Control of translational motion of polar molecules by using superconducting microwave resonators

Katsunari Enomoto,1,2 Yoshiki Moriwaki,1 Walter Hardy,3 Omid Nourbaksh,3 Pavle Djuricic,2 and Takamasa Momose2,3

1Department of Physics, University of Toyama, 3190 Gofuku, Toyama 930-8555, Japan
2Department of Chemistry, The University of British Columbia, Vancouver, BC V6T 1Z1, Canada
3Department of Physics and Astronomy, The University of British Columbia, Vancouver, BC V6T 1Z1, Canada

(Dated: October 8, 2010)

We propose experimental methods to control translational motion of polar molecular beams by using a microwave field enhanced in a superconducting resonator. By using a strong ac electric field in the superconducting resonator, polar molecular beams can be efficiently focused, decelerated, and trapped by a deep ac Stark shift potential. We have made a superconducting resonator for this purpose, and the quality factor is at least \(3 \times 10^5\). This control method can be applied to produce a cold, slow, and high-flux molecular beams, which is favorable for the measurement of the permanent electric dipole moment (EDM) of an electron.

PACS numbers:

I. INTRODUCTION

Ultracold molecular gases have attracted keen interest in this decade [1]. There are several promising research fields of the application of the ultracold molecules. Polar molecules have a strong, long-range, and anisotropic intermolecular interaction due to their electric dipole moment. If such polar molecular gases are cooled to the quantum degeneracy regime, they will show various quantum phases such as supersolid [2]. Ultracold chemical reactions and the controllability of ultracold collisions with external fields have been extensively investigated [1]. Accurate measurements of energy levels using cold and slow molecular beams or trapped molecular gases are inexpensive and promising researches to explore new high-energy physics beyond the standard model such as time variation of fundamental constants and EDMs of an electron and nuclei [3].

There are two main routes to generate ultracold molecules. One method is association of ultracold atoms. Photoassociation and Feshbach resonances are used to convert an atom pair to a molecule. Recently a combination of the Feshbach resonance association and stimulated Raman adiabatic passage coherently transformed ultracold atom pairs to molecules in the rovibrational ground state [4]. Another method is to directly cool molecules and/or to filter low-velocity molecules. This method is applicable to many kinds of molecules including polyatomic molecules, though the lowest temperature achieved so far is in a milliKelvin range. Molecules can be cooled to about 1 K by collisions with carrier gases in supersonic expansion jets or cryogenic helium buffer gases. Static electric and magnetic fields are mainly used to decelerate molecular beam packets [5] and to perform velocity-filtering for the molecular beams [5]. It should be noted that laser cooling of SF\(_6\) molecules has recently been demonstrated [7].

The static fields are powerful tools to control molecular motion, and actually have been used in a long history. However, they have a disadvantage that the field maximum cannot exist in vacuum (Earnshaw’s theorem). This means that high-field seeking (HFS) states cannot be trapped by the stationary fields. Note that the lowest ground state, which is important for various applications, is always the HFS state. A dynamical confinement method with a quasi-static field similarly to an ion trap is instead available to trap the HFS states [9]. DeMille et al. proposed to use a strong microwave standing wave to generate a deep trap potential for molecules [9]. In this case, the microwave frequency is tuned close to a rotational transition, and a strong ac Stark shift can be obtained. Such standing waves can have a field maximum, which can trap HFS state molecules. A time-varying standing wave of a microwave field can decelerate molecules [10]. An advantage of this deceleration method is that the radial confinement is not dynamical. HFS state molecules have already decelerated with a dynamical confinement method for transverse motion, which is called alternate gradient focusing deceleration [11]. This method has to realize the longitudinal deceleration and the transverse dynamical confinement simultaneously, and it is difficult to fulfill both requirements from an initial finite velocity to the final zero velocity. The first step for the control of molecular translational motion with a microwave field is deflection and focusing of molecular beams. The first experiment of deflection dates back to 1975 [12], and recently an obvious focusing of molecular beams with a well-defined velocity and internal state has been demonstrated [13].

In this paper we suggest to use superconducting resonators to improve these microwave tools. The molecular motion is controlled by a potential of an ac Stark shift caused by the microwave electric field \(E \propto \sqrt{PQ_0}\), where \(P\) is the input microwave power and \(Q_0\) is the unloaded quality factor of the resonator. To increase the electric field \(E\), one possibility is to increase \(P\). However, increasing the power \(P\) requires expensive microwave am-
plifiers and other components. Furthermore, a special care of cooling of the resonator is needed to compensate the heat caused by the large input power. The quality factor $Q_0$ is determined by the surface electric conductance, and it can be extremely large if the resonator is made of superconducting material. Thus, a strong electric field can be obtained for that resonator with a not so high $P$. Such superconducting microwave resonators are utilized for charged-particle accelerators and cavity quantum electrodynamics experiments [14].

II. THEORETICAL CALCULATION

Here we will consider a simple example of the interaction between a polar molecule and a microwave electric field. If the microwave frequency $\nu$ is close to the resonance frequency $\nu_{ge}$ of the transition between the upper rotational state $|e\rangle$ and the lower one $|g\rangle$ and the other levels can be neglected, the interaction is described with the rotating wave approximation in the semi-classical treatment as

$$H = \hbar(\nu - \nu_{ge})|e\rangle\langle e| + \frac{\mu_{ee} E_0}{2}|g\rangle\langle e| + |e\rangle\langle g||,$$  
(1)

where $\hbar$ is the Planck constant and $\mu_{ge}$ is the transition dipole moment. If the detuning $\nu - \nu_{ge}$ is negative (positive), the eigenstate correlated with $|g\rangle$ is the HFS (low-field-seeking) state, respectively. For a more general treatment, see Ref. [9]. A standing wave of the microwave generates a series of potential troughs in a resonator. If the standing wave is turned on and off repeatedly in accordance with the timing of the flight of a molecular packet in the resonator, it is possible to make the molecules experience only the upward slopes of the potential so that they lose the kinetic energy. The molecules can be introduced in the resonator through a hole whose size is much smaller than the wavelength of the microwave.

Although our original proposal [10] of the deceleration method is to use a Fabry-Perot type resonator, our current plan is to use a cylindrical waveguide resonator similarly to the focusing experiment [13]. In a circular waveguide the lowest transverse mode is the TE$_{11}$ mode, and it has the following electric field pattern:

$$E_r = 2E_0 \frac{J_1(x)}{x} \sin \theta, \quad E_\theta = 2E_0 J'_1(x) \cos \theta,$$  
(2)

where $x$ is proportional to the distance from the center axis and becomes 1.841 at the waveguide wall, $\theta$ is azimuth, $E_0$ is the electric field at $x = 0$, $J_1(x)$ and $J'_1(x)$ are respectively the first-order Bessel function and its derivative, and $E_r$ and $E_\theta$ are respectively radial and azimuthal components of the electric field. The TE$_{11}$ mode has a maximum electric field on the center axis, and thus provides a radial confinement force for HFS states. For a resonator with the TE$_{11p}$ mode with $p$ the longitudinal mode number, the maximum electric field $E_{\text{max}}$ which is obtained on the center axis at antinodes, is related with $P$ and $Q_0$ as [13]

$$PQ_0 \approx 0.75e_0 \rho V E_{\text{max}}^2,$$  
(3)

where $e_0$ is permittivity of vacuum and $V$ is the inner volume of the cylindrical cavity. If the resonator has $V \sim 10 - 100 \text{cm}^3$ and is made of copper, the quality factor is typically $Q_0 \sim 10^9 - 10^{10}$ for $\nu \sim 10 - 20 \text{ GHz}$ at a room temperature. For superconducting resonators, $Q_0 \sim 10^{10}$ or more is possible [15]. For our deceleration purpose, however, a too high quality factor is not useful since we have to switch on/off the standing wave of microwave rapidly. The criterion is $Q_0 \ll \epsilon/c$ with $c$ the speed of light and $\nu$ the velocity of molecules [10], and thus resonators with $Q_0 \sim 10^9 - 10^{10}$ are practical.
Considering availability of microwave amplifiers and scales of the wavelength and the resonator, we think 10 – 30 GHz is a preferable frequency range. Molecules with large $\mu_g$ are also favorable. We are particularly interested in YbF, BaF, and PbO for future electron EDM measurements, and CH$_3$CN as an example of polyatomic molecules. We have carried out a simulation of deceleration using parameters of the rotational ground state of PbO ($\mu_g = 2.68$ Debye, $\nu_g = 18.37$ GHz, and the mass of 224.0 a.m.u.) and using Eq. (1). We have assumed $PQ_b = 10^6$ W with a resonator size of 14 mm inner diameter and 1 m long. The corresponding electric field is $E_{\text{max}} = 7.3$ kV/cm. Figure 1 shows an example of the simulation. A portion of molecules with the initial kinetic energy of about 9 K are completely decelerated after 70 ms by this microwave decelerator. Note that cold molecular beams with the kinetic energy of a few Kelvin have already been realized by a buffer-gas cooling method [16] and a counter-rotating nozzle method [17, 18]. Therefore, combination of such pre-deceleration methods and this microwave decelerator will enable us to completely decelerate and to trap HFS state molecules.

III. TEST OF RESONATOR

We have made a superconducting resonator and measured the quality factor. The resonator is made of copper, and the inner surface is electro-plated with lead (Pb) and tin (Sn). The superconducting transition temperature of Pb is 7.2 K, and Sn is added for preventing from oxidation. The inner diameter is 12.7 mm, and the length is 112.7 mm. The resonator is composed of two parts and they mate at the middle of the resonator, where an antinode is located for odd $p$ modes, so that the current does not go across the connection part for those modes. 3 mm diameter holes are perforated on the center axis on both ends. A loop antenna made from stainless coaxial cable is inserted into one of the holes to couple to the resonator. By adjusting the insertion depth of the loop antenna, we can optimize the coupling strength to the resonator. The resonator is attached to a liquid helium bath, and is cooled to about 4 K. Microwave whose frequency is between 13 and 20 GHz is introduced through the loop antenna, and it excites TE$_{11p}$ mode standing waves. If the resonator is resonant to the microwave, the reflection from the resonator decreases. We have measured the reflection of the microwave from the resonator with scanning the frequency, and determined the quality factor from the linewidth of the resonance. The experimental setup is schematically shown in Fig. 2(a).

The loaded quality factor $Q_L$ is given by $\nu/\delta\nu$, where $\delta\nu$ is the full-width-at-half-maximum of the resonance signal of the reflection. $Q_b$ is equal to 2$Q_L$ if the impedance matching condition is fulfilled. Firstly we have tested the copper resonator before performing the Pb/Sn coating, and obtained $Q_L \sim 5000$ at a room temperature and $Q_L \sim 16000$ at the liquid nitrogen temperature. Next, after performing the Pb/Sn coating, we have carried out the same measurement. The quality factor is much smaller at a room temperature than the copper resonator, but it drastically increases below the transition temperature. The resonance signal of the resonator at about 4 K is shown in Fig. 2(b). This result indicates $Q_L \sim 3 \times 10^6$ for this superconducting resonator. The limit of the acceptable input power is remaining to be measured. Note that this is a preliminary, first trial to gain experience with the geometry etc. Much higher quality factors are routinely available, and there are many possible ways to improve our resonator, such as substituting with a niobium resonator.

IV. FUTURE APPLICATION

As described in the introductory section, a microwave field is a promising tool to handle HFS state molecules. We are planning to apply it for an electron EDM measurement experiment based on cold molecular beams. An electron can possess a very tiny EDM due to CP and T-violating interactions. According to theories beyond the standard model, such as supersymmetric models, the electron EDM may be too small to detect [19]. The current upper limit of the size of the electron EDM is $\sim 10^{-27}$ e cm determined by a Ti atomic beam experiment [20], and actually some theories have predicted larger electron EDMs than this value. The electron EDM measurement is carried out by measuring a
tiny difference of the electron spin precession frequency of heavy atoms with applying a strong electric field. It is known that the sensitivity of the EDM measurement can be greatly improved if the atoms are substituted with polar molecules, since they have a much stronger anisotropic electric field inside. The electron EDM measurement using YbF molecular beams has been demonstrated by Hinds and co-workers [21]. The upper limit given by this experiment is $\sim 10^{-25}$ e cm, which is two orders of magnitude behind the atom-based experiment.

This is mainly because the flux of the molecular beams in the required rovibrational ground state is much smaller than the atomic beam flux. Therefore, if one can prepare cold molecular beams in which most molecules are in the ground state, the sensitivity will be much improved. Furthermore, if such molecular beams are slow, the precession time for a given flight length is extended and the sensitivity is also improved. Currently several groups are proceeding with the electron EDM measurement using cold and slow molecular beams.

We are constructing a cold molecular beam source based on helium buffer-gas cooling, similarly to Refs. [16, 22]. A copper metal cell is attached to a liquid helium bath, and a helium gas flow of about 10 secm is supplied to the cell. The cell has a hole of about 1 mm diameter to extract molecules produced in the cell along with the helium gas. A solid target of PbO is placed in the cell, and it is ablated with a pulsed Nd:YAG laser (532 nm) to produce PbO molecules. We have detected Pb and O atoms in the gas flow with a residual gas analyzer. We are now making an optical detection system for PbO molecules. We will also try to introduce YbF$^+$ molecules into the buffer-gas cooling cell from an oven inside.

If the microwave lens for molecular beams is incorporated with the beam source, the directionality will be improved and thus the effective beam flux will be enhanced. If the microwave decelerator is connected, a cold and slow molecular packet can be obtained. Such applications will improve the sensitivity of the electron EDM measurement. Moreover, a microwave electric field in a superconducting resonator can be enough strong to focus cold but fast molecular beams produced by a supersonic expansion. We are now constructing an experimental setup to demonstrate the focusing of supersonic jet of CH$_3$CN molecules.

In conclusion, we have proposed the use of superconducting microwave resonators to control translational motion of polar molecules in HFS states. We have shown a simulation result of deceleration of molecules with a $\text{TE}_{11}$ mode in a cylindrical resonator. We have also examined a Pb/Sn coated superconducting resonator, and obtained $Q_L \sim 3 \times 10^5$. Such a superconducting microwave tool is compatible with a cryogenic helium buffer-gas cooling setup. It can improve the electron EDM measurement experiment based on cold and slow molecular beams produced from the buffer-gas cooling system.

Acknowledgments

We acknowledge M. Schnell, S. Merz, H. Odashima, G. Meijer, K. Kobayashi, F. Matsushima, S. Tsunekawa, Y. Takahashi, and D. DeMille for their courtesies and helpful advices. We also thank Y. Kwatada, H. Noguchi, H. Hasegawa, and N. Suzuki for experimental assistances of the buffer-gas cooling system. This work was partially supported by Grant-in-Aid for Scientific Research of JSPS (19840021, 21740300, 22104504), Matsuo foundation, Inamori foundation, and Excellent Young Researchers Overseas Visit Program of JSPS.