The primary purpose of this proposal is to explore the physics of multi-component condensates containing both atomic and molecular species. The adiabatic condition, which sets the limit to the powers and the widths of the laser pulses for an efficient STIRAP, is studied in connection with the collective excitation modes of the CPT state. The resonant Fermi model in which atoms of two opposite spins are coupled to the Feshbach molecules is studied from a quantum optics perspective by equating it to the single-mode laser cavity model. The same model, when a
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List of papers submitted or published that acknowledge ARO support during this reporting period. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)


Number of Papers published in peer-reviewed journals: 11.00

(b) Papers published in non-peer-reviewed journals or in conference proceedings (N/A for none)
(c) Presentations


Number of Presentations: 9.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts): 0

Peer-Reviewed Conference Proceeding publications (other than abstracts):

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts): 0

(d) Manuscripts

Number of Manuscripts: 0.00

Patents Submitted
## Patents Awarded

## Awards

### Graduate Students

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The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields: ...... 3.00
Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale): ...... 3.00
Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering: ...... 0.00
The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense: ...... 0.00
The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields: ...... 2.00

Names of Personnel receiving masters degrees

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Sub Contractors (DD882)

Inventions (DD882)
How to choose the powers and durations of applied laser pulses is a question that one must ask before embarking any STIRAP experiments. The answer to this question can be found from the adiabatic theorem. For linear atomic $\Lambda$ systems, one can always rely on the eigenenergy states as a basis for constructing the adiabatic theorem [1, 2]. For nonlinear atomic-molecular $\Lambda$ systems, the eigenenergy states are no longer useful because the nonlinear quantum system does not support the usual superposition principle [3, 4]. What become important in the nonlinear system are the collective excitation modes.

In this project, by linking nonadiabaticity with population growth in the collective excitation modes of the dark state, we have demonstrated, for the first time, the proper way to derive the adiabatic condition for nonlinear systems is through a linearization procedure similar to the Bogoliubov transformation. In our derivation, we have used the Bogoliubov normal modes (instead of the usual eigenenergy states) and overcome the mathematical obstacles stemming from the Goldstone mode and the biorthonormality of the Bogoliubov normal modes [5].

The establishment of this adiabatic theorem, not only completes the theory of the nonlinear STIRAP from the theoretical point of view, but also makes a qualitative evaluation of the adiabatic condition possible even in the presence of the mean-field shifts, which are an essential part of any condensate system.

Papers:
I. COUPLED TWO-SPECIES ATOM-HETERONUCLEAR-MOLECULAR CONDENSATE SYSTEMS

A. Properties of a coupled two-species atom-heteronuclear-molecule condensate

Most current studies, for converting ultracold atoms into ultracold molecules by means of magnetoassociation (Feshbach resonance) or photoassociation, are concerned with homonuclear molecules. As a natural progression, quantum-degenerate heteronuclear molecules are expected to be the next challenge to the atomic physics community, because heteronuclear molecules possess intriguing properties that will open up many new avenues of research. For example, a large electric-dipole moment can be induced in heteronuclear molecules with the prospect of creating dipole superfluids [6] and with potential applications in quantum computing [7] and quantum simulations [8], and a test of the fundamental symmetry [9].

In this project, we have considered an atom-molecule condensate system as shown in Fig. 2 in which a heteronuclear molecular condensate of mode $|m\rangle$ is coupled to two different atomic species (one in mode $|1\rangle$ and the other in mode $|2\rangle$). An important contribution of this project is that it has generalized the theory of coherent conversion of atoms into molecules through Feshbach resonance from systems with same atomic species to those with different atomic species. Such a generalization has opened the possibility for creating heteronuclear molecular condensates by the method of rapid adiabatic passage, whose application has so far been limited to the coupled atom-molecule condensate systems with same atomic species. In addition, this study also generalized the theory of coherent population oscillations from homonuclear to heteronuclear molecular system coupled to atomic condensates. A measurement of the oscillation frequency can tell us many properties of the system such as the molecular binding energy, atom-molecule coupling strength, etc. We emphasize that proving the integrability of this complex system and having available an analytical formula for the period are important steps in motivating the experimental efforts for demonstrating coherent oscillations in this model.

Papers:
B. Phase Separation in a coupled atom-molecule condensate with interspecies Feshbach Resonance

The question of whether two condensates coexist in the same spatial volume (miscibility) or repel each other into separate spatial regions (immiscibility or phase separation) or even collapse when they are brought together has remained an active research topic [10–14] ever since the experimental realization of multi-component condensate systems in the late-90s. The ability to change the two-body interactions by tuning the magnetic field across the Feshbach resonance in atomic systems has enabled the observation of phase separations, that are otherwise impossible with traditional condensed matter environments, to be accessed experimentally. The recent experimental demonstration of rich phase structure including the phase separation in an unbalanced fermionic superfluidity system [15, 16] serves as a great testament to the remarkable controllability that the technique of Feshbach resonance can bring to the atomic system.

In this project, motivated largely by the recent experimental efforts both in achieving the tunable interaction between $^{41}\text{K}$ and $^{87}\text{Rb}$ [17] and in demonstrating the ability of the Feshbach resonance to control the miscibility of a $^{85}\text{Rb}-^{87}\text{Rb}$ dual-species BEC [18], we consider the bosonic heteronuclear model with interspecies Feshbach resonance as shown in Fig. 2 again. In contrast to the earlier project [19], in which we concentrated on the coherent creation of heteronuclear molecular condensates, in this project, we will focus on constructing zero-temperature phase diagrams, paying particular attentions to phase separation and collapse. We have identified the interspecies Feshbach interaction to be the cause for the absence of the doubly mixed phases, as well as the reason for the presence of both a pure atom phase (AMF) and a pure molecule phase (MSF) in our model. This provides the opportunity, as we have indeed verified numerically, for observing the phase separation not only between mixed atom-molecule phase (AMSF) and a pure molecular phase (MSF) but also those between AMSF and pure ASFs. Under certain conditions, we have even found that our system is able to phase separate into two distinct AMSFs, which, we speculate, is a property unique to the heteronuclear model.

Papers:

Conference Presentations:
II. COHERENT POPULATION OSCILLATION FROM A FERMI ATOM-MOLECULE DARK STATE

Coherent association of ultracold atom pairs into diatomic molecules via Feshbach resonance [20] or photoassociation [21], has made it possible to create coherent superposition between atomic and molecular species at macroscopic level. This ability is the key to applications that employ the principle of the double pulse Ramsey interference interferometer [22] for observing coherent population oscillations between atoms and molecules [23, 24]. A particular kind of state, atom-molecule dark state, has been theoretically proposed [25, 26] and experimentally observed [27], where population is trapped in the superposition between atom pairs and deeply bound molecules in electronic ground state.

So far, macroscopic atom-molecule dark state has only been studied in the context of bosonic atom-molecule systems. The purpose of this project is to explore the possibility of forming a similar dark state and using it to create coherent oscillations in fermionic systems where a two-component atomic Fermi gas is coupled to bosonic molecules as illustrated in Fig. 3. At zero temperature, bosons all condense to zero momentum states and hence are described by discrete internal states only. Fermions, by comparison, are characterized not only by their internal states, but also by their external states which form the momentum continuum. This difference makes the study of the dark state in the fermionic systems far more complex than that of the dark state in the bosonic systems.

We have made two important and original contributions. The existence of the momentum continuum appears to make the two-photon resonance, the necessary condition for creating the dark state, impossible to realize. First, we have shown that this is not the case for systems where the interaction between atoms of opposite spins is attractive. This is because under the attractive interaction, fermi atoms can undergo a phase transition into the fermionic superfluid state (a condensate of cooper pairs of atoms), and it is then possible to achieve the two-photon resonance between the ground molecular state and the fermionic superfluid state by properly tuning the laser frequencies. Second, we have recognized that the fermionic systems, where discrete molecular states are coupled to fermionic momentum continuums, can be regarded as the nonlinear analog of the Fano-Anderson type of models in linear atomic
and condensed matter systems [28]. This analog has enabled us to gain significant insights into the collective excitation spectrum and its relation to the coherent oscillation dynamics. We are confident that these conceptual breakthroughs will greatly help to advance the study of fermionic systems in the coming years.

Submitted Paper:

Conference Presentations:

III. ATOM-MOLECULE BISTABILITY IN THE FERMI FESHBACH RESONANCE MODEL

The ability to cool and trap neutral atoms down to quantum degenerate temperature has created a host of new and exciting problems that are increasingly interdisciplinary both to the atomic-molecular-optical physics and to the condensed matter physics. The rich knowledge and experience accumulated over the past several decades in these interdisciplinary fields have dramatically accelerated the development pace of cold atomic and molecular physics.

In this project, we consider the Fermi Feshbach model as shown in Fig. 4 to illustrate how the
interdisciplinary fields learn and benefit from each other. This resonant Fermi superfluid model has recently attracted significant attention due primarily to the possibility of using it as a test ground for BEC-BCS crossover, a phenomenon thought to be underlying the mechanism of high temperature superconductors and extensively studied in the realm of condensed matter physics. The study of the same model has, however, also benefited greatly from the study of optics. In fact, this model was recently shown to display collective dynamics similar to photon echo and solitonlike oscillations in transient collective coherent optics.

In this project, we have drawn another analogy of the Fermi superfluid, that it can be mapped to the laser model which describes a single-mode laser field interacting with an ensemble of inhomogeneously broadened two-level atoms (Fig. 4). The establishment of such an equivalence enables us to inject new excitement, from the perspective of laser theory or cavity optics, into this problem of significant current research interest. In particular, we have explored the nonlinearity and studied the matter wave analog of the optical bistability in cavity, a popular topic in the 80’s, due chiefly to the possibility of using it as a basic building block for optical computing.

Conference Presentations:


IV. ACHIEVING GROUND POLAR MOLECULAR CONDENSATES BY A CHAINWISE ATOM-MOLECULE ADIABATIC PASSAGE

A condensate of ground polar molecules with large permanent electric dipoles represents a novel state of matter with long-range and anisotropic dipole-dipole interactions, that are highly amenable to the manipulation by DC and AC microwave fields [29]. As such, creation of such a condensate is expected to be celebrated as another milestone that promises to greatly spur activities at the forefront of physics research, particularly with respect to quantum computing and simulation [30], and precision measurement [31]. The road to molecular condensation is, however, complicated by the fact that more degrees of freedom are needed to describe molecules than atoms. In particular, cooling particles by entropy removal, a direct method popular with atoms, has so far been unable to lower the temperature of molecules down to the regime of quantum degeneracy. Thus, most current experimental efforts in creating ultracold heteronuclear molecules have all taken a different approach, using instead a simple stimulated Raman adiabatic passage (STIRAP) involving a single pair of counterintuitive Raman pulses [32–34].

In this project, we focus on the coupled multi-level atomic-molecular condensate systems where the role of the initial transition is played by photoassociation. An example (containing three pairs of Raman pulses) is provided in Fig. 5. A laser field associates atoms from two distinct species \( |0_1 \rangle \) and \( |0_2 \rangle \) into molecules of state \( |1 \rangle \) in the excited electronic manifold with a coupling strength \( \Omega_1 \) proportional to the laser field and the free-bound FC factor. Simultaneously, a series of laser fields of (molecular) Rabi frequency \( \Omega_i \) \( (i \geq 2) \) are applied to move the molecules from the excited to the ground state \( |6 \rangle \) via additional intermediate energy states. Our goal is to apply the concept of atom-molecule dark state, a coherent population trapping (CPT) superposition between stable ground species [35, 36], to generalize chainwise STIRAP from a pure molecular system [37] to a coupled multi-level atomic-molecular condensate systems.
We have found that this scheme has several attractive properties. First, atoms are directly converted into ground molecules. Thus, the loss of atoms typically associated with the initial preparation of Feshbach molecules [32] is never an issue here. Second, pulses of longer durations can be employed to meet the adiabatic condition; we can do so because the atom-molecule dark state is far more stable than the molecular dark state, where the initial state is highly unstable compared to the ground (atom or molecule) states. Finally, the use of intermediate lasers presents us with a new control knob inaccessible to typical three-level models. We have shown that an appropriate exploration of both the intermediate laser fields and the stability property of the dark state can greatly reduce the power demand for the PA laser needed for an efficient conversion. This along with other efforts involving the use of Feshbach resonance may help to combat the weakness in photoassociation, a key concern to STIRAPs starting from free atoms due to the free-bound FC factor being typically very small.

Papers:

Conference Presentations:

VI. ATOM-MOLECULE BISTABILITY WITH AND WITHOUT OPTICAL CAVITY

The ability to cool and trap neutral atoms down to quantum degenerate regime has created a host of new and exciting problems that are increasingly interdisciplinary, bridging in particular the atomic, molecular, and optical physics and the condensed matter physics. The rich knowledge and experience accumulated over the past several decades in these fields have dramatically accelerated the progress of ultracold atomic physics.

In this project, we consider a system where a bosonic molecule is coupled to two bosonic or fermionic
constituent atoms via Feshbach resonance or photoassociation. Such a system not only represents an excellent example that serves to illustrate how the interdisciplinary fields learn and benefit from each other, but also an ideal test ground for studying coupled atom-molecule condensates and the BCS-BEC crossover [38]. The latter is thought to be underlying the mechanism of high temperature superconductors and extensively studied in the realm of condensed matter physics. In addition, the coupled atom-molecule systems have deep quantum optical analogies [39, 40]: bosonic molecules coupled to bosonic atoms is the matter-wave analog of parametric coupling of photons which has important applications in generating nonclassical light fields and, more recently, in quantum information science; while the system of bosonic molecules coupled to fermionic atoms can be mapped to the Dicke model where a light field interacts with an ensemble of two-level atoms, a model having fundamental importance in the field of quantum optics.

In this project, we have further explored these quantum optical analogies of the atom-molecule system and have focused on the important effects of binary collisional interactions between atoms which are largely ignored in previous studies [39, 40]. We have shown that the atom-atom interaction introduces extra nonlinear terms which, under certain conditions, give rise to matter-wave bistability in both bosonic and fermionic models. Further, we have explored the system inside an optical ring cavity (Fig. 6). The ability of an optical cavity to provide feedback between input and output light fields can result in the modification of atom-photon interaction in a highly nonlinear fashion. We have shown that even when the effective Kerr nonlinearity due to s-wave collisions is not sufficiently negative, bistability can still occur, provided that atom-photon coupling, a key parameter describing the cavity-mediated two-body interaction, is sufficiently large. Hence, we have established the connection between the coupled atom-molecule quantum gases and the nonlinear bistable systems [41] that have been extensively studied in the 80’s in the context of nonlinear optics, due both to its fundamental interest, and to its many practical applications in fast optical switches, optical memory, laser pulse shaping, etc.

Papers:
FIG. 7: (a) The bare state picture of our model. (b) The dressed state picture of our model equivalent to (a). (c) A possible realization in $^6\text{Li}$. Here the states labelled by $|i\rangle$ ($i = 1, 2, \ldots, 6$) are the 6 ground state hyperfine states. Most experiments involving $^6\text{Li}$ are performed with a magnetic field strength tuned near a Feshbach resonance at 834G. Under such a magnetic field, the magnetic quantum number for the nuclear spin $m_I$ is, to a very good approximation, a good quantum number. The values of $m_I$ are shown in the level diagrams. Two-photon transition can only occur between states with the same $m_I$. Any pair of the lower manifold ($|1\rangle$, $|2\rangle$, and $|3\rangle$) can be chosen to form the pairing states. In the example shown here, we choose $|1\rangle = |\uparrow\rangle$, $|2\rangle = |\downarrow\rangle$ and $|6\rangle = |g\rangle$. The excited state $|e\rangle$ (not shown) can be chosen properly as one of the electronic $p$ state.

A. Detection of Fermi Pairing via Electromagnetically Induced Transparency

A unique phenomenon of low temperature Fermi system is the formation of correlated Fermi pairs. How to detect pair formation in an indisputable fashion has remained a central problem in the study of ultracold atomic physics. Unlike the BEC transition of bosons for which the phase transition is accompanied by an easily detectable drastic change in atomic density profile, the onset of pairing in Fermi gases does not result in measurable changes in fermion density. Early proposals sought the BCS pairing signature from the images of off-resonance scattering light [42]. The underlying idea is that to gain pairing information, measurement must go beyond the first-order coherence, for example, to the density-density correlation. This is also the foundation for other detecting methods such as spatial noise correlations in the image of the expanding gas [43], and radio frequency (RF) spectroscopy [44, 45], etc.

In this project, we propose an alternative detection scheme, whose principle of operation is illustrated in Fig. 7(a). In our scheme, a relatively strong coupling and a weak probe laser field between the excited state $|e\rangle$ and, respectively, the ground state $|g\rangle$ and the spin up state $|\uparrow\rangle$, form a $\Lambda$-type energy diagram, which facilitates the use of the principle of electromagnetically induced transparency (EIT) to determine...
the nature of pairing in the interacting Fermi gas of two hyperfine spin states: $|↑⟩$ and $|↓⟩$. EIT [46], in which a probe laser field experiences (virtually) no absorption but steep dispersion when operating around an atomic transition frequency, has been at the forefront of many exciting developments in the field of quantum optics [47].

In this project, we have presented the expression of the probe absorption coefficient from both quantum optics and condensed matter physics approach, taking into consideration the pairing fluctuations in the framework of the pseudogap theory [48]. We have also constructed a quasiparticle picture Fig. 7(b) that proves convenient to explain the features of the spectrum, especially, the spectroscopic features at the finite temperature. We have demonstrated that the EIT technique offers an extremely efficient probing method and is capable of detecting the onset of pair formation (i.e., determining $T^*$). We have also compared the RF method and our EIT method. In the method of RF spectrum [44, 45], an RF pulse is applied to the sample followed by a destructive measurement of the atom numbers using laser imaging. The RF signal is defined as the average rate change of the population in state $|g⟩$ during the RF pulse, which can be inferred from the measured loss of atoms in $| ↑⟩$. In the EIT method, by contrast, one can directly measure the absorption spectrum of the probe light. Applying a fast frequency scan to the weak probe field, the whole spectrum can be recorded continuously in an nearly non-destructive fashion to the atomic sample. Furthermore, EIT results from quantum interference and hence is quite sensitive to the two-photon resonance condition.

Papers:

Conference Presentations:


