Photoassociation and the Feshbach resonance are, in principle, feasible means for creating a molecular Bose-Einstein condensate from an already-quantum-degenerate gas of atoms; however, mean-field shifts and irreversible decay place practical constraints on the efficient delivery of stable molecules using either mechanism alone. We therefore propose Feshbach-stimulated Raman photoproduction, i.e., a combination of magnetic and optical methods, as a means to collectively convert degenerate atoms into a stable molecular condensate with near-unit efficiency.

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I. INTRODUCTION

In addition to translation, the molecular degrees of freedom include rotation and vibration, so that the laser cooling techniques used to create an atomic Bose-Einstein condensate (BEC) are difficult, if not impossible, to apply to molecules. Buffer gas cooling and Stark-deceleration methods are thus being developed as replacements for lasers in the precooling step towards a molecular BEC. Meanwhile, for systems where atomic BEC is already available, photoassociation and the Feshbach resonance have been proposed as a shortcut to the production of a molecular condensate.

Photoassociation occurs when a pair of colliding ultracold atoms absorb a laser photon, thereby jumping from the two-atom continuum to a bound molecular state. If the initial atoms are Bose-condensed, then the subsequent molecules will also form a Bose-Einstein condensate. Nevertheless, photoassociation generally occurs to an excited state, and the subsequent irreversible losses defeat the purpose of molecular coherence. Adding a second laser to drive molecular population to a stable electronic state, stimulated Raman adiabatic passage (STIRAP) in photoassociation of a BEC has been proposed as a means for avoiding radiative decay. However, collisions between particles make it difficult to achieve adiabaticity, which ultimately limits the practical efficiency of STIRAP to about fifty percent.

On the other hand, the process known as the Feshbach resonance occurs when two ultracold atoms collide in the presence of a magnetic field, whereby the spin of one of the colliding atoms flips, and the pair jump from the two-atom continuum to a quasibound molecular state. As with photoassociation, the so-formed molecules will constitute a BEC if the incident atoms are themselves Bose-condensed. Unfortunately, the quasibound condensate, while translationally and rotationally ultracold, is vibrationally very hot, and thus highly susceptible to collision-induced vibrational relaxation. Hence, while an adiabatic sweep of the magnetic field across the Feshbach resonance will create a molecular BEC with near-unit efficiency, coherence is again moot in the face of irreversible losses. Although adding a pair of lasers to this scheme allows for a stable molecular condensate, the practical efficiency of STIRAP is limited to about twenty percent.

The purpose of this Article is to develop a means for creating a stable molecular BEC that is near-unit efficient in both principle and practice. We therefore consider Feshbach-stimulated photoproduction, wherein the quasibound state formed in the presence of a static magnetic field is coupled via a Raman laser configuration to a stable molecular state. Again, a version of this scheme, in which STIRAP-inducing lasers are run concurrently with a sweep of the magnetic field across the Feshbach resonance, has been previously proposed; but, the efficiency of STIRAP is limited by the time-dependence of the quasibound energy arising from the changing magnetic field, and large system densities (> 10^{14} cm^{-3}) allow collision-induced mean-field shifts and vibrational relaxation to dominate atom-molecule conversion. On the contrary, we consider a static magnetic field interacting with a modest-density gas of atoms, which minimizes the role of mean-field shifts and vibrational relaxation, and enables the production of a stable molecular condensate with near-unit efficiency.

The development herein is outlined as follows. Section II introduces the model, along with the concept of collective enhancement. Section III illustrates the creation of a stable molecular condensate using a CW two-photon Raman configuration, as well as with transient STIRAP, both of which avoid radiative losses. In Sec. IV we discuss vibrational quenching and dissociation of the quasibounds, and estimate explicit numbers to determine the atomic systems for which such losses are negligible. Besides a summary, Section V considers our results in light of recent experiments with ^{85}\text{Rb}.

II. MODEL

We model a two-component quantum degenerate gas of atoms coupled via a Feshbach resonance to a condensate of quasibound molecules; additionally, one laser drives
Heisenberg equations of motion that govern the evolution of the atomic and molecular fields are

\[ i\dot{\phi}_\pm = (\mp)^\frac{1}{2} \alpha \phi_\pm \psi_1, \]  
\[ i\dot{\psi}_1 = \Omega_1 \psi_2 + \frac{1}{2} \alpha \phi \phi_2 + \frac{1}{2} \Omega_1 \psi_3, \]  
\[ i\dot{\psi}_2 = \delta \psi_2 + \frac{1}{2} \Omega_1 \psi_1 + \frac{1}{2} \Omega_2 \psi_3, \]  
\[ i\dot{\psi}_3 = \Delta \psi_3 + \frac{1}{2} \Omega_2 \psi_2. \]

Here \( \eta = 2 \) (for bosons (fermions), and the amplitudes \( \phi_\pm \) and \( \psi_1 \) have been taken as uniform over the volume of the system \( i = 1, 2, 3 \).

The strength of the Feshbach resonance, the process by which atoms are converted into quasibound molecules, is given by \( \tilde{a} = \frac{8\pi \sigma [a] \mu_{ma} \Delta R / m}{1/2} \), where \( a \) is the off-resonance s-wave scattering length, \( \mu_{ma} \) is the difference in magnetic moments between the quasibound molecule and free-atom pair, and \( \Delta R \) is the resonance width in magnetic-field units. The molecular energies are \( \hbar \omega_1 \), with the quasibound energy is given specifically in terms of the magnetic field by \( \hbar \omega_1 = \text{sgn}(a) (B - B_0) \mu_{ma} / 2 \), where \( B_0 \) locates the resonance position. Note that the magnetic-field dependence of \( \tilde{a} \) and \( \omega_1 \) are chosen so that, upon adiabatic elimination of the quasibound field, the resonance-induced atom-atom scattering length has the correct dispersive form \( \Delta R / |B - B_0| \). Finally, \( \Omega(1/2) \) denotes the Rabi frequency corresponding to laser L1 (L2), which has a frequency \( \omega_{L1,L2} \); correspondingly, the respective intermediate and two-photon detunings are \( \delta = \omega_2 - \omega_{L1} \) and \( \Delta = \omega_3 - \omega_{L1} + \omega_{L2} \).

We also scale the atomic and molecular field amplitudes in Eqs. (1) by the square-root of the total number density of particles: \( x \rightarrow x' = x / \sqrt{n} \), with \( x = \phi_\pm, \psi_1, \psi_2, \psi_3 \). Dropping the primes, the new order-unity field amplitudes evolve in time according to

\[ i\dot{\phi}_\pm = (\mp)^\frac{1}{2} \alpha \phi_\pm \psi_1, \]  
\[ i\dot{\psi}_1 = \Omega_1 \psi_2 + \frac{1}{2} \alpha \phi \phi_2 + \frac{1}{2} \Omega_1 \psi_3, \]  
\[ i\dot{\psi}_2 = \delta \psi_2 + \frac{1}{2} \Omega_1 \psi_1 + \frac{1}{2} \Omega_2 \psi_3, \]  
\[ i\dot{\psi}_3 = \Delta \psi_3 + \frac{1}{2} \Omega_2 \psi_2. \]

The term \( \alpha = \sqrt{n} \tilde{a} \) would have previously been referred to as the Bose-stimulated free-bound coupling \( \Omega \); however, since the above equations of motion are statistics independent, we realize that Bose stimulation (in this case) has nothing whatsoever to do with bosons, but is instead a many-body effect that applies equally well to Fermi-degenerate systems. This idea is implicit to a (Feshbach-) photoassociation-induced Bose-Fermi superfluid, as well as four-wave mixing in a Fermi-degenerate gas of atoms. Hereafter, we therefore coin a more appropriate term for \( \alpha \): the collective-enhanced free-bound coupling.

### III. Avoiding Radiative Decay

Given a Raman configuration, the brute-force way to avoid radiative decay from an electronically-excited state is a large intermediate detuning, and we thus consider the effective one-color problem obtained by adiabatically eliminating the excited-bound molecular field:

\[ i\dot{\phi}_\pm = (\mp)^\frac{1}{2} \alpha \phi_\pm \psi_1, \]  
\[ i\dot{\psi}_1 = \Omega_1 \psi_2 + \frac{1}{2} \alpha \phi \phi_2 - \frac{1}{2} \chi \psi_3, \]  
\[ i\dot{\psi}_3 = \Delta \psi_3 - \frac{1}{2} \chi \psi_1, \]

where \( \psi_1 \leftrightarrow \psi_3 \) transitions result from the two-color Rabi coupling \( \chi = \Omega_1 \Omega_2 / 2 \delta \), and the Feshbach (two-photon) detuning has picked up a Stark shift \( \omega_1 = \omega_1 - \Omega_1^2 / 2 \delta \). In such case, spontaneous decay will now occur at a rate \( \propto \Gamma_s / \delta \), and is managed by choosing \( \delta \) suitably large.

For simplicity, we focus on a single-component BEC as our initial quantum gas: \( \phi_\pm = \phi, i\dot{\phi} = \alpha \phi \psi_1 \). The field amplitudes are treated as \( c \) numbers, with initial conditions \( \phi(0) = 1 \) and \( \psi(t) = 0 \). As shown in Fig. 2, the atomic and stable molecular condensates undergo Rabi-like oscillations consistent with a nonzero but small detuning. This detuning is effectively induced by the presence of the quasibound condensate, so that, while the spontaneous decay of the excited-bounds is essentially eliminated by a large intermediate detuning, losses due to vibrational quenching and dissociation of the quasibound would still be encountered.

Although irreversible decay from the electronically-excited molecules is undoubtedly managed by a large intermediate detuning, we present a more elegant alternative: stimulated Raman adiabatic passage. The hallmark of STIRAP is the counterintuitive pulse sequence \( \delta \), which amounts to adjusting the two lasers so that initially, when most everything is in the quasibound molecular condensate \( \psi_1 \), the \( \psi_2 \leftrightarrow \psi_2 \) coupling is strongest, and finally, when effectively everything is in the stable molecular condensate \( \psi_3 \), the \( \psi_1 \leftrightarrow \psi_2 \) coupling is strongest. As the population moves from quasibound to stable molecules, the state with the larger population is always weakly coupled to the field \( \psi_2 \), thus keeping the population of the excited molecular condensate low (ideally zero) and reducing (eliminating) losses— even for zero intermediate detuning.
The idea is near that a properly-timed counterintuitive pulse sequence, applied to a degenerate atomic gas in the presence of a static magnetic field, will deliver a stable molecular BEC while avoiding radiative losses. Given the appropriate frequency scale as the collective-enhanced free-bound coupling $a$, then atoms will be coherently converted to quasibound molecules on a timescale given by $1/\alpha$, and we need only wait for sufficient population to build before effecting STIRAP. In our model we assume Gaussian laser pulses of the form $\Omega(t) = \Omega_0 \exp[-(t-t_i)^2/T^2]$, where $\Omega_0$ determines the pulse height $\Omega$, $t_i$ locates the pulse center, and $T$ defines the pulse width $\langle i \rangle = (1, 2)$.

This piece of intuition is correct, as shown in Fig. 3. We again use a single-component BEC as an example, with the pulse parameters specified as $\alpha T = 1/2$, $t_2 = 5T$, and $t_1 = 7T$. Adiabaticity is assured by selecting pulse heights such that $\Omega_0 T = 10^8$. The atoms are tuned close to the Feshbach resonance, and the molecules onto laser resonance, by the choices $\omega_1/\alpha = -10^{-2}$ and $\Delta = \delta = 0$. The plot shows the system coherently converting from atoms to quasibounds, and the laser pulses subsequently transferring nearly the entire population to the stable molecular condensate. Characteristic of a counterintuitive pulse sequence, radiative losses are expectedly minimized by a low excited-state population, of the order of $10^{-5}$. Again, the presence of a meaningful fraction of quasibound molecules means that the physics of quenching and dissociation must still be addressed, to which we now turn.

IV. QUENCHING, DISSOCIATION, AND EXPPLICIT NUMBERS

Given either a CW two-photon or transient STIRAP set-up, it is safe to neglect radiative decay from the excited molecular state, and the remaining practical issues involve the mean-field energy shift produced by s-wave collisions between particles, as well as loss-inducing vibrational relaxation and dissociation of the quasibound molecules. Particle-particle collisions shift the system off of resonance by an amount proportional to the s-wave scattering length; since the numbers are not well known for atom-molecule and molecule-molecule collisions, we take $\Delta = 4\pi\hbar\rho a/m$ as approximating the mean-field shift due to s-wave particle collisions in general. Meanwhile, vibrational relaxation in sodium occurs at a rate $2\Gamma \approx \rho \times 10^{-10} \text{ s}^{-1}$, and we use this value to estimate the effects of both atom-molecule and molecule-molecule vibrational quenching. Finally, dissociation of the quasibound back to the atomic continuum will occur analogous to photodissociation [2][21], and necessarily involves “rogue” modes defined as modes lying outside the atomic condensate (Fermi sea). All told, rogue dissociation is non-exponential, i.e., the Fermi Golden Rule and simple inclusion with a non-hermitian term [2][4] do not strictly apply [21], and will impose a rate limit on all-bosonic atom-molecule conversion given roughly by $R_L = 6 \omega_\rho$, where $\omega_\rho = \hbar \omega/\alpha m$ [2][21]. For fermionic atoms, the factor of two difference in the collective-enhanced free-bound coupling $a$ should lead to a doubling of the maximum rate, $R_L = 12 \omega_\rho$. [19]. From Fig. 3, ninety-five percent of the atoms are converted in $3/\alpha$ seconds, corresponding to a rate $R = 0.95\alpha/3$.

Explicit numbers for various systems [24][27] are listed in Table I, where the densities (resonance widths) were chosen as a balance between being low (high) enough to enable safe neglect of s-wave and vibrational-quench-inducing collisions, and large (small) enough to keep atom-molecule conversion below the rate limit. The table indicates that rogue dissociation is the main obstacle. Overall, the $^{23}\text{Na}$ (853 G) and $^{87}\text{Rb}$ (680 G) resonances appear as the strongest candidates. For a CW two-photon Raman scheme, intermediate detunings $\delta \approx 10^4 \text{ ms}^{-1}$ should manage typical spontaneous molecular decay rates $\Gamma_s \approx 10 \text{ ms}^{-1}$ over the timescale $\alpha^{-1}$, while at the same time allowing moderate bound-bound Rabi frequencies: $\Omega_1 = \Omega_2 \approx 0.1 \text{ ms}^{-1}$ for $\chi/\alpha = 1$. The STIRAP peak Rabi frequency $\Omega_0 \approx 1 \text{ ms}^{-1}$ is larger, but not unreasonable. If by chance irreversible quasibound decay proves more than a marginal issue, one might imagine a multi-shot STIRAP scheme (see also Ref. [24]), where a smaller fraction of the population is converted per shot, and losses are further reduced by keeping the population low.

V. SUMMARY

Admittedly, our inclusion of fermions is based on a BEC as an explicit example; nevertheless, the model itself is not dependent on statistics. Collective enhancement applies equally to fermions and bosons, so that the atom-quasibound conversion should still set the timescale $\sim 1/\alpha$, as is evident in Ref. [13]. Absent Pauli blocking, dominant rogue losses render the production of stable molecules inefficient for the $^6\text{Li}$ and $^{40}\text{K}$ resonances considered; nevertheless, in a manner analogous to the bosonic case [21] (see also below), collective oscillations between atoms and correlated dissociation pairs, i.e., Cooper pairs, could still occur.

We have, moreover, identified the 155 G $^{85}\text{Rb}$ resonance as a severely rogue-dominated system ($R/R_L \approx 10$), meaning that it is useless for producing any significant fraction of molecular condensate, stable or otherwise. On the other hand, Feshbach-only experiments have led to the observation of bursts of atoms emanating from a remnant condensate [2], and follow-up experiments using pulsed magnetic fields indicate large-amplitude oscillations ($\sim 30\%$) between remnant and burst atoms with a good many of the initial atoms gone missing—prompting speculation as to the formation of a molecular condensate [24]. Although we have no doubt as to the achievement of atom-molecule coherence, the
amplitude of the number of molecules present: the double-pulse experiments [28] realize a Ramsey-type [29] interferometer, which maps out collective oscillations between atoms and dissociated atom pairs, and which is highly sensitive to a small fraction of molecular condensate (∼1%) [21] (see also Ref. [22]).

In conclusion, we have presented a method for preparing a stable molecular condensate with near-unit efficiency: (i) start with a quantum degenerate gas of atoms, a magnetic field tuned at or near a Feshbach resonance, and a pair of lasers arranged in a Raman configuration; (ii) induce either CW two-photon Rabi flopping or transient STIRAP; (iii) obtain a stable molecular condensate. Favorable systems for implementation are $^{23}$Na and $^{87}$Rb. The advantage of this scheme is near-unit efficiency, which implicitly avoids rather large inelastic losses incurred by molecular collisions with unconverted atoms [28], and delivers a truly stable condensate of molecules. Moreover, experiments with photoassociation alone [33] are just on the verge of coherent conversion [24], and culmination requires a difficult-to-impossible balancing of low density and high laser intensity to simultaneously minimize mean-field shifts and rogue dissociation; a magneto-optical scheme may therefore provide the best opportunity for observing large-scale collective atom-molecule oscillations.

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FIG. 1. Few-level diagram corresponding to Feshbach-stimulated photoproduction of a stable molecular condensate. For ease of illustration, we consider a single-component gas of atoms that has Bose condensed into state \( |0\rangle \). The collective-enhanced Feshbach coupling is \( \alpha \), and the corresponding detuning is \( \omega_1 \). Similarly, the laser 1 (2) coupling is \( \Omega_1(2) \), and the two-photon (intermediate) detuning is \( \Delta(\delta) \). The full two-component case would involve replacing the level \( |0\rangle \) by a pair of (ideally) degenerate levels, with a molecule formed by removing one atom from each level.

FIG. 2. Feshbach-stimulated two-photon atom ↔ stable molecule conversion. Choosing \( \alpha = 1 \) sets the unit of frequency (time). The two-photon Rabi frequency is \( \chi = 1 \), and the system is tuned to Stark-shifted resonance, i.e., \( \omega'_1 = \Delta' = 0 \). (a) The atomic (solid line) and stable molecular (dashed line) condensates oscillate out of phase as if there were a small yet nonzero detuning. (b) The detuning is effectively induced by the presence of the quasibound condensate.

FIG. 3. Creation of a stable molecular condensate via Feshbach-stimulated Raman adiabatic passage. Frequency (time) is again in units of \( \alpha \). (a) As the collective conversion of atoms to quasibounds nears completion (solid line), the lasers transfer the population to the stable molecular condensate (dotted line) with ninety-five percent efficiency. The maximum fraction of electronically-excited molecular condensate (not shown) is of order \( 10^{-3} \). (b) The gaussian laser pulses have equal widths and heights: \( T = 1/2 \) and \( \Omega_0 = 2 \times 10^3 \).

TABLE I. Explicit numbers for various systems. Resonance positions are given in G, densities in \( 10^{14} \text{ cm}^{-3} \), and atom-molecule couplings in ms\(^{-1} \). To estimate \( \alpha \), we assume \( \mu_{ma} = \mu_B \), where \( \mu_B \) is the Bohr magneton.

| Atom | \( B_0 \) | \( \rho \) | \( \alpha \) | \( \Delta/\alpha \) | \( \Gamma/\alpha \) | \( R/R_L \) |
|------|--------|-------|-------|----------|----------|--------|
| \(^7\text{Li}\) | \( 22 \) | \( 800 \) | 1 | 3989 | 0.0008 | 0.0013 | 1.077 |
| \(^{23}\text{Na}\) | \( 23 \) | \( 853 \) | 0.01 | 4.13 | 0.029 | 0.012 | 0.079 |
| \(^{85}\text{Rb}\) | \( 25 \) | \( 155 \) | 0.01 | 142.4 | -0.0013 | 0.0004 | 10.1 |
| \(^{87}\text{Rb}\) | \( 25 \) | \( 680 \) | 0.1 | 8 | 0.061 | 0.062 | 0.013 |
| \(^6\text{Li}\) | \( 26 \) | \( 800 \) | 0.1 | 9635 | 0.0007 | 0.00005 | 5.17 |
| \(^{40}\text{K}\) | \( 27 \) | \( 190 \) | 1 | 1264 | 0.015 | 0.004 | 0.974 |