The specific aim of this project is to produce a Bose-Einstein Condensate (BEC) of rovibronic ground state molecules; i.e. molecules in the lowest vibrational and rotational level of the electronic molecular ground state. This BEC will serve as an ideal starting point for investigation of molecular collisional processes in the zero-temperature limit and for the study of fully coherent chemical processes. This project is intended to bridge the boundaries between atomic, molecular and condensed matter physics with strong linking to the newly emerging field of ultracold chemistry. This goal to produce a rovibronic ground state molecular BEC is a prime example of the broader direction of the research pursued through this endeavor, that of investigating the fundamental physics of the quantum mechanical interactions of matter. Application of this research can be found in precision measurements of fundamental physical constants and preparation of new many-body states which can be used as model systems for studying condensed matter physics. The goal of production of a molecular BEC also requires the joining of many techniques used in cutting-edge atomic physics research, such as atomic quantum gas production, the manipulation of zero-temperature many-body states within the potentials formed by optical lattices, precision spectroscopic and laser control techniques, and the characterization of few-body interactions. The application of these experimental techniques offers the ability to study several physical phenomena. Thus, the multiple results of this project have been the realization of the rovibronic ground state molecular system near quantum degeneracy, the characterization of resonant few body physics within confined geometries and the use of this to realize exotic states of matter and novel types of phase transitions of zero temperature gases, and the characterization of coherent matter-wave physics within the periodic potentials of optical lattices.

We proposed a specific route to create a BEC of rovibronic ground state molecules which is shown schematically in Figure 1. Atoms, initially prepared in a BEC, are associated to weakly bound molecules on a Feshbach resonance, and then adiabatically transferred to the rovibrational ground state of the lowest electronic molecular potential using two stages of coherent optical two-photon transfer. Implementation of this scheme requires precise spectroscopic information on the transitions driven by lasers L1-L4 (see Fig. 1). Previous to this work calculations were available to guide the search for the transitions but the precise knowledge of the frequencies and transition strengths of the lines was not known. Therefore, we began by performing the spectroscopy necessary to identify several possibilities for the transfer driven by laser L1 [1]. With this information we identified the suitable lines for states coupled by laser L2 and realized transfer to deeply lying rovibrational states
To effect the transfer we employed the stimulated Raman adiabatic passage (STIRAP) technique. We demonstrated that we could achieve high transfer efficiencies, greater than 80%, and that the transfer did not heat the original sample. This provided direct evidence that our scheme preserved the quantum character of the original sample and was a significant milestone in demonstrating that the creation of molecular BEC in the rovibronic ground-state via this method could be possible.

The next steps then were to identify the transitions driven by lasers L3 and L4 [3] and to implement the second STRIRAP transfer to the ground state. With this accomplished we added an optical lattice to this scheme. The optical lattice is crucial for this plan as molecules in excited rovibrational states of the electronic ground-state potential undergo inelastic collisions and are immediately lost.

FIG. 1. Molecular level scheme for Cs$_2$ illustrating the optical transfer scheme to realize a BEC of rovibronic ground state molecules. Molecules in a weakly bound Feshbach level, vibrational level $v = 155$ (not resolved near the 6S+6S asymptote), are transferred to the rovibrational level $v = 73$, $J = 2$ ($J$ is the rotational quantum number) of the singlet $X^1\Sigma_g^+$ ground-state potential with a binding energy of 1061 cm$^{-1}$ adiabatically, using a coherent two-photon process involving lasers L1 and L2 near 1126 nm and 1006 nm respectively. The following two-photon transition to the lowest vibrational and rotational level, $v = 0$, $J = 0$, is then driven by lasers L3 and L4 near 1350 nm and 1000 nm, respectively. The first optical transition has L1 and L2 coupled to the $v' = 225$ level of the electronically excited $0_u^+$ system, and the second transfer has L3 and L4 coupled to the $v' = 61$ level of the excited $0_u^+$ system. The position of the vertical arrows is not meant to reflect the internuclear distance at which the transition takes place.
from the sample. We can initialize the lattice with one molecule at each lattice site and thereby shield the molecules from one another during the transfer process. Once the molecules are in the rovibronic ground-state they can be released from the lattice and the collisional properties studied. The optical lattice could impede the transfer however by causing optical excitation or creating unwanted coherences. So with the lattice implemented we demonstrated the first STIRAP transfer and measured the confining potential of the lattice on the weakly bound Feshbach molecules and on the deeply bound molecules in $v = 73$, $J = 2$ of the ground-state potential [4]. Here $v$ is the vibrational quantum number and $J$ is the rotational quantum number. Next, we realized a sample of rovibronic ground-state molecules within the optical lattice [5]. The transfer efficiency from the original very weakly bound molecules to the ground-state molecules is above 50% using either the two sequential STIRAP transfers or one coherent 4-photon transfer. The molecules are trapped in the motional ground-state of each well of the optical lattice and we can selectively populate a specific hyperfine state of the ground-state molecules. Thus a BEC of these molecules is within reach. Furthermore our techniques can be generalized to the case of heteronuclear molecules which opens up the study of dipolar quantum gases within optical lattices.

Given the availability of the optical lattice and an atomic BEC with tunable interactions we also performed studies on the interactions of gases within confined geometries. The lattice can be employed in only two dimensions such that the motion of the atoms is frozen out in these two dimensions and free in one dimension. This creates an effective one-dimensional (1D) system. In a 1D situation the interplay of interactions and confinement amplifies the role of quantum fluctuations and correlations. A remarkable example of this is the so called Tonks-Girardeau gas where the atomic interaction or confinement is strong enough such that bosons minimize interaction energy by avoiding spatial overlap and acquire the properties of noninteracting fermions. We generated this situation and extended it by increasing interactions to such a degree that in the confined geometry they became resonant. In this way we created a super-Tonks-Girardeau (sTG) gas [6]. In this gas the bosons acquire the properties of a gas of interacting fermions. This is a highly-excited and correlated many-body system which nonetheless is metastable. This is of particular interest as it is a novel example of a many-body system which can be used as a model system for studies of highly excited, highly correlated condensed matter systems.

In addition to the realization of the TG gas we investigated the properties of phase transitions of a 1D system of bosons as it becomes fermionized. The phase transition we investigated was the transition from a superfluid to a Mott insulator which can be effected within a three dimensional optical lattice.
Generally, within the context of ultracold atoms, this transition is driven by tuning the ratio of the interaction energy to the kinetic energy of the gas beyond a critical value by increasing the depth of the optical lattice. In 1D however, the increased correlations resulting from the confinement enhance the granularity of the gas and the transition can be driven by an arbitrarily weak lattice which immediately pins the atoms. The description of this phase transition, in this case, is given by an exactly solvable field theoretic model, and as such is of great interest within the physics community. We observed this transition and mapped out the phase diagram between the superfluid and Mott insulator in 1D from the strongly interacting regime all the way to the weakly interacting regime [7]. This observation opens up the study of exactly solvable quantum-field model systems in the context of ultracold atoms with full control over system parameters.

A key to the observation of the pinning phase transition and realization of the sTG was the ability to effect and, in fact, to resonantly enhance interactions in the reduced dimensional geometry. Specifically, during this study we observed for the first time a so-called confinement induced resonance (CIR) [6]. We continued our investigation of atomic interactions within confined geometries and in fact observed the splitting of the CIR into two or more resonances [8] as we tuned the symmetric 1D system to an asymmetric system by gradually reducing the confinement in one direction of the lattice. We followed these resonances as the asymmetry was increased to the limit of a two dimensional system and observed the persistence of one CIR. These observations are of fundamental importance as these CIR’s will allow tunability of the underlying atom-atom interactions of these gases and will afford a host of experimental studies.

We have also investigated the dynamics of matter-waves within the periodic potentials generated by optical lattices. In these experiments we load a BEC into a lattice in only one dimension and then add an offset force. In the case of noninteracting particles this leads to a Bloch oscillation of the matter-wave. Generally particle interactions dephase the coherent momentum evolution of the matter wave and obscure the Bloch oscillation. In one experiment we used high resolution imaging of the momentum distribution of the wave-packet to show that the dephasing induced by interactions was in fact coherent [9]. We went on to show that this dephasing could be reversed. This has fundamental implications for the use of matter-waves in precision measurement experiments. We also showed that a noninteracting matter wave within an optical lattice can be coherently transported while undergoing Bloch oscillations. In this case we found that if the force applied to the sample is modulated the matter-wave will move over several hundred lattice sites. The physics underlying these dynamics is essentially a rescaling of the physics underlying Bloch oscillations and as such these giant oscillations in real space are termed super Bloch oscillations [10]. This finding could find application for instance
in engineering very large area atom interferometers which can be used for precision force sensing.

All of these results greatly extend the role of atomic physics of ultracold gases to investigate the fundamental interactions within nature. The sample of ultracold molecules provides new avenues to investigate the quantum interactions of more complex particles, in particular, to study quantum chemistry. The realization of novel many-body states and observation of novel types of quantum phase changes further extend the role of ultracold physics to investigate the physics of condensed matter systems. Finally, our findings on the dynamics of matter waves within periodic potentials greatly extends the role of BEC’s within the context of high precision interferometric measurements.