A new pathway to the dipolar molecule ground state from Bose polarons

fermi1

We report creation of dipolar molecules at their absolute ground state starting directly from strongly interacting Bose polarons. Bose polarons are quasi-particles formed by immersing impurities inside of a Bose-Einstein condensate (BEC). In a dense bosonic bath, polarons have 50 times longer lifetime than weakly-bound Feshbach molecules, which makes the polaron a stable initial state. We demonstrate the contact, a universal many-body quantity, is proven to be a crucial parameter for binding an atom pair into a ground state molecule. Sufficient polaron contact is observed via photo-association spectroscopy. We perform dark resonance spectroscopy to measure polaron coupling to the absolute ground state via the electronically excited state. A two-photon stimulated rapid adiabatic passage directly creates ground state dipolar molecules inside a T/T_c=0.1 BEC.

Quantum gases of ultracold dipolar molecules are prime candidates for realizing novel many-body systems. Dipolar molecules possess large electric dipole moments and rich internal structures like vibrational, rotational, and nuclear spin degrees of freedom. The long-range nature of electric dipole moments between molecules can create new types of order that are beyond simple contact interactions between atoms. When a dipolar molecule is immersed inside of a Bose-Einstein condensate (BEC), molecule's rotational degree of freedom can couple to the bosonic bath and forms a new quasi-particle called an "angulon", which is still in its experimental infancy [1–3]. Weakly-bound molecules associated via Feshbach resonances are considered a prerequisite step in creation of ground state molecules, because Feshbach molecules provide strong coupling to the excited state. However, the association process yields a finite efficiency and Feshbach molecules are short-lived in a dense bosonic bath due to rapid three-body recombinations, which from the very beginning pose an obstacle for creating ground state molecules in a BEC. This report experimentally realizes a new pathway to the dipolar molecule ground state, which never involves a weakly-bound molecular state as a starting point. The new pathway begins with an attractively interacting Bose-Fermi mixture, directly followed by a two-photon rapid adiabatic passage (STIRAP) that coherently transfers the mixture into the absolute rovibrational ground state.

With fermions at the impurity limit, the Bose-Fermi mixture can be described by a framework of Bose polarons. Bose polarons are quasi-particles formed by impurities dressed by a bosonic bath. Within variational descriptions, the Bose polaron wavefunction has a localized component that has the same characteristics as a Feshbach molecule whose molecule size is set by the polaron energy [4–6]. Polaron energy is related to contact, a many-body quantity that characterizes short-range correlations and captures the change of polaron energy with interaction strength. An intuitive interpretation of the contact is the probability that a fermion finds a boson at a certain proximity [7], which is the very essence for binding two atoms into a molecule. Fig. 1 shows the polaron (blue) energy and contact smoothly evolving from attractive to repulsive interactions, where polarons merge with Feshbach molecules (black). Feshbach molecules exist only at the repulsive side of the resonance, where they are short-lived in the presence of a dense boson bath. In

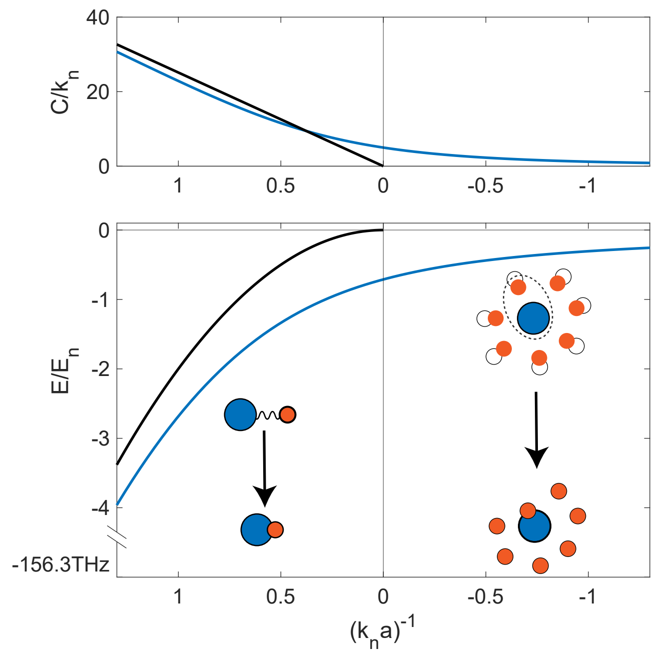

FIG. 1. Pathways to the absolute rovibrational ground state molecules. Polaron contact and energy based on variational descriptions are illustrated in blue line together with Feshbach molecule in black line. The Feshbach molecule branch starts near the Feshbach resonance and extends to the repulsive side. A polaron can be pictured as a Feshbach molecule dressed by a condensate, whose energy smoothly evolves from negative to positive interaction strength and eventually connects to the molecule branch. A polaron, with near unitarity-limited interaction, has a sizable contact that enables couplings to the excited state and a 50 times longer lifetime in a boson bath than a Feshbach molecule. A typical pathway to the ground state molecule from a Feshbach molecule is illustrated on the left. On the right, a new pathway is demonstrated starting from a fermion (blue circle) dressed by bosons (orange circles) to a 156.3 THz deeply bond ground state molecule.

contrast, polarons with attractive interactions have a finite contact and almost two orders of magnitude slower loss rate with bosons, even near unitarity. These two properties of the polaron open up a unique gateway to make ground state molecules in the presence of a BEC.

Similar to our previous work described in [6], we start with fermionic ^{40}K and bosonic ^{23}Na at their ground hyperfine states: $^{40}\mathrm{K}|\mathrm{F}, \mathrm{m}_F\rangle = |\frac{9}{2}, -\frac{9}{2}\rangle$ $\mathrm{Na}|\mathrm{F}, \mathrm{m}_F\rangle = |1, 1\rangle$. After the evaporative cooling in a crossed optical dipole trap, ^{23}Na atoms form a BEC with peak density $n_B = 7 \times 10^{13}$ cm^{-3}, temperature T=100 nK and trapping frequencies $(\omega_x, \omega_y, \omega_z)/2\pi = (120, 116, 10)$ Hz. A magnetic field gradient is applied to levitate both species, which cancels the gravitational sag due to ^{23}Na ^{40}K mass difference. Fermions are fully immersed in the BEC at 81 G where three-body losses between ^{23}Na ^{40}K are minimal. Fermions are at the impurity limit with peak density $n_F = 2 \times 10^{11}$ cm^{-3}. The magnetic field is then ramped to its final value at 78.54 G, where the inter-species scattering length $a = -4000\ a_{\mathrm{Bohr}}$. Impurities are strongly attracted to the BEC with interaction strength $(k_n a)^{-1} = -0.3$, where $k_n = (6\pi^2 n_B)^{1/3}$ is the inter-boson distance. Following a 2 ms thermalization time, impurities are strongly dressed by the BEC and form Bose polaron quasi-particles with an estimated energy $E_p/E_n = -0.5$. E_p is the quasi-particle self-energy and $E_n = \frac{\hbar^2 k_n^2}{4 m_r}$ is the boson degeneracy energy ($m_r = \frac{m_B m_F}{m_B + m_F}$ is the reduced mass). Fig. 1 demonstrates that Bose polarons with attractive interactions have a finite contact. We observe that the contact enhances the impurity coupling to the excited state with photo-association spectroscopy.

An up-leg laser (ω_1) in horizontal polarization is applied to perform the photo-association (PA) spectroscopy shown in Fig. 2. When ω_1 resonates with the initial polaron state $|\mathrm{P}\rangle$ and the excited state $|\mathrm{E}\rangle$, the quasi-particle wavefunction collapses as each dressed impurity finds a nearby boson and photo-associates to an electronically excited molecule. We observe a PA resonance frequency red-shift that is linear to the up-leg laser intensity. Due to the polaron's small energy separation from the free-particle continuum, the continuum dresses the excited state and shifts its energy. Similar AC-Stark-like effects were studied in [8–12]. The continuum- and light-induced spectral shift is measured to be $-(2\pi)2.9$ kHz/(W cm^{-2}). The maximum polaron to excited state coupling is achieved with up-leg laser intensity at 7×10^3 W/cm^2 and at frequency -(2π)19 MHz from the unperturbed resonance. As shown in Fig. 2b, strongly dressed impurities within the BEC Thomas-Fermi radius (black dots) are averaged to have a $\Omega_1 = (2\pi)0.6$ MHz Rabi rate with a 5 μs exponential decay constant. The impurities at the trap center are estimated to have a higher than average Rabi rate since they are dressed by denser bosons. In contrast, weakly dressed impurities outside of the Thomas-Fermi radius (gray dots) are unperturbed by the up-leg laser. These two distinctive photo-association rates manifest the paramount importance of the contact in binding two atoms into a molecule.

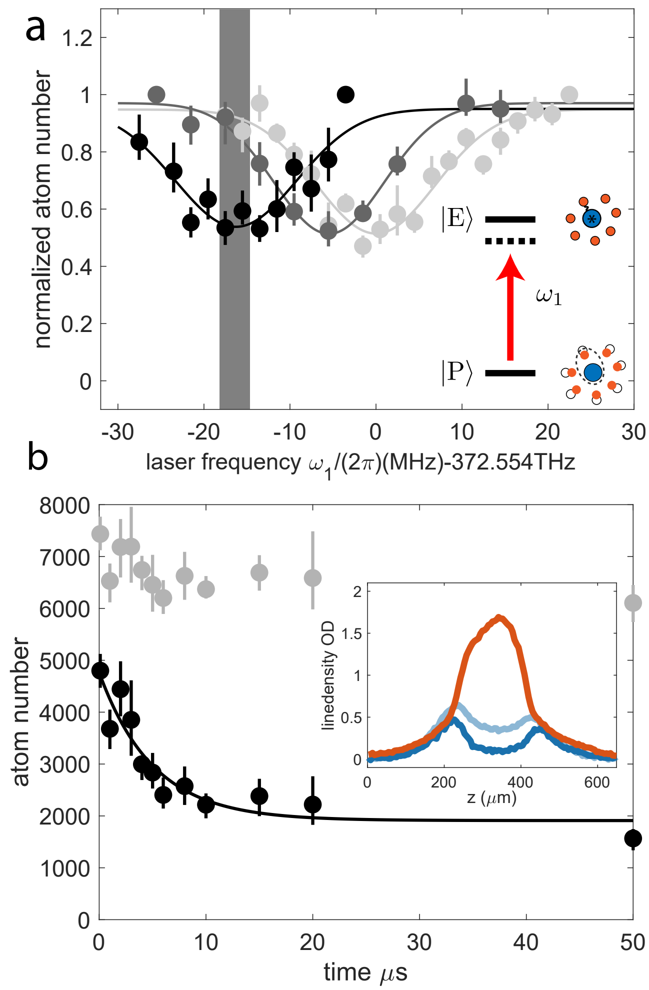

FIG. 2. Polaron photo-association spectroscopy. a. The up-leg laser frequency (ω_1) is scanned to measure photo-association (PA) resonances at 0.2, 0.6 and 1 W power (lighter gray, gray and black data points). The PA resonance frequency is measured to have a red-shift linear to up-leg laser intensity due to the nearby free-particle continuum. The gray shade marks where the maximum polaron to excited state coupling is achieved. b. Polaron PA time dynamics. Locally resolved impurities are measured with 1W up-leg laser power at fixed $-(2\pi)$19MHz detuning. As a result of strong dressing, impurities inside of the BEC Thomas-Fermi radius (black dots) are driven into the excited state with 5 μs exponential time constant. Impurities outside of the BEC Thomas-Fermi radius (gray dots) show little coupling. Inset shows the integrated linedensity of the bosonic majority (orange) and fermionic impurities (light blue at 0 μs and dark blue at 50 μs). The boson linedensity displays a BEC bimodal distribution and stays unperturbed during PA.

We perform dark resonance spectroscopy to characterize the initial polaron state coupling to the ground state

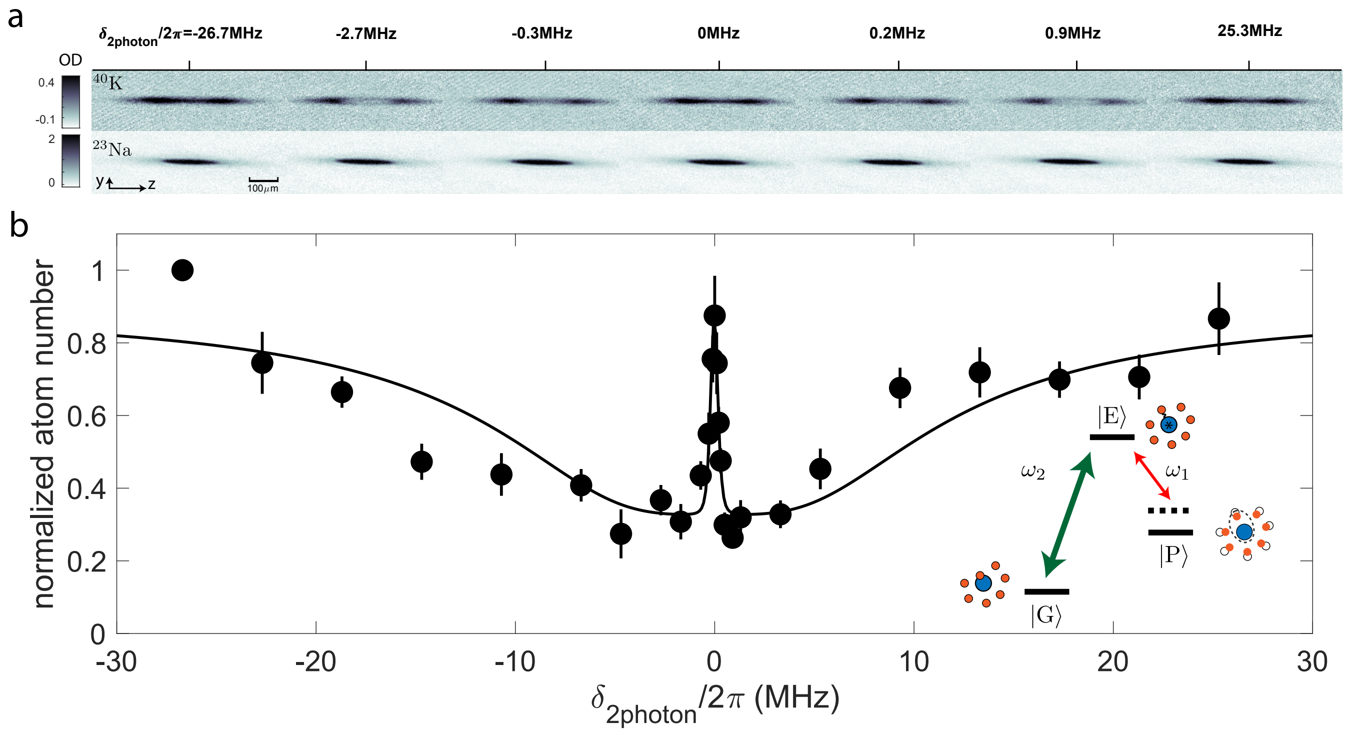

FIG. 3. Observation of polaron coupling to the absolute ground state via dark resonance spectroscopy. a. Exemplary absorption images of the fermionic ^{40}K and the bosonic ^{23}Na. At $\delta_{2\mathrm{photon}}/2\pi = -2.7, 0.9$ MHz, the BEC casts two shadows onto the fermions, manifesting fermions strongly dressed by the BEC are favored to couple to the excited state. b. Dark resonance spectrum with normalized counts of impurities within the BEC Thomas-Fermi radius. The inset illustrates the initial polaron state $|\mathrm{P}\rangle$, the excited state $|\mathrm{E}\rangle$ and the absolute molecular ground state $|\mathrm{G}\rangle$ connected through the up-leg ω_1 in horizontal polarization with 200 mW power and down-leg laser ω_2 in vertical polarization with 30 μW. Up-leg (Ω_1) and down-leg (Ω_2) Rabi rates are fitted to be $\Omega_1, \Omega_2/2\pi = 0.4, 3.2$ MHz.

via the electronically excited state. A down-leg laser (ω_2) in vertical polarization with 30 μW power is introduced as a pumping light with a fixed frequency that resonates with the ground state $|\mathrm{G}\rangle$ and the excited state $|\mathrm{E}\rangle$. Scanning the frequency of the up-leg laser (ω_1) changes the two-photon resonance condition ($\delta_{2\mathrm{photon}}$). When two-photon resonance conditions are dissatisfied, shadows of the BEC are imprinted onto the impurities shown in Fig. 2a $\delta_{2\mathrm{photon}}/2\pi$ =-2.7 and 0.9 MHz. These "negative" BEC prints on the impurities are results of the strongly dressed impurities photo-associating into excited molecules. In the narrow window where two-photon resonance conditions are satisfied ($|\delta_{2\mathrm{photon}}/2\pi| < 200$ kHz), the initial polaron state, the dark state of the system, are transparent to the up-leg and down-leg lasers. The dark resonance spectrum in Fig. 3b precisely determines the ground state molecule binding energy by the up-leg and the down-leg laser frequency differences. The up-leg and the down-leg Rabi rates are fitted to be $\Omega_1, \Omega_2/(2\pi) = 0.4, 3.2$ MHz based on an open three-level model. These results from the dark resonance spectrum pave a clear path to the dipolar molecule ground state.

We achieve dipolar molecule creation directly starting from polarons via a two-photon STIRAP. In the STIRAP sequence, the down-leg (Ω_2) and the up-leg (Ω_1) laser intensities (Ω_1^2, Ω_2^2) are ramped sinusoidally, and the dark state ($\approx \Omega_2 |\mathrm{P}\rangle + \Omega_1 |\mathrm{G}\rangle$) is adiabatically tilted from the polaron state $|\mathrm{P}\rangle$ to the ground state $|\mathrm{G}\rangle$. Fig. 4b demonstrates that the strongly dressed impurities (black dots) are coherently transferred into the absolute rovibrational ground state via a 30 μs STIRAP, and consequently back to polarons after a reverse STIRAP. Such a coherent impurity signal revival is locally resolved and seen only with the impurities within the BEC Thomas-Fermi radius. In contrast, the impurities outside of the BEC Thomas-Fermi radius (gray dots in Fig. 4b) do not participate in the STIRAP due to the lack of up-leg coupling. The STIRAP signal difference between strongly and weakly dressed impurities again reveals that the contact is the critical parameter for making dipolar molecules. We report on average 30% single STIRAP molecule conversion efficiency for the impurities strongly dressed by the BEC. The impurities that locate at the peak boson density are estimated to have a higher than average STIRAP efficiency. We realize 600 fermionic ^{23}Na^{40}K ground state molecules inside of the BEC, which is an inheritance from the initial polaron state. Since bosons are in the majority limit, the BEC is little perturbed by the STIRAP associ-

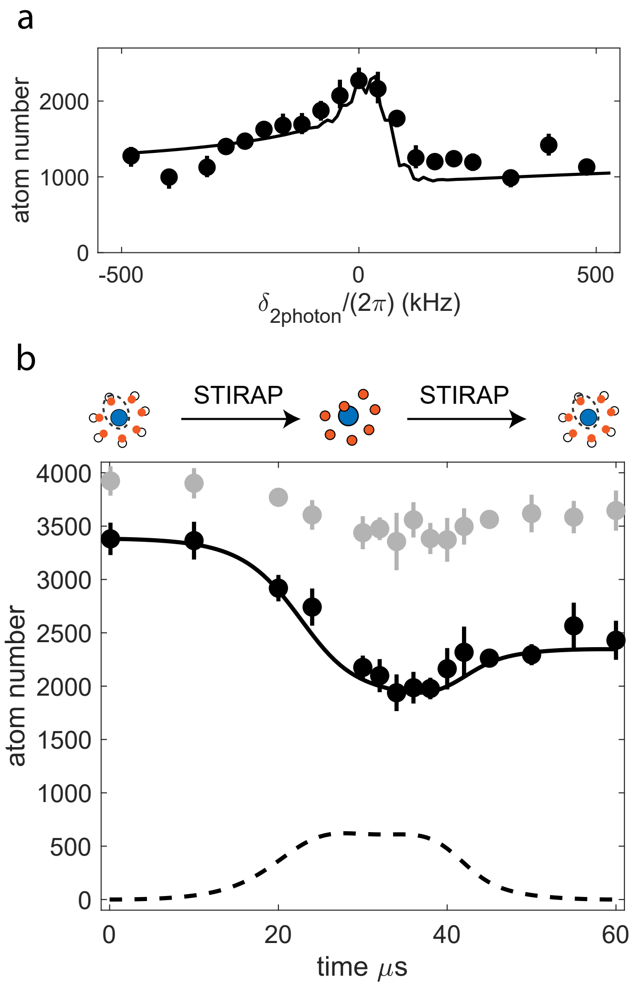

FIG. 4. Coherent population transfer from strongly interaction Bose polarons to the absolute ground state molecules via two-photon STIRAP. a. STIRAP two-photon resonance spectrum. Strongly dressed impurities that go through two consecutive STIRAP pulses are measured. The asymmetric STIRAP resonance lineshape is due to the nearby free-particle continuum. b. Evolution of the number of impurities during the STIRAP sequence. Impurities immersed inside of the BEC Thomas-Fermi radius (black dots) show a coherent revival, in contrast to unperturbed impurities outside of the BEC (gray dots). Black dashed (solid) line is the ground state molecule (impurity) number based on a STIRAP model with an extra spectator continuum state. Thirty percent STIRAP single trip efficiency is observed and six-hundred dipolar molecules are created inside of the BEC.

ation, which remains at its initial temperature T/T_c=0.1 and peak density $n_B = 7 \times 10^{13}$ cm^{-3}.

In conclusion, our method utilizes the contact of a many-body initial state to create ground state molecules immersed in a BEC bath. Feshbach molecules have a finite association efficiency, which is worth mentioning when compared with the polaron pathway. Our locally resolved 30% polaron-to-ground state molecule transfer is not inferior to the Feshbach molecule approach, when both weakly-bound molecule association and STIRAP efficiency are accounted for. Our prior work reported a 11% free fermion-to-ground state molecule efficiency with Feshbach molecules as an intermediate step [13, 14] (15% Feshbach molecule radio-frequency association and 75% single STIRAP efficiency). Recent experiments have achieved $6 - 20\%$ free fermion-to-ground state molecule transfer via magnetic association that converts a large portion of the BEC into Feshbach molecules [15–18]. Feshbach molecules have the disadvantage of a short lifetime due to rapid three-body recombinations with the surrounding bosons. We measure the NaK Feshbach molecules with Na to have a loss rate of $\beta = 10^{-10}$ cm^3/s. Polarons even at near unitarity-limited interactions have 50 times slower loss rate, $\beta = 5 \times 10^{-12}$ cm^3/s, which makes the polaron a much more stable initial state. Direct transfer from polarons creates a unique opportunity for ground state molecules to interact with the BEC. Unfortunately, the NaK ground state molecules are chemically reactive to Na, which results in a two-body loss rate of $\beta = 3 \times 10^{-10}$ cm^3/s. However, the novel BEC-ground state molecule interactions may be achieved with molecule species that are chemically stable [19] or that have Feshbach resonances with their constituent bosons [20]. Polarons created in this work are held in a harmonic trap, which creates a spatially dependent dressing. We propose a box trap as an ideal configuration for creating NaK ground state molecules via the polaron pathway. Bosons held in a box trap have a flattop density distribution and condense without changing their spatial distributions [21]. Fermions can be uniformly dressed by the bosons. We have an opportunity to create an equal Bose-Fermi mixture, where the entire box of atoms can be associated into the ground state molecules.

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Polaron wavefunction

Given the variational description for Bose polaron, the polaron wavefunction consists two parts, delocalized and molecular like components,

$$|\Psi^{(q)}\rangle = (\psi_0^q f_q^\dagger + \sum_{k\neq 0} \psi_k^{(q)} f_{q-k}^\dagger b_k^\dagger)\, |\mathrm{BEC}\rangle \tag{1}$$

The molecular components of the wavefunction has the same momentum distribution to a molecule with size $1/\kappa$. In real position space, the polaron assembles the delocalized (BEC-like) with quasi-particle weight Z and molecule-like part. Based on experiment parameters in this paper, $k_n = (1300a_{\mathrm{Bohr}})^{-1}$, $a = -4000a_{\mathrm{Bohr}}$, we calculate $\kappa^{-1} = 2600a_{\mathrm{Bohr}}$. Feshbach molecules that are transferred into the ground state molecules are created at 85.7G with $a = 1700a_{\mathrm{Bohr}}$, whose size is smaller than the polaron molecular part of the wavefunction thus better coupling the excited state.

Simple STIRAP model with an extra continuum

A simple continuum model is introduced here that's above the polaron state. The continuum has a characteristic coupling ratio to the polaron state.

Polaron and ground state molecule lifetime

Species	loss rate β (cm^3/s)
^{23}Na^{40}K Feshbach molecules with ^{23}Na	$\approx 10^{-10}$ [13]
^{40}K^{87}Rb Feshbach molecules with ^{87}Rb	3×10^{-10} [24]
^{23}Na^{40}K polaron with ^{23}Na	5×10^{-12} (this work)

TABLE I. Feshbach molecule loss rate