Preparation of a Pure Molecular Quantum Gas

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An ultracold molecular quantum gas is created by application of a magnetic field sweep across a Feshbach resonance to a Bose-Einstein condensate of cesium atoms. The ability to separate the molecules from the atoms permits direct imaging of the pure molecular sample. Magnetic levitation enables study of the dynamics of the ensemble on extended time scales. We measured ultralow expansion energies in the range of a few nanokelvin for a sample of 3000 molecules. Our observations are consistent with the presence of a macroscopic molecular matter wave.

Rapid progress in controlling ultracold atomic gases, culminating in the creation of atomic Bose-Einstein condensates (BECs) and opening the door to the realm of coherent matter-wave physics (1–3), has raised the question of whether a similar level of control is possible with molecular samples. Molecules, in contrast to atoms, have a much richer internal structure and can possess permanent vector or tensor properties, such as electric dipole moments, rotational angular momentum, and even chirality. Molecule-atom and molecule-molecule interactions are at least three- and four-body processes in nature, posing new challenges to our theoretical understanding. Exquisite control over the internal and external degrees of freedom of molecules could allow the experimental study of a new coherent chemistry (4), where matter-wave interference, quantum tunneling, and bosonic stimulation dominate the dynamics and where the interaction properties can be externally controlled and engineered with electromagnetic fields. Quantum degenerate molecular gases with permanent dipole moments are also prime candidates for the precise investigation of strongly correlated quantum
systems and for the study of novel quantum phase transitions (5). Several avenues have been investigated to cool and trap molecules. Slowing of a supersonic jet of polar molecules in time-varying electric fields (6) and buffer gas loading and trapping (7) in either electrostatic or magnetic traps both permit large molecular populations with temperatures in the mK range. Alternatively, creation of molecules by photoassociation of precooled atoms has led to molecular samples with temperatures in the μK range (8). For all these techniques, however, the resulting molecular phase-space density is still many orders of magnitude away from quantum degeneracy.

Starting with a sample of ultracold atoms, controlled production of molecules can be realized by the coherent coupling of an atom pair state to a molecular state. For example, a two-photon Raman transition has successfully been applied to produce molecules within an atomic BEC (9). Similarly, the coherent nature of atomic scattering can be exploited on a Feshbach resonance to transfer colliding atoms into molecules, which has been predicted to convert an atomic BEC into a molecular BEC (10–12). A Feshbach resonance occurs when the energy of the atomic scattering state is tuned into degeneracy with that of a bound molecular state (13). Experimentally, Feshbach resonances can be induced by an external magnetic field when both states feature different Zeeman shifts. Consequently, the atom-molecule coupling can be resonantly enhanced at a particular magnetic field value, and a sweep of the field near or across the resonance can convert the atoms into molecules in a single molecular quantum state. Existence of molecules created through atomic Feshbach resonances has been reported previously in a BEC of 85Rb atoms (14), in thermal samples of 133Cs (15), and in degenerate Fermi gases of 40K (16) and 6Li (17). These studies demonstrate the quantum coherence of the Feshbach coupling (14) and the ability to detect molecules within the atomic sample by means of laser-induced (15) or radiofrequency-induced (16) dissociation. However, the resulting molecular samples could not be spatially distinguished from the atoms, nor could the molecular clouds be directly imaged and analyzed. Here, we report the observation of pure molecular quantum matter, achieved by applying a Feshbach sweep to an atomic Cs BEC (18) with immediate spatial Stern-Gerlach separation of the two species. By monitoring the evolution of the coupled-out molecular cloud, we measure ultralow kinetic expansion energies that are consistent with the presence of a coherent molecular matter wave.

The starting point of our experiment was a pure BEC of up to 6 × 10^4 Cs atoms in an optical trap (19) with a radial Thomas-Fermi radius of 8.6 μm and an axial Thomas-Fermi radius of 26.5 μm. The atoms were in the hyperfine ground state with total angular momentum F = 3 and magnetic quantum number m_F = 3. As the optical trap was by far too weak to support the atoms against gravity during the evaporative cooling process, a magnetic field gradient of 30.9 G/cm was applied to levitate the atoms (20). This levitation is very sensitive to the magnetic moment of the trapped particles, and a small levitation is sufficient to render the trap unstable. The state F = 3, m_F = 3 features a narrow Feshbach resonance near 20 G (21) with an estimated resonance width of 5 mG (22). According to an analysis of the Cs scattering properties (23, 24), the corresponding molecular state (25) has a predicted magnetic moment of μ = 0.93 μ_B, where μ_B is Bohr’s magneton, with a small magnetic field dependence (22). We produced molecules from the atomic BEC by sweeping the magnetic field across the resonance from a higher field value with a constant rate of typically 50 G/s (Fig. 1). The duration of the sweep was 3 ms. To turn off the Feshbach coupling, the field was then quickly lowered to a hold field at 17 G for a variable hold time while the optical trap was shut off (20). Because of the large magnetic field gradient along the vertical direction and the narrow resonance width of 5 mG, the Feshbach resonance occurred only within a 2-μm-thin horizontal layer. The conversion was recorded by variation of the hold time. We applied a fit to the image to determine the center position, the size of the spatial distribution, and the number of molecules. The evolution of the molecular cloud was recorded by variation of the hold time. The complete atom-molecule separation is clearly visible in absorption images (Fig. 2).

For reference, the image of a levitated BEC after 12 ms of expansion time is given in Fig. 2A. In Fig. 2B, a coupled-out molecular cloud with ~3000 molecules can be seen below the atomic BEC. The number of atoms in the remaining BEC is reduced by 50% from those shown in Fig. 2A, to ~25,000. The molecular cloud is falling, because the magnetic field gradient needed to levitate the atoms was maintained. For Fig. 2C, the magnetic field gradient was sufficient to render the trap unstable. The resulting molecular phase-space density is still many orders of magnitude away from quantum degeneracy.

![Fig. 1. Energy diagram for the atomic scattering state and the molecular bound state. The Feshbach resonance condition occurs near 20 G, where the Zeeman energy of the atomic scattering state becomes equal to that of a molecular bound state because of the difference in magnetic moments. Molecules at (2) are created from the BEC at (1) by a downward sweep of the magnetic field across the resonance. For detection, a reversed sweep brings the molecules above the dissociation limit. The inset schematically shows the molecular potential that corresponds to the open channel (lower curve) and the molecular potential that supports the bound state (upper curve). U, potential energy; r, interatomic distance.](image)

![Fig. 2. Absorption images of (A) the levitated BEC without the Feshbach sweep, (B) the levitated BEC after the Feshbach sweep with a falling molecular cloud below, and (C) the levitated molecular cloud with an upward-rising BEC above. In (B) and (C), 3000 molecules are produced at a sweep rate of 50 G/s. The separation between the atoms and the molecules is 150 μm in (B) and 240 μm in (C).](image)
raised after the Feshbach sweep in order to levitate the molecules. Hence, the atomic BEC accelerates upward and can be seen at the top of the image above the molecules. Careful adjustment of the magnetic field gradient to null the molecular acceleration allowed a precise determination of the molecular magnetic moment. We find that $\mu_\parallel = 0.930(5) \mu_B$ (20), which is in good agreement with the theoretical calculation (22).

We investigated the atom-molecule conversion as a function of the end value $B_0$ of the creation ramp. The ramp speed was kept constant at 50 G/s by variation of $B_0$ together with the duration of the ramp. We have checked that for final values of $B_0$ well above the resonance, the rapid jump over the resonance to the hold field after the end of the creation ramp did not produce any molecules. As Fig. 3 shows, molecules were created in a steplike manner. Simultaneously, the atomic population in the BEC is reduced. The transition value agrees well with the resonance position of 19.83(2) G as determined from three-body recombination loss measurements (26). From the plot of the atom number, it can be seen that up to 50% of the atoms were lost from the condensate, corresponding to ~25,000 atoms for this experiment. Hence, for a detected number of 3000 molecules, only about 24% of the lost atoms reappeared as partners in molecule formation. Also, we varied the speed of the downward magnetic field ramp across the Feshbach resonance and found that for decreasing ramp speed, the number of detected molecules saturated at a value of ~3000 molecules for speeds less than 50 G/s. The missing atoms and the saturation suggest that collisional relaxation into other molecular states occurs during the creation phase (27). After separation from the atoms, however, we did not detect any substantial loss.

We observed ultralow expansion energies for the molecular cloud in both the vertical and the horizontal directions. This was done in time-of-flight expansion measurements by variation of the hold time and hence the total expansion time. We plotted the vertical and horizontal root-mean-square (rms) widths of the recomputed atomic cloud as a function of total expansion time (Fig. 4, A and B). An apparent anisotropy of the expansion can be seen. The faster vertical expansion corresponds to a mean kinetic energy of $E_z = \frac{1}{2} k_B \times (40 \pm 3 \pm 2) \text{ nK}$ (20), where the first one-standard-deviation error is statistical and the second one is systematic. The origin of this vertical energy was identified as the velocity dispersion of the molecules during the creation phase. The dispersion was caused by the fact that the conversion zone passes through the condensate at a finite speed from below. Hence, molecules created earlier acquire a larger velocity, and those created later acquire a smaller velocity, as a result of the gravity pulling. When the size of the BEC was taken into account, the vertical expansion energy as a result of the velocity dispersion was calculated to be about $\frac{1}{2} k_B \times 30 \text{ nK}$ for the molecular cloud, largely explaining the observed energy. In fact, vertical compression of the BEC did lead to a smaller vertical energy spread. By increasing the dipole trap depth to increase the dipole trap depth to decrease the vertical extent of the BEC by a factor of 1.3, we found that the measured molecular kinetic energy was reduced in the expected way to a value of $E_z = \frac{1}{2} k_B \times (19 \pm 2 \pm 1) \text{ nK}$.

The horizontal expansion shown in Fig. 4B was unaffected by the velocity dispersion effect. However, a repulsive force due to the curvature of the levitation field acted on the molecules. This force resulted in an expansion of the cloud that follows a cosine hyperbolicus expansion dynamics ($20^\circ$), which allows the preparation of a Mott insulator phase ($31^\circ$) with exactly two atoms per lattice site. Molecules created by a subsequent Feshbach sweep will therefore be individually isolated and immune to collisional losses. After the creation of a pure molecular matter wave, one might be able to coherently transfer the molecules to low-lying molecular states by two-photon Raman transitions. Hence, a complete and coherent control over the dynamics of molecular quantum matter can be envisaged.

References and Notes
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Cooling Bose-Einstein Condensates Below 500 Picokelvin


Spin-polarized gaseous Bose-Einstein condensates were confined by a combination of gravitational and magnetic forces. The partially condensed atomic vapors were adiabatically decompressed by weakening the gravito-magnetic trap to a mean frequency of 1 hertz, then evaporatively reduced in size to 2500 atoms. This lowered the peak condensate density to $5 \times 10^{10}$ atoms per cubic centimeter and cooled the entire cloud in all three dimensions to a kinetic temperature of $450 \pm 80$ picokelvin. Such spin-polarized, dilute, and ultracold gases are important for spectroscopy, metrology, and atom optics.

The pursuit of lower temperatures is motivated by the quest to observe phenomena that occur on very low energy scales, in particular, phase transitions to new forms of matter. The achievement of temperatures near 1 K in solids and in liquids led to the discoveries of superconductivity (1) and superfluidity (2), respectively. The advent of laser cooling resulted in microkelvin temperature atomic vapors (3–5), subsequently cooled to nanokelvin and quantum degenerate Fermi gases (6,7). Collectively, these low-temperature systems have a host of applications, including superconducting quantum interference devices (SQUIDs) (9), superfluid gyroscopes (10,11), and atomic clocks (12).

Temperature is a quantity that parameterizes how energy is distributed across the available states of a system, and effective temperatures can be defined for decoupled degrees of freedom or subsets of particles. For example, nuclear spins isolated from the environment precess more slowly than the thermal component and have been characterized by picokelvin effective temperatures for anisotropic (17) and noninteracting (18) gases.

Cooling the atomic motion of entire ensembles in all three dimensions has proven difficult. To date, kinetic temperatures of a few hundred nanokelvin have been achieved with adiabatic and optical cooling (19,20), and evaporative cooling techniques have produced condensates with temperatures of 3 nK (21). By adiabatic expansion and subsequent evaporation, we have cooled partially condensed atomic vapors to picokelvin kinetic temperatures.

Our thermometry is calibrated by the Bose-Einstein condensation (BEC) phase transition temperature, $T_c$, which in the thermodynamic limit for a harmonically trapped ideal Bose gas is (22)

$$k_B T_c = \frac{\hbar \bar{a}}{2} \left(\frac{N}{\xi^3} \right)^{1/3} \approx 0.94\hbar \bar{a} N^{1/3}$$

(1)

where $k_B$ is Boltzmann’s constant, $\bar{a}$ is Planck’s constant divided by $2\pi$, $\xi(n)$ is the Riemann Zeta function, $\bar{a} = (\omega_x \omega_y \omega_z)^{-1/3}$ is the geometric mean of the harmonic trap frequencies, and $N$ is the total number of atoms, both condensed and noncondensed. Thus, the atom number and the trap frequencies set an upper limit for the temperature of a confined Bose-Einstein condensate. In our work, adiabatically weakening the trapping potential to a mean frequency of $\bar{a} = 2\pi \times (1.12 \pm 0.08)$ Hz guaranteed that partially condensed atomic vapors with $N = 8000$ atoms had picokelvin temperatures ($T_c \approx 1$ nK).

Bose-Einstein condensates containing more than $10^7 \ Na$ atoms were created in the weak field seeking $|F = 1, m_F = -1 \rangle$ state in a magnetic trap, captured in the focus of an optical tweezers laser beam, and transferred into an auxiliary “science” chamber as described in (23). In the science chamber, condensates containing $2 \times 10^6$ to $3 \times 10^7$ atoms were transferred from the optical tweezers into a gravito-magnetic trap (Fig. 1A). A small coil carrying current $I_c$ generated a vertical bias field $B_z$ and supported the condensates against gravity with a vertical magnetic field gradient, $B'_{z} = 2$ mg/