

## Optics and Interferometry with Na<sub>2</sub> Molecules

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We have produced an intense, pure beam of sodium molecules (Na<sub>2</sub>) by using light forces to separate the atomic and molecular species in a seeded supersonic beam. We used diffraction from a microfabricated grating to study the atomic and molecular sodium in the beam. Using three of these gratings, we constructed a molecule interferometer with fully separated beams and high contrast fringes. We measured both the real and imaginary parts of the index of refraction of neon gas for Na<sub>2</sub> molecule de Broglie waves by inserting a gas cell in one arm of the interferometer.

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Quantum mechanical treatment of the center-of-mass motion of increasingly complex systems is an important theme in modern physics. This issue is manifest theoretically in studies of the transition from quantum through mesoscopic to classical regimes and experimentally in efforts to coherently control and manipulate the external spatial coordinates of complex systems (e.g., matter wave optics and interferometry). Recently, matter wave optics and interferometry have been extended to atoms with the many techniques for the coherent manipulation of the external degrees of freedom of atoms constituting a new field called atom optics [1]. The present work extends and develops techniques of atom optics into the domain of molecules—systems characterized by many degenerate and nondegenerate internal quantum states. Whereas internal state coherences in complex molecules have long been cleverly manipulated in spectroscopy in both the radio [2] and optical frequency domains [3,4], here we coherently manipulate exclusively the center-of-mass motion [5]. This work, which culminates in the use of a molecule interferometer with spatially separated beams to determine hitherto unmeasurable molecular properties, demonstrates the applicability of molecular interferometers to precision measurements in molecular physics, some of which may have applications to fundamental physics experiments using specific molecules [6].

Our experiment combines several techniques from atom optics to make and use a molecule interferometer whose paths are well separated in position and momentum. Using an incident supersonic beam containing both Na atoms and dimers (Na<sub>2</sub>), we apply resonant light forces to selectively remove the Na atoms, leaving a pure Na<sub>2</sub> beam. We then use nanofabricated diffraction gratings, first to study the characteristics of the molecular beam and subsequently as coherent beam splitters to make a molecule interferometer with high contrast fringes. Finally, we insert a gas cell in one path of the separated beam interferometer and measure the complex index of refraction for Na<sub>2</sub> de Broglie waves in Ne gas. This

provides new information on the long-range part of the Na<sub>2</sub>-Ne potential.

The beam of sodium atoms and dimers was produced in a seeded supersonic expansion using either argon or krypton as the carrier gas. By heating the Na reservoir to 800 °C (Na vapor pressure ~300 torr), we were able to enhance the population of sodium dimers in the beam to as much as 30% of the detected beam intensity [7]. At a carrier gas pressure of 2000 torr with a 70 μm nozzle aperture, we produced a cold Na<sub>2</sub> beam with only 3.5% rms longitudinal velocity spread, corresponding to a (longitudinal) translational temperature of 2 K.

To create a pure molecular beam, we used resonant light pressure forces acting on the atoms to deflect them sideways out of the collimated beam (Fig. 1). The deflecting laser beam was produced with a dye laser (Coherent 599) tuned to the  $F = 2 \rightarrow F' = 3$  transition of the  $D_2$  line in Na (589.0 nm). The frequency was maintained at the Doppler free resonance by serving the laser frequency to keep a fluorescent spot centered in the spreading atomic beam before the first collimating slit [8]. An electro-optic modulator was used to generate frequency sidebands at an offset of 1.713 GHz to deflect atoms in the  $F = 1$  hyperfine state. The laser light was applied at a position centered between two 20 μm collimating slits separated by 85 cm. With this geometry,  $\sim 3\hbar k$  of transverse momentum transfer is required to deflect the atoms out of the beam. A knife edge was positioned directly upstream from the laser beam to prevent any atoms that would normally miss the second collimation slit from being deflected back into the molecular beam. The molecules are not resonant with the deflecting laser beam and are therefore unaffected. (The  $X^1\Sigma_g^+ \rightarrow A^1\Sigma_u^+$  transition to the first excited dimer state is strongest around 680 nm [9].)

We investigated molecular diffraction with 100, 140, and 200 nm period microfabricated diffraction gratings. Since the sodium atoms and molecules coming from the supersonic expansion have nearly the same velocity [10], their momenta and hence their de Broglie wavelengths

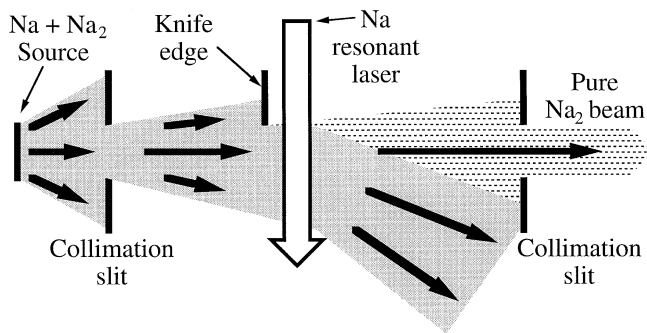


FIG. 1. Removal of the sodium atoms from the beam. The deflecting laser imparts a transverse momentum to the sodium atoms, deflecting them away from the second collimation slit.

differ by a factor of 2 because of their mass difference. For a source temperature of 770 °C and Kr carrier gas,  $\lambda_{dB}(\text{Na}) = 0.21 \text{ \AA}$  and  $\lambda_{dB}(\text{Na}_2) = 0.11 \text{ \AA}$ . A diffraction pattern obtained with a 100 nm grating and the mixed Na-Na<sub>2</sub> beam (deflecting laser off) is shown in Fig. 2(a). The various diffracted orders of the sodium atoms are sufficiently separated to permit easy identification of the intermediate molecular diffraction peaks at half the diffraction angle of the atoms. When the atoms are removed from the beam [Fig. 2(b), deflecting beam on], we resolve the molecular diffraction out to the fourth order. From a comparison of Figs. 2(a) and 2(b) we can determine that our Na<sub>2</sub> beam contains less than 2% residual atoms.

These diffraction patterns provide us with a powerful tool for analyzing the atoms and molecules in the expansion. We determined the mean fraction of molecules and the center and width of the velocity distributions for both atoms and dimers from these diffraction patterns. We observed as much as a 3.5(6)% velocity slip between the atoms and slower moving molecules at low source pressures (400 torr) [10]. At a typical source pressure of 1500 torr the slip was less than 1%. Using diffraction to investigate molecular beams has the advantage of being nondestructive and therefore applicable to very weakly bound systems such as He<sub>n</sub> molecules or other van der Waals clusters. Indeed, our method and our gratings have recently been used to produce unequivocal evidence of the existence of He<sub>2</sub> molecules [11].

We have employed three 200 nm fabricated gratings to construct a Mach-Zehnder interferometer for molecules (Fig. 3), similar to our atom interferometer [12]. This configuration produces high contrast fringes with our pure Na<sub>2</sub> molecular beam (inset, Fig. 4). For both the mixed interferometer (atoms + molecules) and the pure molecule interferometer, the maximum observed contrast  $(I_{\max} - I_{\min})/(I_{\max} + I_{\min})$  was nearly 30%.

The period and phase of the molecular interference pattern is the same as would be observed for atoms, even though the molecules traverse the interferometer on different paths than the atoms (Fig. 3). This is because the three grating geometry produces a white

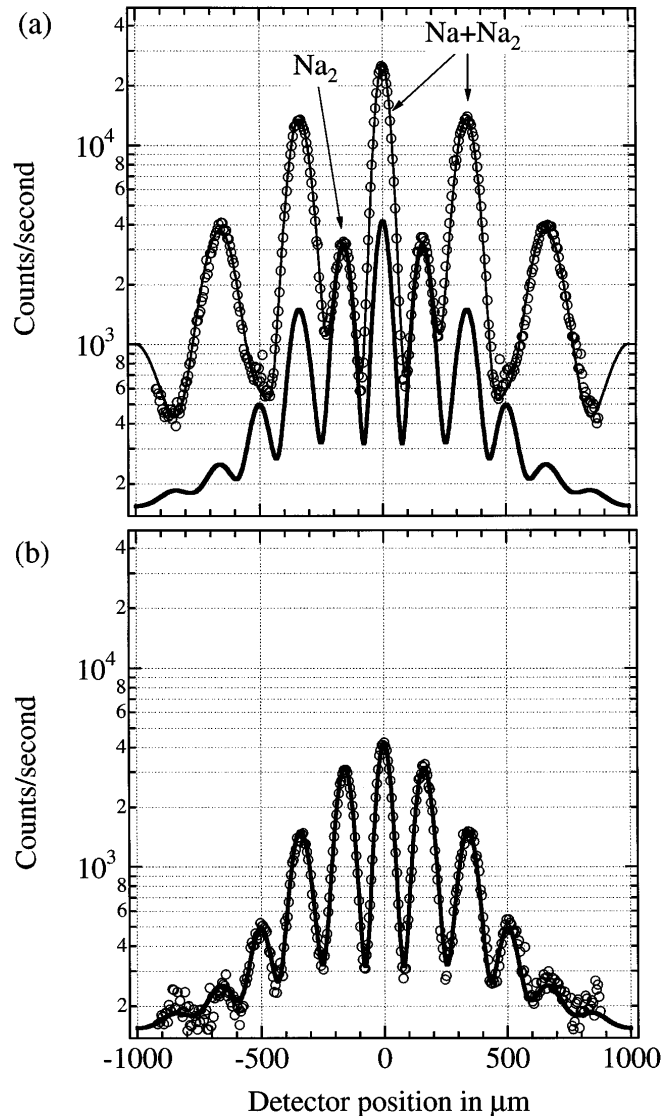


FIG. 2. (a) Diffraction of the mixed atom-molecule beam (deflecting beam off) and (b) molecule beam (deflecting beam on) by a 100 nm period grating. The thin solid line in (a) is a fit to the diffraction pattern, indicating that 16.5% of the detected intensity are molecules. The thick solid line in both graphs is the fit to the Na<sub>2</sub> diffraction pattern in (b). Both fits are for a diffraction grating with 30% open fraction and a beam velocity of 820 m/s (Kr as a carrier gas).

light interference pattern [13] whose period and phase are independent of the de Broglie wavelength of the particle. Therefore, in addition to using the previously described laser deflection scheme to produce a pure beam of Na<sub>2</sub>, we used two other methods to verify that the observed interference is from molecules: We introduced a (decoherence) laser which destroys the atomic interference by scattering resonant light from the split atomic wave function inside the interferometer, and we checked that the interference signal  $(I_{\max} - I_{\min})/2$  varied as expected with the transverse position of the third grating.

The results from a study combining both methods are shown in Fig. 4. For each third grating position, we

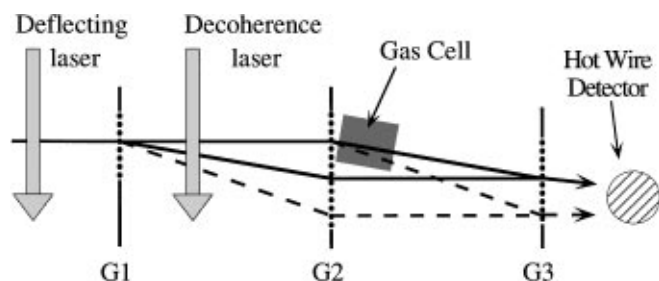


FIG. 3. Schematic of our interferometer showing the paths of Na (dashed line) and Na<sub>2</sub> (solid line). G1, G2, and G3 indicate the three diffraction gratings.

observe the largest interference signal from the combined atom and molecule fringe pattern (both lasers off). With either the deflection or decoherence laser beam on, the interference signal is reduced the same amount, indicating that only molecules contribute to the interference. This interpretation is confirmed because the interference signal does not decrease further when both lasers are on simultaneously.

The measured variation of the interference signal as a function of third grating position is compared with curves calculated by convolving the beam profiles with the detector acceptance. The atom or molecule interference fringes at the third grating have a trapezoidal spatial envelope with a width determined by the collimating slits [2] and an offset from the collimation axis determined by the diffraction angle (different for atoms and molecules) and the

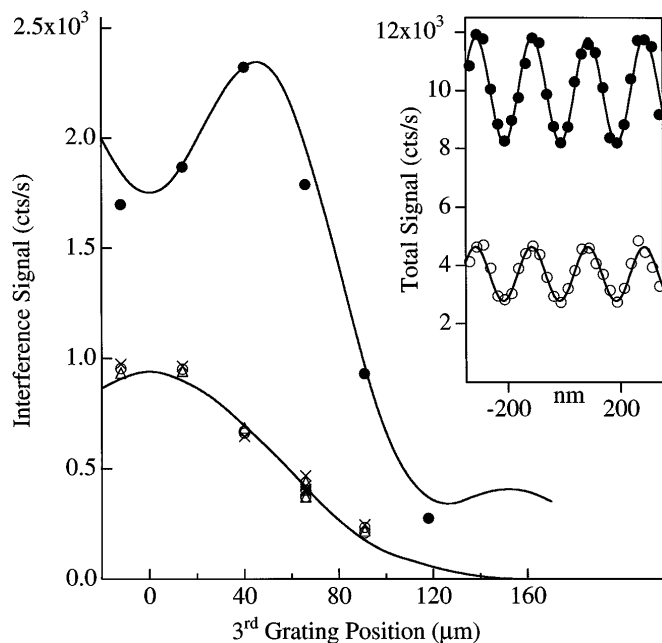


FIG. 4. The variation of the interfering signal vs third grating position relative to the collimation axis is shown for the mixed Na-Na<sub>2</sub> beam (•, no laser on) and the pure Na<sub>2</sub> beam (×, decoherence laser on; Δ, deflecting laser on; ○, both lasers on). Calculated curves are discussed in the text. Inset: The interference fringe data for the mixed Na-Na<sub>2</sub> beam (•) and the pure Na<sub>2</sub> beam (○) observed with the third grating at  $-10 \mu\text{m}$ .

distance between the gratings (Fig. 3). In addition to the interfering paths shown in Fig. 3, there are also interfering paths symmetric to the collimation axis as well as (small) contributions at larger offsets from the third grating involving the first and second order diffractions from the first grating. Thus, the detected interference signal is a convolution of the spatial envelope(s) of the interference signal with the acceptance of the detector, which is determined by the  $50 \mu\text{m}$  width of the third grating and its lateral position. We observe a peak in interference signal for the predominately atomic beam at  $\sim 50 \mu\text{m}$  from the collimation axis, as expected from the diffraction angle for Ar carrier gas ( $v_{\text{beam}} = 1000 \text{ m/s}$ ). A similar peak is not resolved in the Na<sub>2</sub> signal because of the smaller diffraction angle. The upper curve is normalized to the maximum observed interference signal and the lower curve is predicted from the known fraction of molecules (27%) (i.e., it is not independently normalized).

We observed no degradation in molecular interference signal despite the plethora of close lying rovibrational states in the molecules. This is not entirely surprising. The fact that two nearby molecules are very unlikely to have the same quantum numbers for both rovibrational state and total angular momentum projection is not important since the first order interference observed in an interferometer involves only the interference of each particle with itself. Although the 300 K thermal background photon energies typically exceed the internal level spacing of molecules ( $\sim 1 \text{ cm}^{-1}$  for rotations and  $\sim 100 \text{ cm}^{-1}$  for vibrations), decoherence effects due to transitions between vibrational or rotational levels or spontaneous emission are minimized because electric dipole transitions between rovibrational levels in the same electronic state are not allowed in a homonuclear diatomic molecule [9]. Scattering of the molecules on the microfabricated diffraction gratings could also cause rotational or vibrational transitions, since a beam velocity of 1000 m/s and a grating thickness of 200 nm produces Fourier components up to 5 GHz (or  $0.17 \text{ cm}^{-1}$ ). However, this is less than the smallest rotational splitting  $2B$  ( $B$  is the rotational constant) of  $0.31 \text{ cm}^{-1}$  and much smaller than the vibrational spacing of  $159 \text{ cm}^{-1}$ . As expected, we did not observe any contrast reduction, indicating an absence of these transitions.

Using Kr as the carrier gas, our Na<sub>2</sub> interferometer produces a molecular beam separation of  $38 \mu\text{m}$  at the second grating. This just exceeds the beam width at that position and allows insertion of an interaction region with a thin Mylar barrier between the interfering beams. The foil casts a shadow  $20 \mu\text{m}$  wide, which partially blocks the edges of the two beams, reducing the contrast from 19% without the foil to 7% with the foil. The lower observed contrast with krypton as the carrier gas (even without the inserted foil) is attributed to the slower beam velocity which enhances the inertial sensitivity of the interferometer, making it more vulnerable to vibrations

of the apparatus. A similar contrast reduction is observed with atoms.

We have used this separated beam molecule interferometer to measure the ratio of the real to imaginary parts of the index of refraction for the  $\text{Na}_2$  de Broglie waves passing through a Ne gas cell in one path of the interferometer, as we have done with atoms [14]. In analogy to light optics, the wave propagation through the gas cell of length  $L$  is modified by the factor  $\exp[i(n-1)k_{\text{lab}}L]$ , where the index of refraction  $n-1 = 2\pi Nf(k,0)/k_{\text{lab}}k$  is characterized by the complex forward-scattering amplitude  $f(k,0)$ , the number density  $N$  of the Ne gas, and the wave vector in both the laboratory ( $k_{\text{lab}}$ ) and center-of-mass ( $k$ ) frame of the collision. By measuring both the phase shift and attenuation of the interferometer for different target densities, we have measured the ratio  $\text{Re}[f(k,0)]/\text{Im}[f(k,0)] = 1.4(3)$  for  $\text{Na}_2$  scattering from Ne. The corresponding value for atomic Na waves passing through Ne is 0.98(2), a value considerably in excess of the 0.72 expected for a pure  $C_6/R^6$  potential [14]. We have separately measured the total absorption of  $\text{Na}_2$  by Ne (i.e.,  $(4\pi/k)\text{Im}[f(k,0)]$ ) to be 57(2)% larger than the corresponding absorption of Na, whereas a 32% increase would be expected for a pure  $C_6/R^6$  potential if  $C_6$  for  $\text{Na}_2$ -Ne were twice that for Na-Ne. Our measurements are in qualitative agreement with Na-Ne potentials from [15] if extended to  $\text{Na}_2$  using combination rules from Ref. [16]. More detailed studies will allow us to investigate how the long-range interactions for combined systems can be predicted from the simple systems.

In conclusion, we have demonstrated the production and diffraction of an intense beam of sodium dimers. We find that the diffraction of a seeded supersonic sodium beam by a transmission grating can serve as a sensitive intrinsically nondestructive probe of the constituents of an atomic beam. Further, we built a three-grating interferometer for molecules and observed no degradation in the interference signal compared with atoms. We then spatially isolated the two interfering molecular beams and measured the index of refraction of neon gas as seen by the molecular matter waves.

This work opens up the field of molecular optics and molecular interferometry with new possibilities of making fundamental and precision experiments on molecules similar to the ones recently conducted in atom interferometry. Separated beam molecule interferometry provides a new way to study interactions of the molecular ground states. For example, these techniques could be used to investigate the electric polarizability of diatomic molecules [17]. By measuring the phase shift and contrast as a function of an applied electric field one could determine both the parallel and perpendicular (to the molecular axis) polarizabilities. This experimental technique may also be used to measure the interaction of a polar molecule with the background electromagnetic field to investigate decoherence of external degrees of freedom caused by the interaction with a thermal bath.

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