

Coherent Molecular Optics Using Ultracold 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, Massachusetts 02139, USA*

(Received 13 September 2004; published 3 February 2005)

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.

DOI: 10.1103/PhysRevLett.94.040405

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

Similar to the field of optics, where the high intensity and coherence of lasers allow for the observation of effects such as frequency doubling and wave mixing, atom optics has benefited greatly from the realization of Bose-Einstein condensates (BECs). High phase-space density (atoms per mode) and a uniform phase [1,2] give the condensate its laserlike qualities. Although not fundamentally required [3,4], BECs have led to the observation of such phenomena as four-wave mixing [5], matter wave amplification [6,7], and atom number squeezing [8].

The current state of molecular optics is similar to atom optics prior to the realization of BECs. Diffraction and interferometry of thermal molecular beams has been demonstrated [9–12], 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 [13–15] and fermions [16–20] 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 [21].

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 [22]). Here we demonstrate the ability to apply Kapitza-Dirac and Bragg diffraction [23] 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 $|F, m_F\rangle = |1, -1\rangle$ state. Trap frequencies of $(\omega_x, \omega_y, \omega_z) = 2\pi \times (146\ 105\ 23)$ Hz yielded a typical peak density of $n_0 \simeq 4.3 \times 10^{14}$ cm⁻³ for 25×10^6 atoms. Atoms were then spin-flipped to the $|1, 1\rangle$ state, in which a 1 G wide Feshbach resonance exists at 907 G [24].

The magnetic field sequence used to create and detect Na₂ molecules was identical to our previous work [15,25]. 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 (~ 1 μ s) to jump through the resonance with minimal atom loss. After waiting 1200 μ s for the transient field to die down, the field was ramped back down to 903 G in 50 μ s to form molecules. In order to remove nonpaired atoms from the trap, the sample was irradiated with a 10 μ s pulse of resonant light. Because 903 G is far from the Feshbach resonance, the mixing 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 detuned ~ 50 MHz from the apparent resonance, yielding a Rabi frequency of 2 kHz.

Figure 1 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 λ_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. Figures 1(c) and 1(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 μ s).

In order to study the coherence properties of the sample, Bragg spectra [1] were taken with ~ 1 kHz resolution by pulsing on the two laser beams (250 μ s square pulse) before releasing the particles from the trap. For noninteracting particles, the Bragg resonance occurs at a relative detuning of $\nu_0 = \pm p_r^2/2mh$ between the beams, where the sign of ν_0 dictates the direction of outcoupling. Interactions in a condensate give rise to a mean-field shift of the resonance $\delta\nu = 4n_0U/7h$, where $U = 4\pi\hbar^2 a/m$ and a is the scattering length. Figure 2 shows three spectra for (a) atoms above the Feshbach resonance, as well as (b) atoms and (c) molecules after sweeping through the reso-

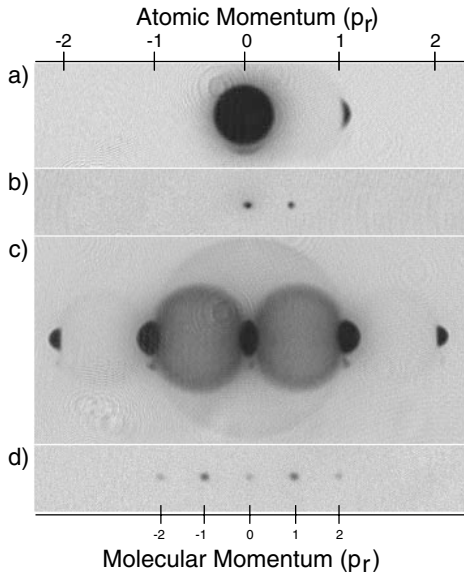


FIG. 1. Bragg diffraction of (a) atoms and (b) molecules. Scattered particles recoil with identical momenta, p_r . However, during ballistic expansion, diffracted molecules expand with half the velocity of atoms, due to their doubled mass. The pulse duration in each image was 200 μ s. A shorter pulse (5 μ s) populated multiple (c) atomic and (d) molecular momentum states. The halos in (c) are due to collisions between different momentum states [36]. The time of flight in each image is 17 ms.

nance. The reduced mean-field shift for atoms below the resonance [Fig. 1(b)] 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 outcoupled 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. 1(c)]. 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 [1,26]. For a parabolic density distribution, the finite size of the sample yields a momentum spread

$$\Delta\nu_p = 1.58 \frac{p_r}{2\pi m x_0}, \quad (1)$$

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

$$\Delta\nu_n = \sqrt{\frac{8}{147}} \frac{n_0 U}{h}. \quad (2)$$

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, $\bar{\sigma}$, 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 $\Delta\nu = 1.39$ kHz, compared to the measured value of

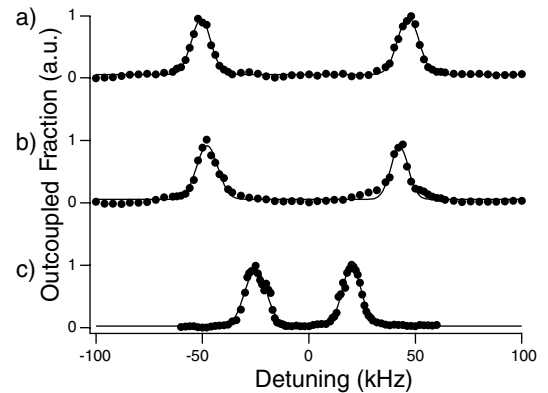


FIG. 2. Bragg spectra for atoms and molecules. In (a) the spectrum is taken for a pure atomic sample above the Feshbach resonance (913 G). (b) and (c) are spectra of atoms and molecules, respectively, below the resonance (903 G). In (c) atoms were removed from the trap with resonant light to limit losses due to atom-molecule collisions. The Bragg resonance condition for molecules occurs at half the frequency of the atomic resonance.

4.46 kHz. Therefore, another broadening mechanism must contribute ~ 4 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 outcouple particles in both $\pm x$ directions. For particles at rest, the outcoupling should always be symmetric. However, we observe the ratio of outcoupled 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 ≈ 2 mm/s, or a vibrational amplitude of $\approx 2 \mu\text{m}$ (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.

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 $\bar{\sigma}$, the corresponding temperature for a thermal distribution of particles is

$$T = \frac{mh^2\bar{\sigma}^2}{k_B p_r^2}. \quad (3)$$

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 [15]. The BEC transition temperature for our trap parameters [27] and 5×10^4 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 [28].

The molecular Bragg spectrum showed a surprisingly large shift of $\delta\bar{\nu} = 625$ 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 [29], 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 μs), applied after 2 ms of ballistic expansion. The copies, with momentum $\pm p_r$, moved away from the zero momentum peak for time Δt before an identical pulse recombined them with the original. This type of interferometer has three readout ports, with momenta 0, $\pm p_r$. The straightline interference fringes are characteristic of a quadratic spatial phase. We observe fringe contrast as high as 50% and a fringe spacing consistent with $\lambda_f = ht/md$ [30], where $d = p_r\Delta t/m$ is the distance the copies moved between pulses [31]. 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 [32,33].

The conversion of atoms to molecules may be viewed as the atom optic equivalent of frequency doubling [34]. The relevant Hamiltonian for the atom-molecule coupling has the same form as that for the optical frequency doubling process:

$$a_{2m}^\dagger a_m a_m, \quad (4)$$

where a_m is the annihilation operator for the atomic field and a_{2m}^\dagger is the creation operator for the molecular field. The measurement of the Bragg spectrum shows that the sharp “linewidth” of the seed (atom) laser is inherited by the molecular laser. In nonlinear optics, photon interactions are

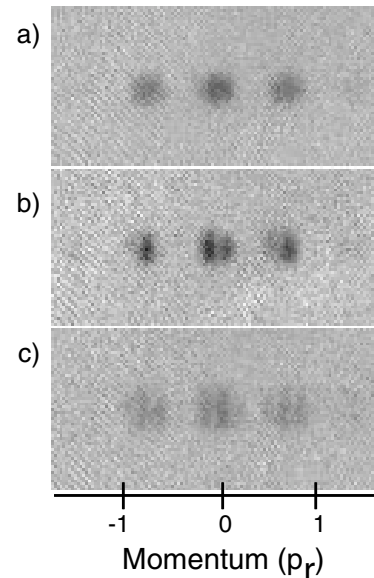


FIG. 3. Matter wave interference of molecules. (a) The molecular sample is split with a short Kapitza-Dirac pulse, creating two identical copies with momenta $\pm p_r$. (b),(c) After the copies have separated for time $\Delta t = 100 \mu\text{s}$, a second pulse recombines them, giving rise to interference in each momentum component. The time of flight in each image is 12 ms.

TABLE I. Fundamental width ($\Delta\nu$), measured rms width ($\bar{\sigma}$), and the corresponding temperature (T), assuming a thermal distribution.

Spectrum	$\Delta\nu$ (kHz)	$\bar{\sigma}$ (kHz)	T (nK)
Atoms (above)	1.39	4.46(17)	10
Atoms (below)	1.03	4.50(60)	10
Molecules	0.36 ^a	4.53(14)	20

^aThis lower bound assumes that the molecules have the same spatial profile as the atoms, which our results indicate is not the case.

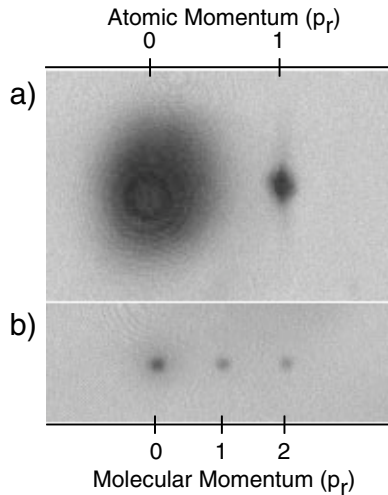


FIG. 4. Sum-frequency generation of atomic matter waves. (a) Atoms were initially prepared in momentum states 0, 1. (b) By sweeping through the Feshbach resonance, atoms combine to form molecules with momenta 0, 1, and 2. Momentum state 1 is the sum frequency of the two atomic matter waves. The “nonlinear medium” is provided by the atomic interactions. The time of flight in each image is 17 ms.

typically mediated by a refractive medium. Here, the nonlinearity arises from the atoms themselves, in the form of s -wave interactions. The high density, or “brightness,” of the source, together with the enhanced interactions at the Feshbach resonance provide the means to combine two matter waves. By combining two disparate matter waves, rather than identical ones, we extend the analogy of frequency doubling to the more general process of sum-frequency generation. To do this, atoms were initially prepared in momentum states 0, 1 (in units of p_r). By sweeping through the resonance, molecules were produced in three momentum states: 0, 1, and 2 (see Fig. 4). States 0 and 2 are simply the frequency doubled components of the two initial matter waves. State 1, however, results from cross pairing between the initial momentum states and is thus their sum frequency. This is the first time that a Feshbach resonance was observed between atoms colliding with a controlled nonvanishing momentum. The Feshbach resonance should be slightly shifted compared to the resonance for atoms at rest, which reflects the same physics encountered in the temperature dependence of the position of the resonance [35].

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 Letter 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.

*Present address: Lawrence Berkeley National Laboratories, MS88R0192, Berkeley, CA 94720-8101, USA.

Email address: jrabo-shaer@lbl.gov

†Electronic address: http://cua.mit.edu/ketterle_group

- [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 (London) **398**, 218 (1999).
- [6] S. Inouye *et al.*, Nature (London) **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**, 2794 (1998).
- [13] S. Dürr, T. Volz, A. Marte, and G. Rempe, 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, C. Ticknor, J. L. Bohn, and D. S. Jin, Nature (London) **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, G. B. Partridge, and R. G. Hulet, Phys. Rev. Lett. **91**, 080406 (2003).
- [21] D. J. Heinzen, R. Wynar, P. D. Drummond, and K. V. Kheruntsyan, Phys. Rev. Lett. **84**, 5029 (2000).
- [22] S. L. Rolston and W. D. Phillips, Nature (London) **416**, 219 (2002).
- [23] M. Kozuma *et al.*, Phys. Rev. Lett. **82**, 871 (1999).
- [24] S. Inouye *et al.*, Nature (London) **392**, 151 (1998).
- [25] T. Mukaiyama *et al.*, Phys. Rev. Lett. **92**, 180402 (2004).
- [26] J. Stenger *et al.*, Phys. Rev. Lett. **84**, 2283(E) (2000).
- [27] For an optical dipole trap using 1064 nm light, the trap parameters for both atoms and molecules are the same.
- [28] E. A. Donley, N. R. Claussen, S. T. Thompson, and C. E. Wieman, Nature (London) **417**, 529 (2002).
- [29] J. Simsarian *et al.*, Phys. Rev. Lett. **85**, 2040 (2000).
- [30] M. R. Andrews *et al.*, Science **275**, 637 (1997).
- [31] Strictly speaking, this analysis applies only to the out-coupled ports. The center port is comprised of three copies.
- [32] I. Bloch, T. W. Hänsch, and T. Esslinger, Nature (London) **403**, 166 (2000).
- [33] D. Miller *et al.*, cond-mat/0412672.
- [34] B. Saleh and M. Teich, *Fundamentals of Photonics* (Wiley, New York, 1991).
- [35] V. Vuletić, C. Chin, A. J. Kerman, and S. Chu, Phys. Rev. Lett. **83**, 943 (1999).
- [36] A. P. Chikkatur *et al.*, Phys. Rev. Lett. **85**, 483 (2000).