Thermal equilibrium and efficient evaporation of an ultracold atom-molecule mixture

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We derive the equilibrium conditions for a thermal atom-molecule mixture near a Feshbach resonance. Under the assumption of low collisional loss, thermodynamical properties are calculated and compared to the measurements of a recent experiment on thermal fermionic lithium atoms. We discuss and evaluate possible collision mechanisms which can lead to atom-molecule conversion. Finally, we propose a novel evaporative cooling scheme to efficiently cool the molecules toward Bose-Einstein condensation.

I. INTRODUCTION

Recent experiments witness a fast and remarkable progress in creating molecules from atomic Bose-Einstein condensates (BEC) [1–3], degenerate Fermi gases [4–6] or ultracold thermal gases [7,8]. These results have initiated new pursuits toward molecular BEC, Cooper-paired Fermi gas [9,10] as well as matter-wave interferometry based on ultracold molecules.

In all these experiments, the creation of molecules is based on magnetically tunable Feshbach resonances [11,12], which allow interacting atom pairs to couple to molecules in a single internal quantum state. Extensive studies on atomic Feshbach resonances not only lead to a thorough understanding of the cold atom collision properties, but also suggest intriguing possibility to convert atomic BEC or degenerate Fermi gas to a molecular BEC with high efficiency [13–16].

However, converting atoms into molecules via a Feshbach resonance is accompanied by other effects: three-body collisions [17–19], atom-molecule interactions [5], molecule-molecule interactions [8], as well as many-body effects in quantum degenerate gases [1]. These processes can lead to a low conversion efficiency or a short molecular lifetime, both limit the ability to observe and manipulate the molecules. As the detailed mechanisms of the conversion process require future investigations, it remains an experimental task to identify the best system and strategy to reach molecular condensation.

In most recent experiments, molecules are created by ramping the magnetic field through a Feshbach resonance. High conversion efficiencies of 50 % to 80 % are found in degenerate Fermi gases [4–6], which agree with the calculations [20,21]. Surprisingly, a recent experiment showed that a similar efficiency can be achieved in a thermal gas of fermionic ^{6}Li atoms at fixed magnetic field [8]. Furthermore, the molecule sample can be purified, trapped and remains stable with a lifetime of up to 10 s. The results of Ref. [8] prompt us to investigate the thermodynamics of a non-degenerate atom-molecule mixture and the possibility to cool the molecules to a molecular BEC.

In this paper, we introduce a thermodynamical model to calculate the atom-molecule conversion efficiency in a thermal gas (Sec. II). Using the model, we derive the atom-molecule (molecule-atom) conversion efficiency and compare the results with the experimental data (Secs. III and IV).

To further cool the molecules, we propose a novel evaporative cooling scheme in an atom-molecule mixture (Sec. V). Finally, we identify the relevant conversion mechanisms and calculate the time scale of the proposed cooling process (Sec. VI).

II. MODEL

Based on the fermionic lithium system, we consider a classical thermal sample with N and N' atoms in two internal states and M molecules. The molecules individually consist of two non-identical atoms and are in a single internal state with binding energy E, defined as the difference between the dissociation threshold and the uncoupled molecular state eigenvalue. See Fig. 1. Notice that when the molecular state is tuned into the dissociation continuum, the binding energy is negative ($E<0$). In this case, the molecular state is quasi-bound with a lifetime determined by the (Feshbach) coupling to the continuum.

We assume the system has negligible collision loss and energy exchange with the environment. The conservation of particle number and energy thus imposes two constraints in the thermalization process: the former is simply $N+M$
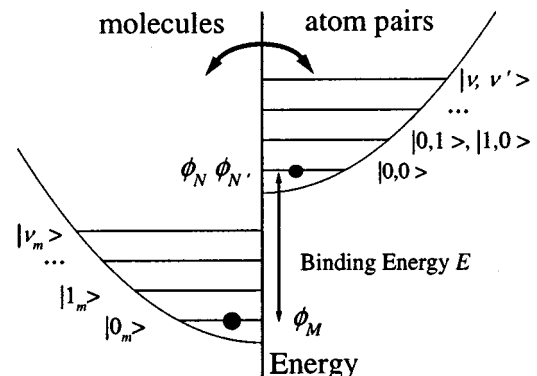

FIG. 1. Thermal equilibrium in an atom-molecule mixture. Molecular states $|\nu_m\rangle$ are introduced to the two-body states of nonidentical atom pairs $|\nu, \nu'\rangle$, where ν, ν' (ν_m) are the motional quantum numbers of a single atom (molecule). Population in $|0,0\rangle$ is the product of that in single-particle state $|\nu=0\rangle$ and $|\nu'=0\rangle$, namely, $\phi_N\phi_{N'}$. Molecular phase-space density ϕ_M is enhanced by the Boltzmann factor $e^{E/kT}$ when $E>0$.

$=$const and $N'+M=$const, while the latter is given by $Ne_n +N'e_{n'}+M(e_m-E)=$const, where $e_n(e_{n'})$ and e_m are the mean external energy per atom and molecule. We assume the gas is dilute with negligible interaction energy among particles.

The thermal equilibrium condition is obtained assuming that the atom-molecule mixture is in a quantum-mechanical canonical ensemble. This assumption is generally valid in the limit of large particle number [22]. Given the temperature T and single-particle partition function $Z_n(Z_{n'})$ and $Z_m e^{-E/kT}$ for atoms and molecules, we derive the equilibrium condition by minimizing the free energy $F=-kT \ln Z$, subjected to only the particle number conservation constraint, where $Z =Z_n^N Z_{n'}^{N'} Z_m^M e^{-ME/kT}(N!N'!M!)^{-1}$ is the partition function of the system, and k Boltzmann's constant. The result constitutes the key equation we investigate in this paper,

$$\phi_M = \phi_N \phi_{N'} e^{E/kT}. \qquad (1)$$

Here $\phi_N=N/Z_n$ ($\phi_M=M/Z_m$) is the atomic (molecular) phase-space density in the motional ground state.

This equation is valid for systems with atoms in two internal states and molecules in one state, as well as those with two atomic species, say, Rb atoms, Cs atoms with RbCs molecules. For single component systems with N atoms and M molecules, Eq. (1) is rewritten as $\phi_M=\phi_N^2 e^{E/kT}$ [7]. The only assumptions are the low collision loss, interaction energy and phase-space density. The applicability of this classical treatment to quantum degenerate gases has recently been suggested in Ref. [23].

The thermalization condition given in Eq. (1) can be understood in a simple picture. As the atomic phase-space density ϕ_N and $\phi_{N'}$ are the ground state populations of the two atomic components, $\phi_N \phi_{N'}$ is the population of the non-identical atom pairs in the lowest two-atom state. Equation (1) states that the motional ground state of the molecule can be regarded as one of the two-atom states. An additional Boltzmann factor $e^{E/kT}$ accounts for the binding energy. This is illustrated in Fig. 1. Notice that this result is independent of the details of the molecule formation and dissociation mechanisms.

Interesting consequences can be directly observed from Eq. (1). At constant temperature, molecular density is proportional to atomic density squared, even in the case when the molecules are formed, say, by a three-body process, whose rate depends on density cubed. This is because the reverse molecule dissociation rate also depends on atomic density. The balance of both the formation and dissociation processes in thermal equilibrium gives the density squared dependence. We will come back to the dissociation process in Sec. VI.

In the following sections, we calculate the atom (molecule) number $N(M)$ and temperature T in thermal equilibrium based on Eq. (1) and the conservation laws: we first determine the constants of the conservation laws from the initial conditions, then express partition function Z_n, $Z_{n'}$, and Z_m in terms of trapping potential and temperature T; mean energy in terms of partition function and temperature,

$e_i=-Z_i^{-1}\partial_\beta Z_i$, where $\beta=(kT)^{-1}$. In cases where no analytic solutions are available, we solve the equations numerically.

A special case is calculated here when the molecules and two atomic components are identically trapped $Z_n=Z_{n'}=Z_m$ with initial atom numbers N_0, N_0' and molecule number $M_0 =0$. The conversion efficiency $f=2M/(N_0+N_0')$ can be derived from Eq. (1) and particle conservation law as

$$f=\left(1+\frac{\phi_N+\phi_{N'}}{2\phi_N\phi_{N'}}e^{-E/kT}\right)^{-1}. \qquad (2)$$

When the atomic gas is tuned right on resonance $E=0$ with final phase-space density $\phi_N=\phi_{N'}=1$, an conversion efficiency of 50 % is obtained. Notably, the unity phase-space density assumption is on the border of the applicability of our model.

On the other hand, given an atom sample with low initial phase-space densities $\phi_{N_0}=\phi_{N_0'}\ll 1$ and zero binding energy, the conversion efficiency is simply $f=\phi_N=\phi_{N'}\sim \phi_{N_0}$. This result indicates that creating molecules from a cloud of thermal atoms by tuning the field right on Feshbach resonance is inefficient. In the following sections, we show that an appreciable gain in conversion fraction can be obtained when we tune the molecular state below continuum.

III. CONVERTING ATOMS TO MOLECULES

The atom-molecule conversion can be qualitatively understood from Eq. (1). When the molecular state is far below (far above) the atomic continuum, all particles should accumulate in the lower molecular (atomic) state and the atom (molecule) number is exponentially suppressed. This naive picture, however, is incorrect when one tries to convert cold atoms into molecules. At large binding energy, the internal energy released during the molecule formation process heats up the sample significantly and reduces the atomic phase-space density. In the following calculation, we show that the final temperature goes up approximately linearly with the binding energy and the molecule fraction is therefore limited. In the limit of infinite binding energy, counter-intuitively, no molecules are formed.

To show the conversion in the vicinity of a Feshbach resonance, we consider both atoms and molecules are harmonically trapped with identical single-particle partition function, $Z_n=Z_{n'}=Z_m=\prod_i(1-e^{-\hbar\omega_i/kT})^{-1}$, where ω_i is the trap vibration frequency in the ith direction. The external energy of the particles is $e_n=e_n'=e_m=3kT$. The assumption that both species have the same trap vibration frequencies is generally valid for atoms and long-range atomic dimers in a deep far-detuned dipole trap where the trap depth and mass for a molecule are both twice as large as those for an atom.

Based on the experiment parameters given in Ref. [8]: axial (radial) trap frequency of ω_z (ω_r)$=2\pi \times 260$ (0.39) kHz, initial atom number $N_0=N_0'=1.25\times 10^6$, molecule number $M_0=0$, and temperature $T_0=2.5$ μK, we calculate the molecule (atom) fraction, phase-space density and temperature in thermal equilibrium, shown in Fig. 2. A maximum conversion efficiency of $2M/(N_0+N_0')=M/N_0 \sim 50$ % agrees with the experimental results [24].
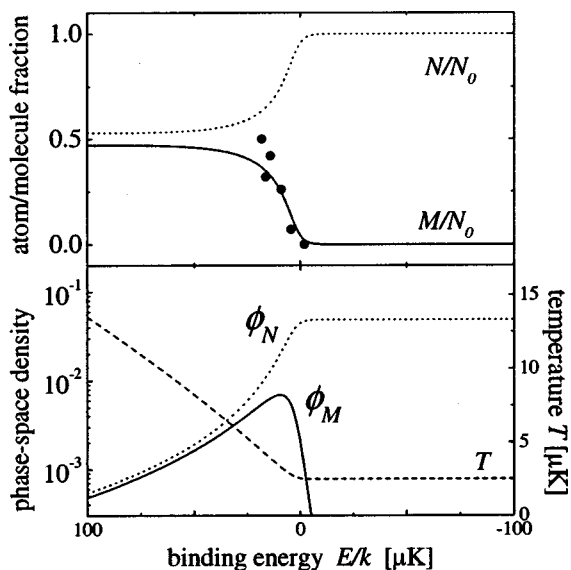
FIG. 2. Atom and molecule fraction in equilibrium starting with a pure atom sample. Trap parameters are specified in the text. In the upper figure, the atom fraction N/N_0 (dotted line) and the molecule fraction M/N_0 (solid line) are calculated and compared to the measurements (solid circles). The atom(molecule) fraction of 50 % at large binding energy is accidental. In the lower figure, the final phase-space density for molecules ϕ_M (solid line) and for atoms ϕ_N (dotted line) are shown together with the temperature T (dashed line).

Remarkably, the maximum conversion efficiency is reached at the binding energy of $E > k$ 20 μK. This value greatly exceeds the initial temperature of the sample and qualitatively explains the displacement of the maximum recombination loss position by ~ 150 G relative to the Feshbach resonance [25,26]. However, a generalization of our treatment to quantum-degenerate gas is necessary to predict the exact field shifts in those experiments. For even more negative binding energy, an abrupt turn-off of the molecule formation rate is observed and no experimental data on the molecule number in thermal equilibrium is available.

To calculate the best atom-molecule conversion efficiency, we assume initially $N_0 = N_0'$ atoms are in an isotropic harmonic trap with $Z_n = Z_{n'} = Z_m$, phase-space density $\phi_{N_0} = \phi_{N_0'}$ and binding energy E, we calculate the maximum molecular phase-space density ϕ_M or conversion fraction M/N_0 by varying the binding energy. Over the range of $\phi_{N_0} = 10^{-4} \sim 1$, the maximum phase-space density of the molecules ϕ_M is typically a factor of $5-20$ lower than ϕ_{N_0}, while the maximum conversion fraction varies from $30-55$ %, shown in Fig. 3. This very weak dependence on the initial atomic phase-space density is remarkable and suggests that an efficient conversion of atoms into molecules in thermal clouds is possible.

IV. CONVERTING MOLECULES TO ATOMS

Converting a pure molecule sample into atoms, as demonstrated in Ref. [8], shows different features. First, at large
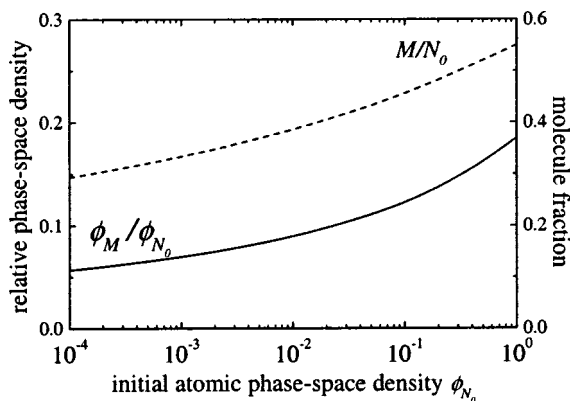

FIG. 3. Maximum conversion efficiencies in molecular phase-space density ϕ_M and in molecule number M from a pure atom sample. The calculation is performed assuming an isotropic harmonic trap, an initial atomic phase-space density ϕ_{N_0} and atom number $N_0 = N_0'$. Notice that maximum molecular phase-space densities ϕ_M (solid line) and numbers M (dashed line) are obtained at different binding energies E.

negative binding energy, all molecules dissociate into atoms. Beginning with $M_0 = 3 \times 10^5$ molecules at 2.5 μK with the same trap parameters described earlier, we calculate the molecule (atom) fraction M/M_0 (N/M_0) in thermal equilibrium, shown in the upper figure of Fig. 4. A full conversion from molecules to atoms is achieved when the molecular state is high above the scattering continuum. Shown together with the calculation is the experimental data [24].

The agreement between the experiment and calculation is excellent as there are no free parameters in the calculation. Both the experiment and calculation support the possibility
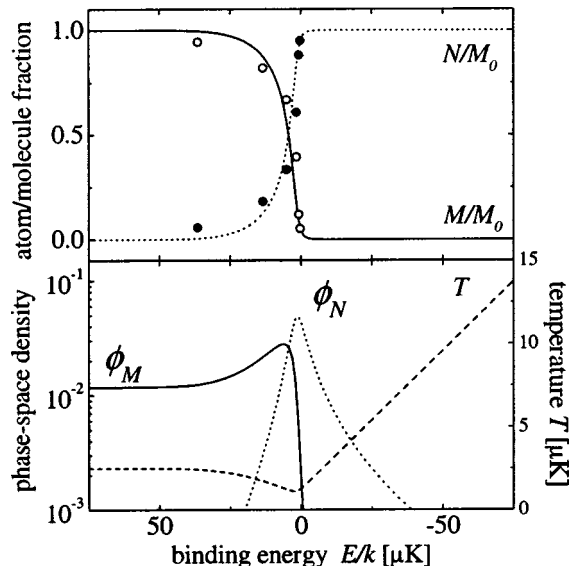

FIG. 4. Atomic and molecular fractions in equilibrium starting with a pure sample of M_0 molecules. In the upper figure, the fractions of molecules (solid line) and atoms (dotted line) are calculated and compared to the experimental data: atoms (solid circles) and molecules (open circles). In the lower figure, the phase-space density of the molecules (solid line) and atoms (dotted line) are shown together with the final temperature (dashed line).

to dissociate molecules into atoms even when the binding energy is positive and large compared to the initial molecular temperature. This is due to the much larger phase space of the atomic scattering continuum as compared to that of the molecular bound state.

Furthermore, a gain in phase-space density after thermalization is predicted at small positive binding energies where temperature drops and phase-space density peaks up, shown in the lower figure of Fig. 4. This cooling occurs due to two different processes. First, the dissociation process is endoergic and reduces the total external energy. Second, the total particle number, $N+N'+M$, increases after molecules dissociate and further reduces the mean energy per particle. When the equilibrium is reached, we find a gain in molecular (atomic) phase-space density of 2.4 (4.2), compared to that of the initial molecular phase-space density. This cooling effect is studied in more detail in Sec. V.

V. NOVEL EVAPORATIVE COOLING IN AN ATOM-MOLECULE MIXTURE

In conventional evaporative cooling schemes, energetic particles are removed from the trap. The remaining particles rethermalize and acquire a lower temperature and higher phase-space density.

The molecule-atom conversion process resembles the above process. By locating the atomic scattering continuum above the molecular state with positive binding energy, molecules with higher thermal energy are more probable to collisionally dissociate into atoms. While these particles are immediately removed in the conventional evaporative cooling recipe, the atoms from the dissociated molecules have a greatly reduced thermal energy and a rethermalization among these atoms and the remaining molecules is advantageous to cool the sample. To evaluate the cooling performance, we consider an initially pure sample of M_0 molecules in an isotropic harmonic trap with phase-space density ϕ_{M_0}, temperature T_0 and binding energy $E=\eta kT_0$, where η defines a truncation parameter, in analogy of the energy cutoff in the conventional evaporative cooling scheme. After the equilibrium is reached, we calculate the mean evaporative cooling efficiency based on $\gamma=-\ln(\phi_M/\phi_{M_0})/\ln(M/M_0)$ [27] for a wide range of truncation parameter η, shown in Fig. 5, where $\phi_M(M)$ is the molecular phase-space density (number) in equilibrium. At constant η, we find the atom-molecule thermalization permits a better cooling efficiency γ than does the conventional one.

On this basis, we propose a scheme to continuously cool molecules toward high phase-space density and provide a quantitative estimate on its performance. Given a harmonically trapped molecule sample with negligible collisions loss, temperature T and total external energy K, we control the molecular binding energy according to the cloud temperature and truncation parameter according to $E=\eta kT$. We assume atoms resulting from molecule dissociation are quickly thermalized with the sample before they are removed. The removal of only atoms from an atom-molecule mixture is demonstrated in Ref. [8]. During the thermalization process, a small molecular fraction ϵ that dissociates
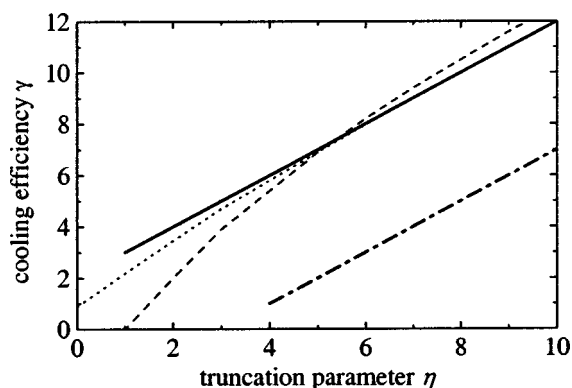

FIG. 5. Evaporation efficiency in an atom-molecule mixture. Evaporation efficiency γ during the thermalization process is evaluated for $\eta=E/kT_0=10^{-4}$ (dashed line), and $\eta=10^0$ (dotted line). Estimations based on the proposal (solid line) and on conventional evaporation (dash-dotted line) are also shown for comparison. No collision loss is considered in the calculation.

will lower the total external energy by $\delta K=-K\epsilon\eta/3$; next, the thermalization process shares the energy among the remaining $1-\epsilon$ molecules and 2ϵ atoms and lowers the temperature by $\delta T=-T\epsilon(\eta+3)/3$. Given the phase-space density in a harmonic trap $\phi_M=MZ_m^{-1}\propto MT^{-3}$, we obtain the evaporation efficiency as $\gamma=\eta+2$. The $\eta+2$ factor comes from the increase in particle number, together with the endoergic nature of the dissociation process. Compared to the conventional evaporation with $\gamma_c=\eta+k-4$ [27], where $0<k<1$, the new scheme with, say, $\eta=4$ can have an evaporative cooling performance comparable to that in conventional method with $10>\eta>9$, shown in Fig. 5.

However, this evaporation method involves additional atom-molecule conversion and thermalization steps and can potentially be slow due to either a slow thermalization rate or atom-molecule conversion rate. In the two-component fermion system, in particular, a large s-wave scattering length and elastic collision cross section for atom-molecule (molecule-molecule) scattering near an atom-atom Feshbach resonance are expected [19,28]. A fast thermalization rate on the order of $(1\text{ ms})^{-1}$ is possible from recent experiments. The conversion rate from molecules to atoms, however, depends on the details of the atom-molecule interaction mechanism and will be discussed in Sec. VI.

VI. REACTION MECHANISMS AND EVAPORATION RATE

To estimate the conversion rate or evaporation rate, specific molecule formation and dissociation mechanism need to be identified. We will consider two-component Li system as a specific example.
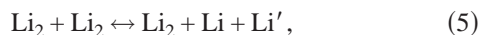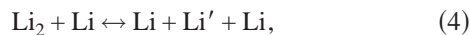
In a dilute gas with negative binding energy, molecules dissociate according to the following process:

$$\text{Li}_2 \leftrightarrow \text{Li} + \text{Li}'. \tag{3}$$

This spontaneous process is dominant since it is density independent. The molecular dissociation rate in this case is

characterized by the Feshbach resonance linewidth [29]. In typical experiments, Feshbach resonance linewidth of 0.1 MHz–10 MHz indicates a very short dissociation time of $\leqslant 1 \ \mu$s.

When the binding energy is tuned to positive values $E > 0$, the process described by Eq. (3) is forbidden due to the energy and momentum conservation. The leading binary processes to dissociate molecules and the corresponding reversed ternary or quaternary processes to form molecules are

$$Li_2 + Li \leftrightarrow Li + Li' + Li, \tag{4}$$

$$Li_2 + Li_2 \leftrightarrow Li_2 + Li + Li', \tag{5}$$

$$Li_2 + Li_2 \leftrightarrow Li + Li' + Li + Li'. \tag{6}$$

Notice that the dissociation (formation) processes are shown from left to right (right to left) and are allowed only when the total energy of the incident channel is sufficient to support the internal energy of the outgoing channel.

In thermal equilibrium, each molecule formation rate is exactly balanced by the corresponding dissociation rate, due to detailed balancing. For the above processes, we have $\overline{C_1 mn} = \overline{R_1 n^2 n'}$, $\overline{C_2 m^2} = \overline{R_2 mnn'}$ and $\overline{C_3 m^2} = \overline{R_3 n^2 n'^2}$, respectively, where $C_i(R_i)$ are the associated dissociation (formation) rate coefficients, n, n' (m) are the atomic (molecular) densities and $\overline{X}$ denotes the averaged value of X in a canonical ensemble. To directly relate the dissociation and formation coefficients, we assume $n(n')$ atoms and m molecules are uniformly distributed in a box with unity volume. The single particle partition function is given by $Z_n = Z'_n = 2^{-3/2} Z_m = \lambda_{dB}^{-3}$, where $\lambda_{dB} = h(2\pi m_0 kT)^{-1/2}$ is the thermal de Broglie wavelength of the atom, m_0 atomic mass and h Plank's constant. Using Eq. (1), we obtain the relationship between the coefficients in processes described by Eqs. (4)–(6):

$$\overline{C_1} = \overline{R_1} h^{-3} (\pi m_0 kT)^{3/2} e^{-E/kT}, \tag{7}$$

$$\overline{C_2} = \overline{R_2} h^{-3} (\pi m_0 kT)^{3/2} e^{-E/kT}, \tag{8}$$

$$\overline{C_3} = \overline{R_3} h^{-6} (\pi m_0 kT)^3 e^{-2E/kT}. \tag{9}$$

The above "formation-dissociation" relation reveals the rate coefficient when the counterpart coefficient is calculated or measured.

In samples which consist of mostly atoms, the dominant atom-molecule conversion process is given in Eq. (4). The formation reaction is widely studied in cold atom systems and is called three-body recombination [17–19]. For a two-component fermionic system, the rate coefficient is $\overline{R_1} = 167 a^6 kT/\hbar$ for $E/kT \gg 1$ [19]. Using the relationship $E = \hbar^2/m_0 a^2$, and Eq. (7), we immediately obtain the dissociation coefficient $\overline{C_1} = 3.75 \hbar^2 m_0^{-3/2} (kT)^{5/2} E^{-3} e^{-E/kT}$ for $E/kT \gg 1$ [30].

The situation is different when we start with a pure sample of molecules, since the dissociation process in Eq. (4) does not happen in the absence of atoms. Comparing Eq. (8)

and Eq. (9), we identify Eq. (5) as the dominant dissociation process at low temperature since $\overline{C_3}$ is exponentially suppressed relative to $\overline{C_2}$. This suppression can be understood as the molecule-molecule collision energy in Eq. (6) should be sufficiently high to support four atoms in the continuum, while in Eq. (5), only two atoms are in the continuum.

In contrast to that in Eq. (4), the formation process in Eq. (5) involves scattering of three non-identical particles: Li_2, Li, and Li' and is expected to scale as $R_2 \sim \epsilon^0$ at low collision energy ϵ in the three-body scattering channel. Consequently, dissociation coefficient scales as $C_2 \sim \epsilon^2$ [18]. Compared to $R_1 \sim \epsilon$ and $C_1 \sim \epsilon^3$ [30], the dominant conversion process in an atom-molecule mixture at low temperature limit is actually Eq. (5), which involves four atoms. A detailed calculation will be necessary to quantitatively determine either C_2 or R_2.

Knowing the possible dissociation mechanism for molecules, we estimate the speed of the proposed evaporative cooling based on Eq. (4), in which the rate coefficients are known. Given the evaporation parameter of $\eta = E/kT = 4$, temperature of 2.5 μK, and a small atom fraction with density $n = 2 \times 10^{11}$ cm^{-3}, we obtain an evaporation rate of $\overline{C_1} n = (2.5 \ s)^{-1}$, which is indeed much slower than the two-body collision rate. However, in conjunction with the predicted evaporation efficiency of $\gamma \sim 6$, we expect an increase of molecular phase-space density by six orders of magnitude can be achieved in $6s$, which is still within the lifetime of the molecular cloud $10s$ reported in Ref. [8]. Furthermore, during the evaporation process, temperature decreases and the dissociation process in Eq. (5) will eventually dominate and speed up the evaporation.

VII. CONCLUSION

We have described the equilibrium condition and conversion mechanisms in a thermal mixture of atoms and molecules under the assumption of negligible collisional loss. Near a Feshbach resonance, the molecular state extends the phase space of that of two atoms. A full thermalization in this extended phase space provides the equilibrium condition given in Eq. (1) and permits a quantitative estimation on the thermodynamical properties of an atom-molecule mixture.

In particular, we calculate the atom-molecule conversion efficiencies under various conditions and the results agree with the experimental data very well [8,24]. We show that by properly locating the binding energy of the molecules, the conversion of a significant fraction of atoms to molecules is possible even in a thermal gas with low phase-space density. Our result suggests an alternative scheme to reach quantum degeneracy: first, convert the fermionic atoms into bosonic molecules; second, evaporatively cool them to a molecular condensate and finally, convert them back to fermionic atoms. The last process would allow a creation of atomic Cooper pairs [9,10].

To cool the molecules to a molecular condensate, we suggest a novel cooling method based on the endothermic

molecule-atom conversion process at positive binding energy. We estimate a much higher evaporative cooling efficiency can be obtained as compared to conventional cooling in magnetic traps or dipole traps.

To estimate the time scale of thermalization and atom-molecule conversion, we identify a new collision process given in Eq. (5), which dominates the atom-molecule conversion at low temperature. The evaporation rate of the proposed scheme is estimated and an increase of molecular phase-space density by six orders of magnitude within the typical lifetime observed in the Li_2 thermal gas is expected [8]. This result highlights great prospect of attaining a molecular Bose-Einstein condensate.

Note added in proof. Recently, Bose-Einstein condensation of molecules was reported in Ref. [31].

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