due to the focuser, as shown below, and is not due to simple deflection, that is, the displacement of the maximal point of the flux density. The atomic beam spread at 156 cm is limited mainly by the entrance aperture of the microwave cavity, and is about 8 mm full width at half maximum.

There are two components in the cold PbO beam as shown in figure 2(b). A fast component is initially emitted, and a slow one trails it. A possible explanation for this is that...
the heat of the ablation increases the temperature and also causes desorption of helium gas from the PbO target, which is made by compressing a powder, and this sudden increase of the temperature and pressure generates the fast component. The slow component corresponds to the standard buffer-gas-cooled beam in the hydrodynamic regime, where the beam is moderately slow and has a high flux [40]. Figure 2(b) shows time profiles of the fluorescence at 5, 45, and 156 cm. These time profiles are well reproduced by assuming temporal and velocity distributions at the cell aperture as

\[ g(v_z, t) = \sum_{i=1,2} A_i \theta(t - T_i) \exp\left[-\frac{(v_z - u_i)^2}{2w_i^2}\right] - \frac{t}{\tau_i}, \]  

where \( \theta \) is the Heaviside step function and \( \{A_i, u_i, w_i, \tau_i, T_i\} \) are parameters to be fitted. The time origin \( t = 0 \) corresponds to the pulse laser irradiation. The center velocity and velocity distribution width \( (u_i, w_i) \) in unit of ms\(^{-1}\) are (370, 100) for the \( i = 1 \) fast component and (145, 25) for the \( i = 2 \) slow component, respectively. The decay times are \( \tau_1 = 0.025 \text{ ms} \) and \( \tau_2 = 0.37 \text{ ms} \). The beginnings of the beam are \( T_1 = 0 \text{ ms} \) and \( T_2 = 0.16 \text{ ms} \). The approximate distribution described by equation (5) is used for the simulation of the focusing to be described later. Note that there have been several works on the characterization of the helium buffer-gas-cooled beams. The slow component of our beam has a similar forward velocity \( u_2 \) to the previous works on heavy molecules [43, 44], and has a relatively short decay time \( \tau_2 \) close to that of the beam source optimized for generating a short pulse [45].

When the microwave is applied to the resonator, the molecular beam is focused and the flux density in the detection area, 135 cm from the source, is increased. The detection area is about 6 mm\(^2\), which is determined by the excitation laser beam diameter of 2 mm and the photon detection system. This area is much smaller than the molecular beam spread. The microwave pulse is applied for a duration of 1 to 16 ms, when most of the molecular beam fully experiences the microwave standing wave in the cavity.

4. Results

Figure 3(a) shows examples of time-of-flight profiles with and without the microwave. To minimize the effect of the gradual change of the yield of the laser ablation, the fluorescence with and without microwave is observed alternately, and the time-of-flight data with and without microwave are respectively accumulated. A photon count rate of \( 10^5\text{s}^{-1} \) in figure 3(a) corresponds to a flux density of about 1.7 \( \times \) 10\(^8\) cm\(^{-2}\) s\(^{-1}\) of the \(^{208}\text{PbO}\) beam for \( v_z = 145 \text{ ms}^{-1} \). Thus, the flux density of the slow component of the molecular beam within the detection area is increased from about \( 10^7 \) to \( 10^8 \text{ cm}^{-2}\text{s}^{-1} \) at the temporal peak by the focuser. The flux density is estimated from the photon collection and detection efficiency of about 2.4%, the excitation laser intensity of 0.01 W cm\(^{-2}\), and spectroscopic characteristics of PbO such as the lifetime of the B(1) state [46]. Simulated profiles are also shown in figure 3(a). The temporal and longitudinal velocity distribution of equation (5) is assumed with the amplitudes \( A_1 \) and \( A_2 \) reproducing the data for no microwave. All other parameter values are already obtained in section 3, and thus there are no fitting parameters to simulate the profiles with microwave. The initial position \( (r, \phi) \) on the cell aperture and the initial flight direction are assumed to have uniform distribution. Good agreement between the observation and simulation proves the focusing effect quantitatively.
The profile for $P = 0.6$ W has a longer tail than, for example, that for $P = 1.4$ W. This power dependence reflects the velocity dependence of the focal length of equation (2). Figure 3(b) shows simulated results of the enhancement of the flux density as functions of the axial velocity. The maximum enhancement factor is about 9, and it moves to larger $v_c$ with increasing $P$. This means that a high input power causes overfocusing to very slow molecules, which are responsible for the tail of the profile. Figure 3(c) shows the power dependence of the enhancement of the flux density integrated for $7 \leq t \leq 14$ ms, which almost covers the slow component profile. Experimental data shows that the number of molecules passing the detection area increases by a factor of about 5 for the slow component for $0.5 < P < 2$ W.

5. Conclusion

In conclusion, a proof-of-principle experiment of an efficient focuser for a polar molecular beam in a HFS state is performed by using an intense standing wave of the TM$_{010}$ mode in a cylindrical superconducting microwave cavity. The deep confinement potential is able to efficiently focus the PbO molecular beam despite its heaviness of 224 u. Only about 1 W microwave power is enough for our experiment. This power can easily be handled with usual cryogenic systems. Since the focal length of equation (2) scales with $v_c/\alpha \propto v_c \sqrt{m}$, a conventional supersonic expansion beam with a heavy carrier gas of about 300 ms$^{-1}$ would be able to be similarly focused for moderately light molecules of about 50u. Since the microwave resonator has a more closed structure than a hexapole focuser, more careful skimming of the beam would be required to bring it very close to the beam source. However, the cryogenic surface of the resonator would be beneficial for adsorption pumping of the heavy carrier gas.

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Appendix A. Details of the theory of the focuser

The $J = 0$ ground state is adiabatically connected to the HFS dressed ground state in the case of $\nu < \nu_{eg}$. The ac Stark shift potential $U$ of this state for the electric field amplitude $E$ is well approximated as

$$U = \frac{h(\nu_{eg} - \nu)}{2} - \left[ \frac{h(\nu_{eg} - \nu)}{2} \right]^2 + \left( \frac{E\mu_{eg}}{2} \right)^2$$  \hspace{1cm} (A.1)

where $h$ is the Planck constant. When $h|\nu_{eg} - \nu| \ll |E\mu_{eg}|$, the first-order Stark shift $U \simeq -E\mu_{eg}/2$ is obtained. This first-order Stark shift and the approximation of $J_0(x) \simeq 1 - x^2/4$ for equation (1) give the cylindrically symmetric harmonic potential.

When the azimuthal velocity component $v_\phi$ of the molecular beam is neglected, the radial position and velocity $(\rho_{in}, v_{in})$ of a molecule at the entrance of the cavity and those at the exit $(\rho_{out}, v_{out})$ are related for this harmonic potential as [38]

$$\left( \begin{array}{c} \rho_{out} \\ v_{out} \end{array} \right) = \left( \begin{array}{cc} \cos \alpha & \frac{1}{\alpha} \sin \alpha \\ -\alpha \sin \alpha & \cos \alpha \end{array} \right) \left( \begin{array}{c} \rho_{in} \\ v_{in} \end{array} \right)$$  \hspace{1cm} (A.2)

Molecules with $v_{in} = 0$ are focused at the distance $f$ of equation (2) from the exit of the cavity.

Appendix B. Evaluation of the input microwave power

The evaluation of the input microwave power $P$ is not straightforward in our experiment. Since our vacuum feed-through for the microwave has a non-negligible reflectance, the semi-rigid cable between the feedthrough and the antenna becomes a lossy low-$Q$ resonator. The combination of this low-$Q$ resonator and the high-$Q$ superconducting resonator causes a distortion of the reflection spectral profile and a deviation of the reflection minimum as compared to the transmission maximum seen by the monitor antenna (see, figure 2(a)). These behaviors are well explained by a model of two combined optical cavities composed of three mirrors as shown in figure B1. The electric fields $E_i$ are related one another as

$$E_2 = t_1 \alpha_1 E_4 \exp(ikL_1) + r_1 E_1,$$
$$E_3 = t_1 E_1 - r_1 \alpha_1 E_4 \exp(ikL_1),$$
$$E_4 = t_2 \alpha_2 E_6 \exp(ikL_2) + r_2 \alpha_1 E_3 \exp(ikL_1),$$
$$E_5 = t_2 \alpha_1 E_3 \exp(ikL_1) - r_2 \alpha_2 E_6 \exp(ikL_2),$$
$$E_6 = -r_3 \alpha_2 E_5 \exp(ikL_2),$$
$$E_7 = t_3 \alpha_2 E_5 \exp(ikL_2),$$  \hspace{1cm} (B.1)

where $r_i$ is the amplitude reflection coefficient, $t_i$ is the
amplitude transmission coefficient with $r_i^2 + t_i^2 = 1$, $L_i$ is the spacing between mirrors, $k$ is the wavenumber, and $\alpha_i$ represents the loss. The reflection and transmission of the power are proportional to $|E_{2i}|^2$ and $|E_{1i}|^2$, respectively. The fitted curves (dots in figure 2(a)) of the solutions of equation (B.1) reproduce well the experimental data (lines in figure 2(a)). The power $P$ entering the resonator is also determined from this model. The energy $W$ stored in the superconducting resonator is proportional to $|E_d|^2 + |E_a|^2$.

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