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Speckle Patterns with Atomic and Molecular de Broglie Waves

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We have developed a nozzle source that delivers a continuous beam of atomic helium or molecular hydrogen having a high degree of transverse coherence and with adequate optical brightness to enable new kinds of experiments. Using this source we have measured single slit diffraction patterns and the first ever speckle-diffraction patterns using atomic and molecular de Broglie waves. Our results suggest fruitful application of coherent matter beams in dynamic scattering and diffractive imaging at short wavelength and with extreme surface sensitivity.

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After de Broglie first postulated [1] and others verified [2–4] the wave nature of massive particles, applications using matter waves expanded quickly into a diverse array of mainstream scientific instruments, including many electron microscopies and a host of scattering and diffraction techniques. Later, invention of the laser vastly facilitated exploration and exploitation of wave coherence. The intersection of these two developments has been slow to develop, largely due to the lack of sources with high coherent flux. A notable exception is the field of atom optics [5], which has produced a dazzling array of results relying on atom coherence in the long wavelength limit. There remains a keen interest in developing short wavelength coherent sources to enable diffractive imaging and coherent scattering techniques with higher resolution than is available in the optical regime. Much progress along these lines has been accomplished by extracting the coherent fraction from field emission cathodes in electron microscopy [6,7]. The high brightness available at third generation synchrotron radiation sources has spawned efforts to use coherence in the soft and hard x-ray regimes [8–10]. High flux helium beams have been developed to study surface dynamical phenomena [11], and collimating these to achieve high coherence in one dimension has led to several pioneering experiments [12–14]. Here we describe a new apparatus that produces continuous beams of thermal energy helium atoms and hydrogen molecules having a high degree of transverse coherence in two dimensions and a coherent flux that will be useful for a variety of new kinds of experiments. The particles have de Broglie wavelengths comparable to x rays, are neutral and nondamaging, and are scattered with high surface selectivity. We report the results of preliminary tests in which we have used this apparatus to measure single slit diffraction patterns and a speckle-diffraction pattern of an irregularly shaped object.

Figure 1 shows a schematic of our experimental setup. A key ingredient of any experiment utilizing wave coherence is source brightness (B), as measured by particle flux per unit area and solid angle, since this is directly related to the available coherent flux (F_{coh}) by [8]:

$$F_{\mathrm{coh}} = B\left(\frac{\bar{\lambda}}{2}\right)^2, \tag{1}$$

where $\bar{\lambda}$ is the de Broglie wavelength. Our He or H_2 source is a glass capillary which has been drawn into a 2 μm diameter nozzle [15]. Gaseous He or H_2 at variable stagnation temperature T_0 and pressure P_0 expands freely through the nozzle into vacuum. Using a small nozzle diameter and high P_0 will produce a beam of high brightness [15]. The resulting gas jet passes through a conical skimmer with an opening diameter of 300 μm into a differential pumping region, and then into an ultrahigh vacuum chamber and through a spatial filter consisting of 5 μm and 1 μm diameter pinholes separated by 8 cm. For a surface scattering experiment, the spatially filtered coherent beam would be incident on a scattering sample. Scattered atoms or molecules are field ionized with nearly unit efficiency using a tungsten tip having a nominal radius of ~100 nm held at a temperature of ~100 K and a voltage of typically +10 kV. Field ionization detection of molecular beams has been used for some time, and a recent article describes the current state of the pursuit [16,17]. The resulting ions are detected with a channel

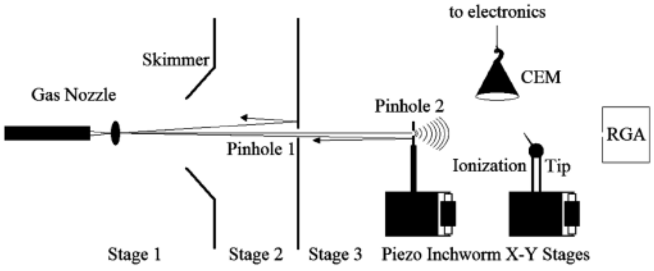

FIG. 1. Schematic representation of our apparatus. Shown are the glass capillary nozzle mounted in a metal high-pressure chamber, two spatial filter pinholes, the micropositioning stages that hold the second pinhole and the detector consisting of a tungsten field ionization tip and the channel electron multiplier (CEM), and a calibrated residual gas analyzer (RGA) used for alignment and to measure the coherent flux.

electron multiplier (CEM). The advantage of using field ionization is that diffraction patterns can be measured with the spatial resolution necessary to resolve interference fringes by scanning the tip with piezoelectric actuated stages. The very small ionization volume of the tip also effectively discriminates against the ambient gas.

Coherence is the property of waves that leads to interference; that is, if coherent waves are overlapped in space and time, there will be coherent addition of wave amplitudes rather than incoherent addition of wave intensities [18]. Longitudinal coherence pertains to the monochromaticity or degree of regularity of the wave fronts in the direction of propagation. For a free-jet expansion, the longitudinal coherence length l_L can be expressed in terms of the speed ratio, S [19]:

$$l_L = \bar{\lambda} S = \bar{\lambda} \frac{\bar{\nu}}{\Delta \nu} = \frac{\bar{\lambda}^2}{\Delta \lambda}, \tag{2}$$

where $\bar{\nu}$ is the average speed. By extrapolating previous results [11] for our nozzle parameters we estimate $S \sim 30$–40 for H_2 and $S \sim 100$–110 for He at $T_0 \sim 100$ K. Combining this with the de Broglie wavelengths determined below, we determine l_L to be several nanometers. High diffraction fringe contrast will be observed if the scattering path length difference is less than the longitudinal coherence length. l_L does not limit contrast in the results presented below, but it does provide a fundamental limit on our ultimate spatial resolution.

Transverse coherence refers to the degree of regularity of wave fronts perpendicular to the propagation direction [18] and in molecular beams is related to the less-well-studied perpendicular velocity distribution [15,20]. To improve the limited transverse coherence of a free-jet source, we insert a spatial filter composed of two pinholes that set the transverse phase space acceptance at the level of the Heisenberg Uncertainty Principle. The correlation of wave amplitudes at two points in an observation plane will diminish over a scale given by the transverse coherence length l_T, which can be predicted using the Zernicke–van Cittert theorem [18] to be

$$l_T = \frac{R\bar{\lambda}}{2d_1}, \tag{3}$$

where d_1 is the first pinhole size and R is the distance between the pinhole and the plane. A wave field with an average degree of coherence of ~90% will fill the second pinhole in the spatial filter if its diameter d_2 is smaller than the transverse coherence length l_T, leading to the constraint

$$\frac{2d_1 d_2}{R} < \bar{\lambda}. \tag{4}$$

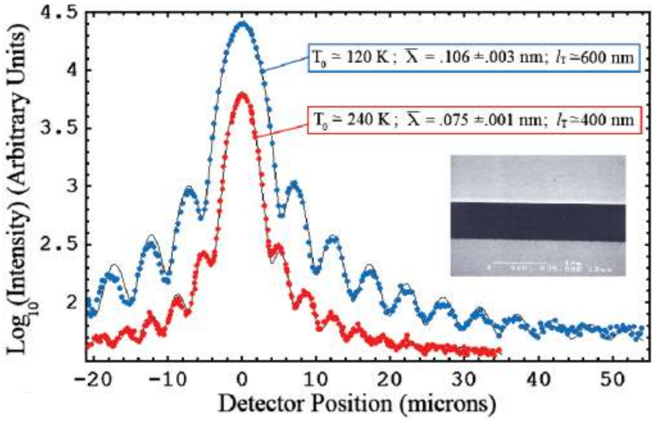

FIG. 2 (color). Measured and fitted single slit diffraction patterns using hydrogen gas at $P_0 = 700$ kPa and $T_0 = 120$ K and 240 K (calculated). Each scan took ~20 min to record. The inset shows a scanning electron micrograph of our single slit produced in a silicon nitride membrane.

To test our approach, we diffracted He and H_2 beams off several different apertures fabricated in 100 nm thick silicon nitride membranes using a focused ion beam. The inset in Fig. 2 shows a micrograph of a 0.7 μm wide slit that replaced the second spatial filter pinhole. The resulting diffraction patterns shown in Fig. 2 exhibit fringes to at least seventh order before they become lost in the background produced by ionization of ambient gas. We have fitted our results to an ideal single slit diffraction pattern convoluted with a Gaussian distribution of transverse wave vectors of width $\delta k_T = \pi / l_T$ [21], with addition of a constant background. The resulting fits and derived values of $\bar{\lambda}$ and l_T are shown in the figure. We cannot accurately measure the nozzle temperature T_0, and therefore assume conversion of stagnation gas enthalpy into directed kinetic energy [19] to infer the nozzle temperatures shown. The derived values δk_T actually correspond to the width of the transverse wave vector distribution projected onto the detector plane convoluted with the unknown size of the detector ionization region and the amplitude of any residual vibrations of the apparatus. The true transverse coherence lengths, therefore, are larger than those reported in Fig. 2.

The results in Fig. 2 provide a good characterization of beam properties but do not require coherence in both transverse directions. They also are not fundamentally new, since researchers have produced thermal energy particle waves with good coherence in one transverse direction and have used these in single slit geometries to perform various precision measurements [12–14,22–24]. A beam approaching full transverse coherence will allow qualitatively new classes of scattering experiments, while also further reducing the signal by factor of $>10^4$ due to increased collimation. To demonstrate good transverse coherence requires that we measure the diffraction pattern of an object that is small in both dimensions. We have measured Airy fringes from single pinholes, but a result that is closer to our planned experiments is to produce a speckle-diffraction pattern of a random structure. Mechanical limitations precluded scattering the beam off an imperfect surface. Instead, we measured a speckle pattern produced in transmission through the random pinhole pattern shown in Fig. 3(a). In Fig. 3(b) we show the expected speckle amplitude, which is the magnitude of the Fourier transform of Fig. 3(a). The raw speckle amplitude, measured reproducibly using a raster scan, is shown in Fig. 3(c). The diffraction pattern of a real object must have inversion symmetry, and aside from uncertainty in the measurement, our pattern does indeed exhibit this symmetry, as evidenced by the similarity of the image in Fig. 3(c) to its symmetrized version in Fig. 3(d). The observed and predicted speckle patterns have similar features, although finite coherence again diminishes the visibility of the measured fringes. The quality of these data is not sufficient to extract a coherence length as was done for the single slit results in Fig. 2. Instead, Fig. 3(e) shows the result of convoluting the ideal diffraction pattern in panel (b) with a two-dimensional distribution of transverse wave vectors having an isotropic width equal to that deduced in Fig. 2. The predicted fringe contrast in panel (e) is comparable to that in the symmetrized experimental pattern in panel (d), and the similarity between measured and predicted fringe patterns is clear.

These results constitute the first speckle-diffraction pattern collected with atomic or molecular de Broglie waves. More importantly, we have achieved a brightness and coherent flux high enough to enable several future research directions. Table I summarizes the typical brightness, wavelength, and coherent flux of this and other short wavelength particle and photon sources. We have shown elsewhere how to produce a source brightness in excess of 10^{28} atoms/sec $\mathrm{m}^2\,\mathrm{rad}^2$ [15], which is a few orders of magnitude below the average brightness of undulators at third generation synchrotron radiation facilities. Given its simplicity, relative low cost, continuous availability, and the unit probability for helium scattering from surfaces, our source will be very competitive in some applications. For example, we have adapted an existing theory for diffuse x-ray and neutron surface reflectivity [25] to estimate the average flux of coherent helium atoms scattered into a single speckle from a surface with typical roughness characteristics. Atoms scatter diffusely with unit efficiency into a distribution that is roughly the Fourier transform of the surface height-height correlation function. For an optimized apparatus, the predicted average flux/speckle is 3–4 orders of magnitude below the incident flux—a substantial signal that could be used to measure surface fluctuations or to do diffractive imaging.

A remaining instrumentation issue that is crucial to the long-term application of coherent atomic and molecular beams is the detector. With a dwell time of a few sec/pixel, ~2000 pixels, and a duty cycle of <50%, the image in Fig. 3 required a total accumulation time of several hours. A larger momentum range was not accumulated due primarily to the thermal stability of our apparatus. Diffractive imaging, where $\sim 10^6$ pixels are required, is not possible using a single channel detector. This atom optical experiment would benefit from developing an "atom camera" that would measure the entire speckle pattern in one exposure. Field emission and ionization tip arrays have been developed and used in various sensor and display applications [26,27], and integrating these with the necessary ion optics and area detector would provide a sound approach for such a development. If such an atom camera were

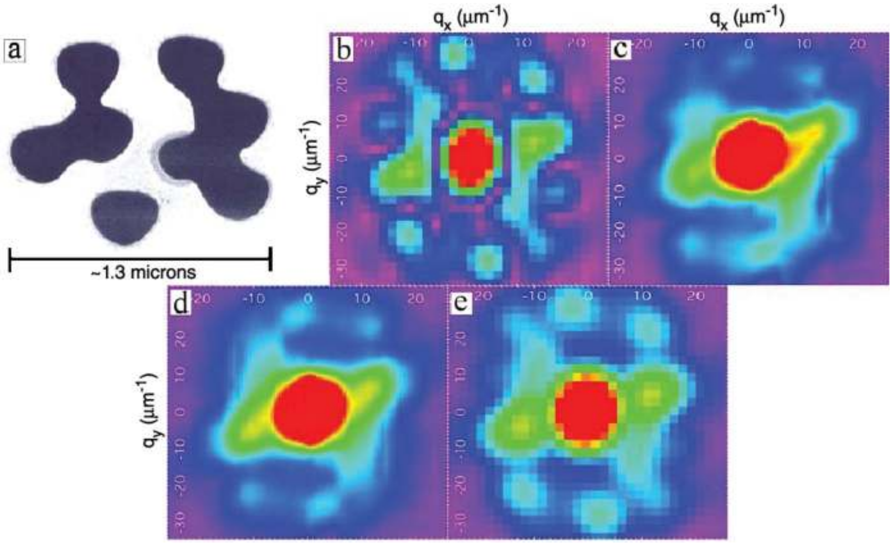

FIG. 3 (color). (a) Scanning electron micrograph of the random pinhole structure produced in a silicon nitride membrane; (b) expected speckle-diffraction amplitude, which is simply the magnitude of the Fourier transform of the structure in panel (a); (c) the measured speckle-diffraction amplitudes from the structure in panel (a); (d) inversion-symmetrized speckle-diffraction pattern; (e) expected speckle-diffraction amplitude corrected for finite coherence as explained in the text. Amplitudes rather than intensities are plotted to make the weaker fringes more easily visible.

TABLE I. Typical brightness, wavelength, and coherent flux of various short wavelength particle and photon sources. Coherent flux values are derived from Eq. (1) and assume no monochromatization beyond the natural bandwidth of the source.

	Brightness (particles/sec m^2 rad^2)	Wavelength (nm)	Coherent flux (particles/sec)
Helium micronozzle			
This study	3×10^{27}	0.1	1×10^{7}
Optimized [15]	3×10^{28}	0.1	1×10^{8}
X-ray undulator	10^{32}	0.1	2×10^{11}
Field emission tip	10^{32}	0.004	4×10^{8}

employed, a full diffraction pattern could be produced in a few minutes with adequate signal to provide a real-space resolution, limited by longitudinal coherence, of a few nanometers. Such a detector would also benefit the atomic analog of dynamic light scattering to measure soft, diffusive modes at surfaces on time scales longer than a microsecond or on energy scales less than a nanovolt. Finally, our source and such a detector would also facilitate several experiments at the interface of quantum optics and quantum physics. Our degeneracy parameter [28]—the number of particles per coherence volume, $V_{\mathrm{coh}} \approx l_T{}^2 l_L$—is presently $\delta \sim 10^{-4}$, and will increase to 10^{-3} with a next-generation source design [15]. While this is small compared to liquid helium and Bose-Einstein condensates, where $\delta \sim 1$, and to lasers, where δ is macroscopically large, Hanbury Brown and Twiss [29] accomplished pioneering quantum optical experiments 50 years ago using mercury lamps with degeneracy parameters comparable to ours.

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