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Strongly Correlated Quantum Gases of Atoms and Dipolar Molecules

Final report W911NF1310189

PI: Martin Zwierlein

MIT-Harvard Center for Ultracold Atoms, Research Laboratory of Electronics, and Department of Physics, MIT, Cambridge, MA 02139, USA

A. Accomplishments

1. Research Objectives

In this work we established a Fermi gas of ultracold dipolar molecules of NaK as a novel platform for the discovery of novel quantum phases of matter with designed properties. With this novel species of dipolar quantum gases, we may hope to create high-temperature superfluids and quantum magnets. To this end, we harvested novel long-range and anisotropic interactions between molecules, mediated by the dipolar interaction, and by laser light between atoms. This could ultimately lead to the discovery of topological superfluids that might enable topologically protected quantum computation. Our experiments employed quantum mixtures of bosonic ^{23}Na and fermionic ^{40}K and ^6Li atoms. The observation of a high-temperature fermionic superfluid and its topological excitations, the achievement of spin-orbit coupling in a Fermi gas and the formation of fermionic molecules by the PI had previously demonstrated that these quantum mixtures are ideal for the discovery of novel many-body systems of strongly correlated fermions. With mixtures of bosons and fermions, we may realize novel forms of superfluidity mediated by the surrounding bath. Out of bosonic ^{23}Na and fermionic ^{40}K atoms we formed, in this project, the first chemically stable degenerate Fermi gas of ground-state molecules. Thanks to their large electric dipole moment, novel forms of topological p-wave superfluidity as well as high-speed quantum simulation of magnetism are within reach.

This research, proposed in 2013, led to the first realization of chemically stable, fermionic, ground-state dipolar molecules in 2015. The properties of this new molecule were beyond what we could have hoped for at the time of writing the proposal. We were able to demonstrate second-scale coherence times for the nuclear spins, and managed to demonstrate strong dipolar interactions via microwave-dressing. This technique is now used by other competing groups working on NaK (I. Bloch, Max-Planck Munich) to produce a hydrodynamic gas of dipolar molecules. Also the group by Jian-Wei Pan in China has profited enormously from our pioneering work creating NaK molecules, as they have also built competing experimental setups that demonstrated further remarkable properties, from Feshbach resonances between molecules to triatomic molecules.

This molecule is extremely promising for future research on dipolar quantum gases. We hope to acquire novel funding for this research from DoD to continue our work. Understanding and controlling fermionic systems with complex interactions is imperative if we are to harness the full potential of modern materials.

2. Major activities

2.1 Ultracold Dipolar Gas of Fermionic $^{23}\text{Na}^{40}\text{K}$ Molecules in their Absolute Ground State

Jee Woo Park, Sebastian A. Will, Martin W. Zwierlein
Phys. Rev. Lett. 114, 205302 (2015)

A major outcome of this grant was the creation of ultracold dipolar Fermi gases of NaK molecules in their absolute ground state. This was the first time a chemically stable, degenerate Fermi gas of molecules was created. We report on the creation of an ultracold dipolar gas of fermionic $^{23}\text{Na}^{40}\text{K}$ molecules in their absolute rovibrational and hyperfine ground state. Starting from weakly

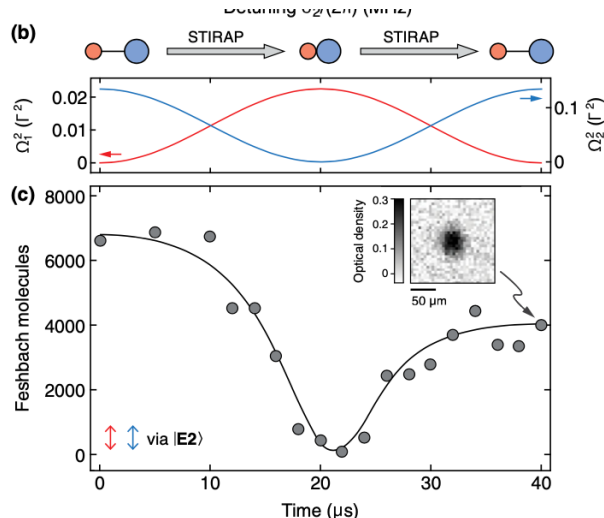


Figure 1: Creation of dipolar molecules of NaK via a STIRAP process.

bound Feshbach molecules, we demonstrate hyperfine resolved two-photon transfer into the singlet $X^1\Sigma^+|v=0, J=0\rangle$ ground state, coherently bridging a binding energy difference of 0.65 eV via stimulated rapid adiabatic passage. The spin-polarized, nearly quantum degenerate molecular gas displays a lifetime longer than 2.5 s, highlighting NaK's stability against two-body chemical reactions. A homogeneous electric field is applied to induce a dipole moment of up to 0.8 Debye. With these advances, the exploration of many-body physics with strongly dipolar Fermi gases of $^{23}\text{Na}^{40}\text{K}$ molecules is in experimental reach.

<http://dx.doi.org/10.1103/PhysRevLett.114.205302>

2.2 Two-Photon Pathway to Ultracold Ground State Molecules of $^{23}\text{Na}^{40}\text{K}$

Jee Woo Park, Sebastian A. Will, Martin W. Zwierlein
New J. Phys. 17, 075016 (2015)

To produce these gases of ultracold dipolar molecules, a sophisticated two-photon step associates weakly bound Feshbach molecules into ground-state dipolar molecules. To even find the necessary optical transitions is an extremely difficult task, akin to finding a needle in a haystack. This paper details the pathway we found. We report on high-resolution spectroscopy of ultracold fermionic NaK Feshbach molecules, and identify a two-photon pathway to the rovibrational singlet ground state via a resonantly mixed B-c intermediate state. Photoassociation in a $^{23}\text{Na}^{40}\text{K}$ atomic mixture and one-photon spectroscopy on NaK Feshbach molecules reveal about 20 vibrational levels of the electronically excited c-triplet state. Two of these levels are found to be strongly perturbed by nearby B-singlet states via spin-orbit coupling, resulting in additional lines of dominant singlet character in the perturbed complex $B^1\Pi|v=4\rangle \sim c^3\Sigma^+|v=25\rangle$, or of resonantly mixed character in $B^1\Pi|v=12\rangle \sim c^3\Sigma^+|v=35\rangle$. The dominantly singlet level is used to locate the absolute rovibrational singlet ground state $X^1\Sigma^+|v=0, J=0\rangle$ via Autler-Townes spectroscopy. We demonstrate coherent two-photon coupling via dark state spectroscopy between the predominantly triplet Feshbach molecular state and the singlet ground state. Its binding energy is measured to be 5212.0447(1) cm, a thousand-fold improvement in accuracy compared to previous determinations. In their absolute singlet ground state, NaK molecules are chemically stable under binary collisions and possess a large electric dipole moment of 2.72 Debye. Our work thus paves the way towards the creation of strongly dipolar Fermi gases of NaK molecules.

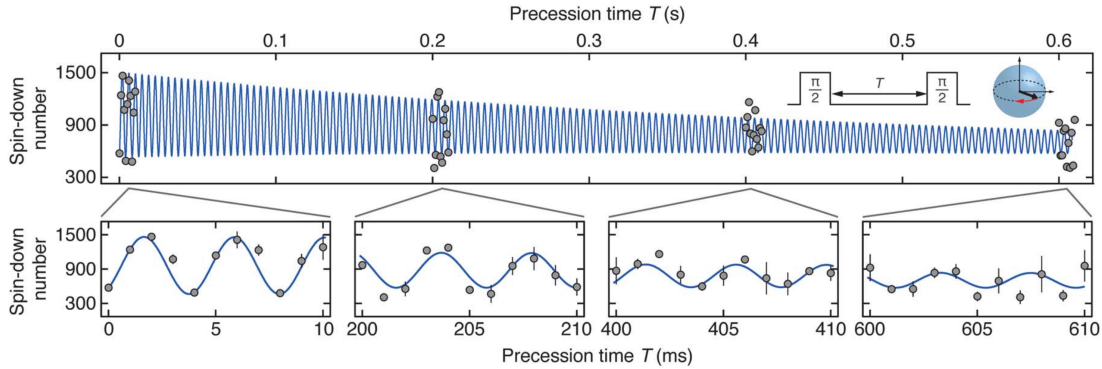


Figure 2: Second-long spin coherence times observed for the first time in a gas of ultracold molecules.

<http://dx.doi.org/10.1088/1367-2630/17/7/075016>

2.3 Coherent Microwave Control of Ultracold $^{23}\text{Na}^{40}\text{K}$ Molecules

Sebastian A. Will, Jee Woo Park, Zoe Z. Yan, Huanqian Loh, Martin W. Zwierlein
 Phys. Rev. Lett. 116, 225306 (2016)

We demonstrate coherent microwave control of rotational and hyperfine states of trapped, ultracold, and chemically stable $^{23}\text{Na}^{40}\text{K}$ molecules. Starting with all molecules in the absolute rovibrational and hyperfine ground state, we study rotational transitions in combined magnetic and electric fields and explain the rich hyperfine structure. Following the transfer of the entire molecular ensemble into a single hyperfine level of the first rotationally excited state, $J=1$, we observe collisional lifetimes of more than 3 s, comparable to those in the rovibrational ground state, $J=0$. Long-lived ensembles and full quantum state control are prerequisites for the use of ultracold molecules in quantum simulation, precision measurements and quantum information processing.

<http://dx.doi.org/10.1103/PhysRevLett.116.225306>

2.4 Second-Scale Nuclear Spin Coherence Time of Trapped Ultracold $^{23}\text{Na}^{40}\text{K}$ Molecules

Jee Woo Park, Zoe Z. Yan, Huanqian Loh, Sebastian A. Will, Martin W. Zwierlein
 Science 357, 372-375 (2017)

Coherence, the stability of the relative phase between quantum states, lies at the heart of quantum mechanics. Applications such as precision measurement, interferometry, and quantum computation are enabled by physical systems that have quantum states with robust coherence. With the creation of molecular ensembles at sub- μK temperatures, diatomic molecules have become a novel system under full quantum control. Here, we report on the observation of stable coherence between a pair of nuclear spin states of ultracold fermionic NaK molecules in the singlet rovibrational ground state. Employing microwave fields, we perform Ramsey spectroscopy and observe coherence times on the scale of one second. This work opens the door for the exploration of single molecules as a versatile quantum memory. Switchable long-range interactions between dipolar molecules can further enable two-qubit gates, allowing quantum storage and processing in the same physical system. Within the observed coherence time, 10^4 one- and two-qubit gate operations will be feasible.

<http://dx.doi.org/10.1126/science.aal5066>

2.5 Resonant dipolar collisions of ultracold molecules induced by microwave dressing

Zoe Z. Yan, Jee Woo Park, Yiqi Ni, Huanqian Loh, Sebastian Will, Tijs Karman, Martin Zwierlein
 Phys. Rev. Lett. 125, 063401 (2020)

<http://dx.doi.org/10.1103/PhysRevLett.125.063401>

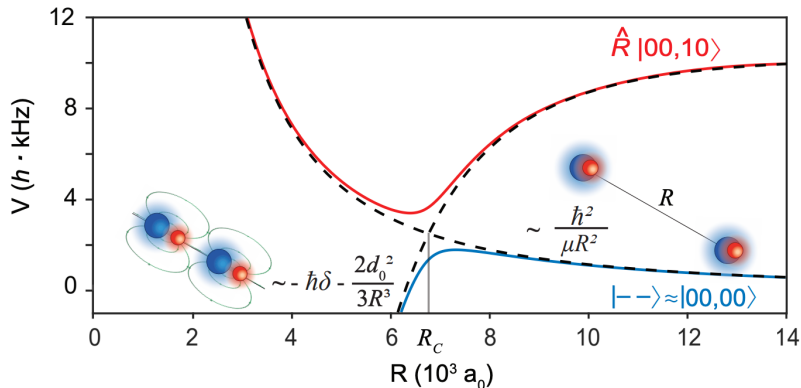


Figure 3: Resonant collision between microwave-dressed molecules.

This work demonstrated the creation of resonantly interacting dipolar molecules via a novel method, microwave dressing, which is now also widely used in other groups to manipulate molecular interactions. The work was performed during this grant period, while the analysis and understanding took significantly longer, so that this work was published only in 2020.

We demonstrate microwave dressing on ultracold, fermionic $^{23}\text{Na}^{40}\text{K}$ ground-state molecules and observe resonant dipolar collisions with cross sections exceeding three times the s -wave unitarity limit. The origin of these collisions is the resonant alignment of the approaching molecules' dipoles along the intermolecular axis, which leads to strong attraction. We explain our observations with a conceptually simple two-state picture based on the Condon approximation. Furthermore, we perform coupled-channels calculations that agree well with the experimentally observed collision rates. While collisions are observed here as laser-induced loss, microwave dressing on chemically stable molecules trapped in box potentials may enable the creation of strongly interacting dipolar gases of molecules.

2.6 Bose polarons near quantum criticality

Zoe Z. Yan, Yiqi Ni, Carsten Robens, Martin W. Zwierlein
 Science, 368, 190-194 (2020)
<http://dx.doi.org/10.1126/science.aax5850>

While the above studies focused on the creation and study of dipolar molecules, the same apparatus also enabled novel studies of strongly interacting Bose-Fermi mixtures. This particular highlight, still funded in its early stages by the grant, reports on the discovery of a novel quasi-particle, the Bose polaron.

The emergence of quasiparticles in strongly interacting matter represents one of the cornerstones of modern physics. However, when different phases of matter compete near a quantum critical point, the very existence of quasiparticles comes under question. Here we create Bose polarons

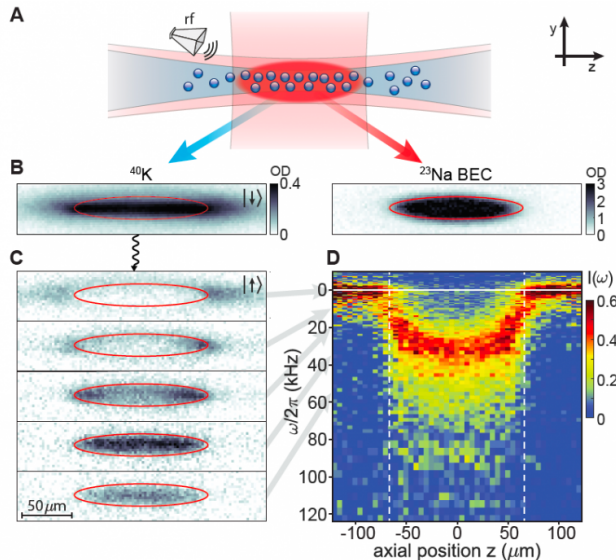


Figure 4: Observation of Bose polarons via radiofrequency spectroscopy.

near quantum criticality by immersing atomic impurities in a Bose-Einstein condensate (BEC) with near-resonant interactions. Using locally-resolved radiofrequency spectroscopy, we probe the energy, spectral width, and short-range correlations of the impurities as a function of temperature. Far below the superfluid critical temperature, the impurities form well-defined quasiparticles. However, their inverse lifetime, given by their spectral width, is observed to increase linearly with temperature at the Planckian scale $\frac{k_B T}{\hbar}$, a hallmark of quantum critical behavior. Close to the BEC critical temperature, the spectral width exceeds the binding energy of the impurities, signaling a breakdown of the quasiparticle picture.

3. Dissemination

All results were disseminated on the preprint server <https://arxiv.org/> and, for published items, appeared in *Science*, *Physical Review Letters* and other significant journals. In several public talks over the years the PI related the findings to the general public.

B. Impacts

1. Development of the principal discipline of the project

Our experiments realized fundamental models of condensed matter physics in a fully controllable environment, testing them with the precision of atomic physics, atom by atom. The experiment established the equation of state of fermions on a lattice and gave insight into the workings of strongly correlated fermionic matter. These advances are enabling theorists to test their models of strongly-interacting fermions and many-fermion systems, which is imperative if we are to harness the full potential of modern materials. The development of the spin- and density-resolved bilayer imaging represents a major step forward in the field of quantum simulators and is likely to be adapted by other groups.

2. Other disciplines

Experiments in ultracold quantum gases sometimes realize situations that cannot be realized in condensed matter systems. The most prominent example from the above results is the spin transport measurement in a Mott insulator, something that had not been achieved in condensed matter experiments.

3. Describe the impact in this reporting period on the development of human resources

Work on ultracold quantum gases of atoms and molecules trains students and postdocs in a wide variety of technical and theoretical skills relevant for the work force. On the experimental side, students and postdocs master ultrahigh vacuum technology, lasers, optics, optomechanics, high-current electromagnets, water cooling, high-voltage electric fields, electronics, computer control, ultra-high-precision laser spectroscopy, frequency standards etc. On the theoretical front, the group learns about many-body quantum theory of atoms and molecules, dipolar interactions, interactions between atoms and light, molecular spectroscopy.

Students and postdocs graduating from this experiment have gone on to prestigious jobs in industry and academia. In particular, Huanqian Loh, a female postdoc in the group, is now professor in Singapore. Another former postdoc, Sebastian Will, is professor at University of Columbia. Jee Woo Park, a former graduate student, is professor at Postech university, South Korea. Former female student Zoe Yan is doing a highly successful postdoc at Princeton University, and is on the short list of several prestigious university search committees.

My students are successful in selections for the finalists of the DAMOP thesis prize (e.g. Lawrence Cheuk) and for the Martin Deutsch Prize of Experimental Physics from MIT (Lawrence Cheuk and Thomas Hartke).

C. Honors

During the grant period, the PI was elected fellow of the American Physical Society (2016) and received the I.I. Rabi Prize of the American Physical Society (2017). He received the Thomas A. Frank Chair of Physics at MIT (2018-current) and, as a direct outcome of the grant's successes, he received a Vannevar Bush Faculty Fellowship from the Department of Defense (2019-current).

In 2021 the PI was awarded the Toptica Junior BEC award. The citation was “for his pioneering contributions to the field of ultracold quantum gases, specifically Fermi and Bose polarons, rotating condensates, spin and charge transport and the unitary Fermi gas.”

1. Describe the impact on teaching and educational experiences

I regularly use material from research in my teaching, in particular the two atomic physics courses at MIT.

D. Technical Updates

We here note useful news items written up for the public:

1. MIT team creates ultracold molecules

<https://news.mit.edu/2015/ultracold-molecules-0610>



Figure 5: MIT researchers have successfully cooled a gas of sodium potassium (NaK) molecules to a temperature of 500 nanokelvin. In this artist's illustration, the NaK molecule is represented with frozen spheres of ice merged together: the smaller sphere on the left represents a sodium atom, and the larger sphere on the right is a potassium atom. Credits: Illustration: Jose-Luis Olivares/MIT