Coherent Molecular Optics using Ultra-Cold Sodium Dimers

J.R. Abo-Shaeer*, D.E. Miller, J.K. Chin, K. Xu, T. Mukaiyama, and W. Ketterle†

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

(Dated: November 13, 2018)

Coherent molecular optics is performed using two-photon Bragg scattering. Molecules were produced by sweeping an atomic Bose-Einstein condensate through a Feshbach resonance. The spectral width of the molecular Bragg resonance corresponded to an instantaneous temperature of 20 nK, indicating that atomic coherence was transferred directly to the molecules. An autocorrelating interference technique was used to observe the quadratic spatial dependence of the phase of an expanding molecular cloud. Finally, atoms initially prepared in two momentum states were observed to cross-pair with one another, forming molecules in a third momentum state. This process is analogous to sum-frequency generation in optics.

PACS numbers: PACS 03.75.Fi, 34.20.Cf, 32.80.Pj, 33.80.Ps

Similar to the field of optics, where the high intensity and coherence of lasers allowed for the observation of effects such as frequency doubling and wave-mixing, atom optics has benefited greatly from the realization of Bose-Einstein condensates (BEC). High phase-space density (atoms per mode) and a uniform phase give the condensate its laser-like qualities. Although not fundamentally required, BEC has led to the observation of such phenomena as four-wave mixing, matter wave amplification, and atom number squeezing.

The current state of molecular optics is similar to atom optics prior to the realization of BEC. Diffraction and interferometry of thermal molecular beams has been demonstrated, yet monoenergetic beams lack the density necessary to observe nonlinear effects. However, recent experiments using Feshbach resonances have demonstrated the conversion of degenerate atomic bosons and fermions into ultracold molecules. These sources have the potential to greatly advance molecular optics. Furthermore, atom-molecule coupling can be studied as the first steps towards “superchemistry”, where chemical reactions are stimulated via macroscopic occupation of a quantum state.

The coherent manipulation of atomic matter waves via stimulated light scattering has been crucial in the development of nonlinear atom optics (for a review see[23]). Here we demonstrate the ability to apply Kapitza-Dirac and Bragg diffraction to cold molecules. Using optical standing waves of suitably chosen frequencies, sodium dimers were coherently manipulated with negligible heating or other incoherent processes. First, we characterized the coherence of our “source” molecules, created via Feshbach resonance. By measuring the Bragg spectrum of the molecules immediately after their creation, the conversion from atoms to molecules was shown to be coherent - the matter wave analog to frequency doubling in optics. The quadratic spatial dependence of the phase of the expanding molecules was observed using an autocorrelation interference technique. By creating a duplicate sample of molecules and overlapping it with the original, matter wave interference was observed. Finally,
the matter wave analog to sum-frequency generation was demonstrated. Atoms prepared in two momentum states, prior to creating molecules, were observed to cross-pair, generating a third momentum state.

To produce molecules, sodium condensates were prepared in a crossed optical dipole trap in the state \( |F, m_F\rangle = |1, -1\rangle \) state. Trap frequencies of \( \omega_x, \omega_y, \omega_z \) were \( 2\pi \times (146, 105, 23) \) Hz yielding a typical peak density of \( n_0 \simeq 4.3 \times 10^{14} \text{ cm}^{-3} \) for 25 million atoms. Atoms were then spin-flipped to the \(|1,1\rangle\) state, in which a 1 G wide Feshbach resonance exists at 907 G.

The magnetic field sequence used to create and detect Na$_2$ molecules was identical to our previous work \[15, 26\]. Briefly, the axial (z-axis) magnetic field was ramped to 903 G in 100 ms. In order to prepare the condensate on the negative scattering length side of the resonance, the field was stepped up to 913 G as quickly as possible (\( \sim 1 \mu s \)) to jump through the resonance with minimal atom loss. After waiting 1200 µs for the transient field to die down, the field was ramped back down to 903 G in 50 µs to form molecules. In order to remove non-paired atoms from the trap, the sample was irradiated with a 10 µs pulse of resonant light. Because 903 G is far from the Feshbach resonance, the magnetic field mixed between atomic and molecular states was small, and therefore molecules were transparent to this “blast” pulse. By ramping the field back to 913 G, molecules were converted back to atoms. Absorption images were taken at high fields (either at 903 G or 913 G), with the imaging light incident along the axial direction of the condensate. Bragg scattering of atoms and molecules was carried out using two nearly orthogonal beams (\( \theta_B = 84^\circ \)), aligned such that particles were scattered along the x-axis of the trap. The beams were far-detuned from atomic/molecular transitions to limit spontaneous scattering. For atoms the detuning was \( \simeq 4 \) nm from the sodium D lines. To find a suitable transition for the molecules, we scanned the laser wavelength and measured the Rabi frequency for Bragg transitions. Several narrow transitions were observed, but not carefully characterized. For this work the laser was set to 590.159 nm and the detuning \( \sim 50 \) MHz from the apparent resonance, yielding a Rabi frequency of 2 kHz.

Fig 1a shows time-of-flight images of Bragg scattering for atoms and molecules. Because the kinetic energy of the scattered particles was much larger than their mean-field energy, individual momentum states were well-resolved in ballistic expansion. Both atoms and molecules receive equal two-photon recoil momentum, \( p_r = 2h \sin(\theta_B/2)/\lambda_L \), where \( \lambda_L \) is the wavelength of the Bragg beams. However, scattered molecules move away from the central peak with half the velocity of atoms, owing to their doubled mass. Fig 1b-d show Kapitza-Dirac scattering, where multiple atomic and molecular momentum states were populated due to the broad frequency distribution of the short pulse (\( 5 \mu s \)).

In order to study the coherence properties of the sample, Bragg spectra \[b\] were taken with \( \sim 1 \) kHz resolution by pulsing on the two laser beams (250 µs square pulse) before releasing the particles from the trap. For noninteracting particles, the Bragg resonance occurs at a relative detuning of \( \nu_0 = \pm p_r^2/(2m\hbar) \) between the beams, where the sign of \( \nu_0 \) dictates the direction of out-coupling. Interactions in a condensate give rise to a mean-field shift of the resonance \( \delta \nu = 4n_0U/7\hbar \), where \( U = 4\pi \hbar^2 a/m \) and \( a \) is the scattering length. Fig 2 shows three spectra for (a) atoms above the Feshbach resonance, as well as (b) atoms and (c) molecules after sweeping through the resonance. The reduced mean-field shift for atoms below the resonance (Fig 1b) can be attributed to inelastic losses caused by passing through the resonance. Atoms below the resonance coexisted with a small fraction of molecules (2%). The peak output for each set of data is normalized to 1. The actual peak out-coupled fractions were 0.06 for the atoms and 0.5 for the molecules. The atomic signal was kept intentionally low to minimize collisions, which make the data analysis difficult (see halos in Fig 1c). As expected from the resonance condition, molecular resonances occur at half the frequency of atomic resonances.

Two mechanisms contribute to the fundamental width of the Bragg resonance \[a, 27\]. For a parabolic density distribution, the finite size of the sample yields a momentum spread

\[
\Delta \nu_p = 1.58 \frac{p_r}{2\pi m x_0},
\]

where \( x_0 \) is the Thomas-Fermi radius. In addition, the inhomogeneous density distribution of the sample produces a spread in mean-field energy

\[
\Delta \nu_m = \sqrt{8 \frac{n_0 U}{147}} \frac{1}{\hbar},
\]

FIG. 2: Bragg spectra for atoms and molecules. In (a) the spectrum is taken for a pure atomic sample above the Feshbach resonance (913 G). (b) and (c) are spectra of atoms and molecules, respectively, below the resonance (903 G). In (c) atoms were removed from the trap with resonant light to limit losses due to atom-molecule collisions. The Bragg resonance condition for molecules occurs at half the frequency of the atomic resonance.
The fundamental width is approximately given by the quadrature sum of these two broadening mechanisms
\[ \Delta \nu = \sqrt{\Delta \nu_n^2 + \Delta \nu_p^2}. \]

The fundamental width, \( \Delta \nu \), and measured rms width, \( \sigma \), are compared for each case in Table I. \( n_0 \) and \( x_0 \) were determined from the size of the condensate in time-of-flight. The measured widths cannot be accounted for by fundamental broadening mechanisms alone. For atoms above resonance, the fundamental width is \( \Delta \nu = 1.39 \text{ kHz} \), compared to the measured value of 4.46 kHz. Therefore, another broadening mechanism must contribute \( \sim 4 \text{ kHz} \) to the width. Most likely this is due to Doppler broadening caused by random center-of-mass motion and other collective excitations of the condensate. This was investigated by mixing two frequencies into each Bragg beam to out-couple particles in both \( \pm x \) directions. For particles at rest, the out-coupling should always be symmetric. However, we observe the ratio of out-coupled particles in either direction to fluctuate. In addition we measure a small, consistent shift in the Bragg spectrum, indicating a drift velocity. A line broadening of 4 kHz corresponds to a velocity of \( \sim 2 \text{ mm/s} \), or a vibrational amplitude of \( \sim 2 \mu \text{m} \) (compared to \( x_0 = 13 \mu \text{m} \)). This is not unreasonable, because the field ramping scheme used to bring the atoms to high field is violent and may induce collective excitations such as breathing modes.

- **Atoms (Above)**: \( \Delta \nu = 1.39 \text{ kHz}, \sigma = 4.46(17) \text{ kHz}, T = 10 \text{ nK} \)
- **Atoms (Below)**: \( \Delta \nu = 1.03 \text{ kHz}, \sigma = 4.50(60) \text{ kHz}, T = 10 \text{ nK} \)
- **Molecules**: \( \Delta \nu = 0.36 \text{ kHz}, \sigma = 4.53(14) \text{ kHz}, T = 20 \text{ nK} \)

| Spectrum   | \( \Delta \nu \) (kHz) | \( \sigma \) (kHz) | T (nK) |
|------------|------------------------|--------------------|--------|
| Atoms (Above) | 1.39                   | 4.46(17)          | 10     |
| Atoms (Below) | 1.03                   | 4.50(60)          | 10     |
| Molecules  | 0.36 \( \dagger \)     | 4.53(14)          | 20     |

**TABLE I**: Fundamental width (\( \Delta \nu \)), measured rms width (\( \sigma \)), and the corresponding temperature (T), assuming a thermal distribution.

\( \dagger \)This lower bound assumes that the molecules have the same spatial profile as the atoms, which our results indicate is not the case.

Despite vibrational noise making the dominant contribution to the width of the spectra, the measured values are still narrow enough to indicate quantum degeneracy. For a given \( \sigma \), the corresponding temperature for a thermal distribution of particles is

\[ T = \frac{m \hbar^2 \sigma^2}{k_B p_f^2}. \]  

Thus, for an rms width of 4.5 kHz, the temperature for a thermal distribution of molecules would be 20 nK, comparable to a previous value obtained using a time-of-flight technique \[32]. The BEC transition temperature for our trap parameters \[28\] and \( 5 \times 10^4 \) molecules is much higher (115 nK). This demonstrates a deeply degenerate, purely molecular sample, where as previous experiments have demonstrated coherent admixture of molecular character into an atomic BEC \[22\].

The molecular Bragg Spectrum showed a surprisingly large shift of \( \delta \sigma = 625 \text{ Hz} \). If interpreted as a mean-field shift, this would imply either a very high density (possible due to a spatial collapse) or an anomalously large molecular scattering length outside the Feshbach resonance. Further study is necessary.

The spatial phase of the expanding molecular cloud was directly imaged using an autocorrelation method \[30\], which gives rise to the self-interference of the molecular sample (see Fig 3). To accomplish this, two identical copies of the sample were made using a short Kapitza-Dirac pulse (10 \( \mu \text{s} \)), applied after 2 ms of ballistic expansion. The copies, with momentum \( \pm p_r \), moved away from the zero momentum peak for time \( \Delta t \) before an identical pulse recombined them with the original. This type of interferometer has three readout ports, with momenta 0, \( \pm p_r \). The straight-line interference fringes are characteristic of a quadratic spatial phase. We observe fringe contrast as high as 50% and a fringe spacing consistent with \( \lambda_f = \hbar/\mu d \) \[31\], where \( d = p_r \Delta t/m \) is the distance the copies moved between pulses \[32\]. Interference fringes can only be resolved for small \( d \). Therefore, this method cannot be used to observe coherence lengths longer than those inferred from Bragg spectroscopy. It should be noted that the appearance of interference fringes does not imply that the sample is condensed. Rather, it demonstrates only that the coherence length in time-of-flight is longer than the separation \( d \). Therefore, similar interference can also be observed for a cloud of thermal atoms \[32, 54\].

The conversion of atoms to molecules may be viewed as the atom optic equivalent of frequency doubling \[33, 34\]. The relevant Hamiltonian for the atom-molecule coupling has the same form as that for the optical frequency doubling
In conclusion, we have demonstrated coherent molecular optics using standing light waves. The ability to coherently convert atoms into molecules makes molecular optics even richer than atom optics. In addition, the techniques demonstrated in this paper could prove useful for probing molecules formed in quantum-degenerate fermi systems, and possibly even Cooper pairs.

The authors would like to acknowledge M. Boyd and W. Setiawan for experimental contributions and thank A.E. Leanhardt and M. W. Zwierlein for critical readings of the manuscript. This research is supported by NSF, ONR, ARO, and NASA.

1. J. Stenger et al., Phys. Rev. Lett. 82, 4569 (1999).
2. E. W. Hagley et al., Phys. Rev. Lett. 83, 3112 (1999).
3. M. G. Moore and P. Meystre, Phys. Rev. Lett. 86, 4199 (2001).
4. W. Ketterle and S. Inouye, Phys. Rev. Lett. 86, 4203 (2001).
5. L. Deng et al., Nature 398, 218 (1999).
6. S. Inouye et al., Nature 402, 641 (1999).
7. M. Kozuma et al., Science 286, 2309 (1999).
8. C. Orzel et al., Science 291, 2386 (2001).
9. C. Bordé et al., Phys. Lett. A 188, 187 (1994).
10. M. S. Chapman et al., Phys. Rev. Lett. 74, 4783 (1995).
11. L. Hackermüller et al., Phys. Rev. Lett. 91, 090408 (2003).
12. H. Sakai et al., Phys. Lett. A 57, 27942801 (1998).
13. S. Dürr et al., Phys. Rev. Lett. 92, 020406 (2003).
14. J. Herbig et al., Science 301, 1510 (2003).
15. K. Xu et al., Phys. Rev. Lett. 91, 210402 (2003).
16. C. A. Regal et al., Nature 424, 47 (2003).
17. S. Jochim et al., Science 302, 2101 (2003).
18. M. W. Zwierlein et al., Phys. Rev. Lett. 91, 250401 (2003).
19. J. Cubizolles et al., Phys. Rev. Lett. 91, 240401 (2003).
20. K. E. Strecker et al., Phys. Rev. Lett. 91, 080406 (2003).
21. D. J. Heinzen et al., Phys. Rev. Lett. 84, 5029 (2000).
22. A. P. Chikkatur et al., Phys. Rev. Lett. 85, 483 (2000).
23. S. L. Rolston and W. D. Phillips, Nature Insight 416, 219 (2002).
24. M. Kozuma et al., Phys. Rev. Lett. 82, 871 (1999).
25. S. Inouye et al., Nature 392, 151 (1998).
26. T. Mukaiyama et al., Phys. Rev. Lett. 92, 180402 (2004).
27. J. Stenger et al., Phys. Rev. Lett. 84, 2283 (2000), erratum.
28. For an optical dipole trap using 1064 nm light, the trap parameters for both atoms and molecules are the same.
29. E. A. Donley et al., Nature 417, 529 (2002).
30. J. Simsarian et al., Phys. Rev. Lett. 85, 2040 (2000).
31. M. R. Andrews et al., Science 275, 637 (1997).
32. Strictly speaking, this analysis only applies to the outcoupled ports. The center port is comprised of three copies.
33. I. Bloch et al., Nature 403, 166 (2000).
34. D. E. Miller et al., in Preparation (2004).
35. B. Saleh and M. Teich, Fundamentals of Photonics (Wiley, New York, 1991).
36. V. Vuletić et al., Phys. Rev. Lett. 83, 943 (1999).