Fraunhofer Diffraction of Atomic Matter Waves: Electron Transfer Studies with a Laser Cooled Target

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We have constructed an apparatus combining the experimental techniques of cold target recoil ion momentum spectroscopy and a laser cooled target. We measure angle differential cross sections in $Li^+ + Na \rightarrow Li + Na^+$ electron transfer collisions in the keV energy regime with a momentum resolution of 0.12 a.u. yielding an order of magnitude better angular resolution than previous measurements. We resolve Fraunhofer-type diffraction patterns in the differential cross sections. Good agreement with predictions of the semiclassical impact parameter method is obtained.

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The phenomenon of Fraunhofer diffraction is one of the more spectacular manifestations of the wave nature of light. When a beam from a coherent light source of wavelength λ is confined to pass through a hole of radius r, the spot pattern formed at a distant screen consists of a central peak surrounded by concentric circles with a nearly equidistant angular spacing of $\Delta\theta = \lambda/2r$. Airy [1] was the first to derive the exact expression for the angular intensity distribution in the spot by integration of the coherent contributions to the scattering amplitude from the different parts of the hole, expressed in the celebrated formula

$$I(\theta) \propto \left| \int_0^r db\, bJ_0(kb\theta) \right|^2 \propto \left| \frac{J_1(2\pi\theta r/\lambda)}{\theta} \right|^2 \quad (1)$$

with J_i being the Bessel functions and $k = 2\pi/\lambda$. For visible light and typical geometries, however, the characteristic angles in question are very small, explaining why it was not until the carefully designed investigation of Fraunhofer that these properties were clearly appreciated.

By analogy, if an atomic matter wave of *de Broglie* wavelength λ_{dB} is confined by an aperture, a Fraunhofer-type diffraction pattern should appear. Again, in typical cases the scattering angles in question are prohibitively small for observation. This Letter demonstrates how, by combining the recently developed powerful techniques of cold target recoil ion momentum spectroscopy (COLTRIMS) [2–4] and laser cooling, an angular resolution may be achieved in an atomic scattering experiment that enables Fraunhofer patterns to become clearly observable for coherent matter waves produced in a judiciously selected electron transfer process.

When a $^6Li^+$ ion with a kinetic energy of a few keV hits a sodium atom in the Na($3s$) ground state with an impact parameter b less than about $10a_0$, the probability for transfer of the $3s$ valence electron into the Li($2s$) state is very high, as detailed in several experimental and theoretical studies [5–10]. As a result of this S-S transfer, a coherent Li($2s$) atom wave will emerge from a "hole" of radius $r = 10a_0$. Recent semiclassical theoretical studies of this process solve the close-coupled scattering equations using 26/28 state basis sets of atomic/molecular (AO/MO) orbitals and straight line trajectories [8–11]. The corresponding angular differential cross section (DCS) for a reduced mass μ and incident velocity v, obtained by the Eikonal transformation, indeed reveals a Fraunhofer pattern for the S-S electron transfer channel, as evaluated by the formula

$$\frac{d\sigma(\theta,\phi)}{d\Omega} = \sigma(\theta) = k^2 \left| \int_0^\infty db\, bJ_0(kb\theta)a(b) \right|^2, \quad (2)$$

where $k = 2\pi/\lambda_{dB} = \mu v/\hbar$, and $a(b)$ is the scattering amplitude for S-S electron transfer, which falls off rapidly for impact parameters above $r = 10a_0$. The diffraction rings have a characteristic angular spacing of about $\Delta\theta = \lambda_{dB}/2r = \pi/kr = 0.008°$ for 6 keV $^6Li^+$ ions ($v = 0.2$ a.u.). However, corresponding experimental studies using conventional scattering geometries [5–7] have been performed with angular resolutions of 0.01° (FWHM), or worse. As demonstrated below, our novel combination of laser cooling and 3D COLTRIMS spectroscopy enables us to achieve an angular resolution which is about an order of magnitude higher, and we clearly resolve a rich structure in the DCS which allows us to perform a detailed comparison of theory and experiment.

The experimental technique of COLTRIMS has in recent years enabled a range of kinematically complete investigations of atomic collision processes (see [2–4] and references therein) with unprecedented angular resolution. The idea is to evaluate the scattering angle from the momentum ratio $\theta = \Delta p/p$, where Δp is the transverse momentum transfer and p is the incident momentum. The momentum transfer is determined by analysis of the recoiling target ion, which is extracted by an electric field. The resolution of this technique is limited by the thermal motion of the target, which accordingly is cooled. COLTRIMS studies of He have led to new insight into a diversity of problems. Recent highlights with slow ionic projectiles include results in electron transfer [12,13], single ionization [14,15], and molecular breakup [16]. The requirement of a cold target in COLTRIMS spectroscopy has restricted the targets to precooled supersonic gas

jets such as He, H_2, Ar, and Xe. A new generation of spectrometers incorporating the laser cooling technique will extend the applicability of COLTRIMS to a much larger number of atomic targets.

The magneto-optical trap (MOT) has been a very successful tool in a large variety of atomic physics experiments. Few, however, use the MOT as a target in scattering experiments. Schappe *et al.* [17] used a Rb MOT as a target in a pioneering measurement of absolute total electron scattering cross sections. Wolf and Helm [18] measured the recoil ion energy of photoionized Rb^+ ions from a Rb MOT, and Eike *et al.* [19] tested the use of a Cs MOT as a probe of the profile of a stored heavy-ion beam. Only in the experiment of Wolf and Helm is the low temperature of the trapped atoms essential for success.

Our new experimental setup can be summarized as follows: A Na MOT acts as a target for a Li^+ beam. The recoiling Na^+ ions resulting from electron transfer collisions are momentum analyzed in a COLTRIMS spectrometer, which employs 3D focusing in order to achieve high resolution of all momentum components [3]. The recoil ion time of flight (TOF) is measured through coincidence detection of the neutralized Li projectile and Na^+ recoil ion. A schematic diagram of the experimental setup is shown in Fig. 1. (Note the choice of the coordinate system: x is along the ion beam direction, and z is along the spectrometer axis.)

From such an experiment the full collision kinematics of the charge transfer reaction can be determined. In particular, measurement of the recoil ion momentum component p_x longitudinal to the projectile direction allows one to derive the energy defect in the collision. In this way we distinguish capture into the Li($2s$) and Li($2p$) channels.

Special care has been taken in the construction of the COLTRIMS spectrometer in order to create a well-defined extraction field while allowing for the access of six large-diameter laser beams needed for the MOT. The spectrometer is divided into an extraction region and a field free drift region before the recoiling ions hit a position sensitive detector (Z-stack microchannel plate detector with a delay line readout). The extraction field is defined by a cage of thin (40 μm) tungsten wires. Each wire is electrically isolated and connected to a resistor chain, which divides the applied spectrometer voltage equally between each wire. A resistor halfway between the collision volume and the drift region can be adjusted individually, thereby creating a lensing field. Four germanium coated glass plates with holes for the laser and projectile beams surround the extraction cage. Along the surface of each glass plate is applied a voltage drop that matches that of the extraction cage inside. In this way, external fields are shielded efficiently while the extraction field is disturbed as little as possible.

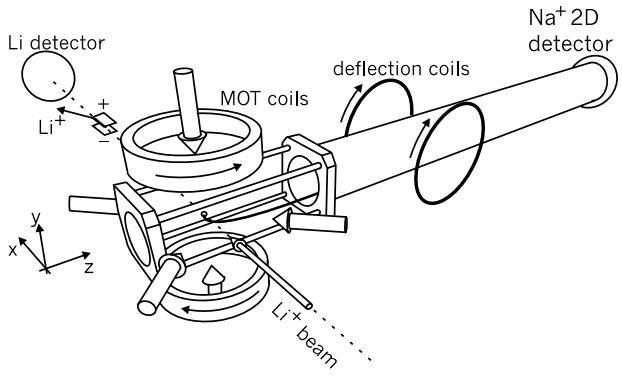

FIG. 1. Schematic diagram of the experimental setup. The large arrows indicate trapping laser beams. Current direction is indicated by arrows on the coils. The distance from the collision region to the position sensitive detector is 620 mm.

The atomic target is a standard vapor cell MOT in a vacuum chamber (pressure 3×10^{-8} mbar) with a quadrupole magnetic field created by a pair of anti-Helmholtz coils (gradient 1.5 mT/cm along the coil axis). We monitor the MOT through both the collision rate with our projectile beam and by fluorescence detection. We capture 5×10^7 atoms in the MOT with a central density of 4×10^{10} atoms/cm^3 using the Na $3\,^2S_{1/2}(F = 2) \rightarrow 3\,^2P_{3/2}(F = 3)$ transition. An 860 MHz electro-optical modulator [20] is used for simultaneous pumping of the $F = 2 \rightarrow F = 3$ and $F = 1 \rightarrow F = 2$ hyperfine transitions so that trapping of the atoms in a dark state is avoided. With the COLTRIMS spectrometer around the MOT, the trapping laser beams have to pass the cage of wires acting as electrodes for the extraction field. Diffraction from these wires affects the wave front of the laser beams so that the size of the atom cloud increases as compared to the "free" MOT, but the MOT is robust against these imperfect conditions. The MOT position can be adjusted by means of a weak static magnetic field along the y and z directions made by bias coils to ensure optimal alignment of MOT and projectile beam (typical values of B_y and B_z are 0.1 mT). A typical temperature of our MOT is below 1 mK [11] which corresponds to a momentum spread of 0.02 a.u.

The electrical field of our spectrometer is computed numerically [21]. A typical extraction field in the experiment is 500 mV/cm which gives a recoil ion TOF of about 90 μs. (From the 2D image from recoiling ions we derive the p_x and p_y momentum components.) For the three momentum components we obtain the resolution $(\delta p_x, \delta p_y, \delta p_z) = (0.30, 0.80, 0.12)$ a.u. The relatively low resolution of the p_y component is caused by distortion from the MOT quadrupole magnetic field. The p_z component derived from the recoil ion TOF is not affected appreciably by the magnetic field.

A typical image from the position sensitive detector is shown in Fig. 2.

The MOT target produces low-energy molecular Na_2^+ ions through the process of photoassociative ionization [22]. Accidental coincidences of these ions and neutralized projectiles are avoided by separating them from the

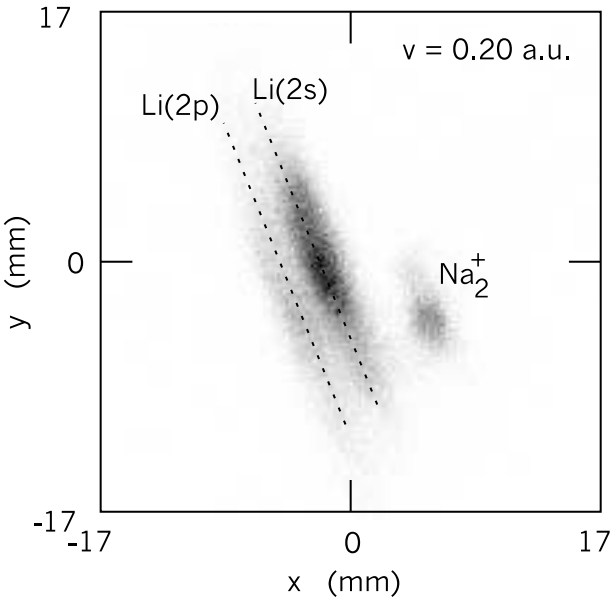

FIG. 2. Image obtained with the position sensitive detector. The lines corresponding to different collision channels are tilted from vertical due to the influence of the MOT magnetic field on the recoiling Na^+ trajectories. The rightmost spot corresponds to accidental coincidences with Na_2^+ ions produced by the MOT.

Na^+ recoil ions on the position sensitive detector with a perpendicular deflection magnetic field along the spectrometer drift tube. The p_x longitudinal momentum spectrum of Na^+ ions resulting from electron transfer collisions is mainly distributed between the Li($2s$) and Li($2p$) channels, respectively. Further analysis of the p_x spectrum reveals that transfer into unresolved Li final states with $n > 2$ is negligible at the present collision velocities.

A fraction of the trapped atoms in the MOT is excited to the Na($3p$) state by the near-resonant trapping light. Electron transfer from these excited atoms does occur predominantly into the quasiresonant Li($2p$) channel ($\Delta E = 0.50$ eV). These Na($3p$) $\rightarrow$ Li($2p$) transfer collisions cannot be resolved from the Na($3s$) $\rightarrow$ Li($2s$) collisions in the present experiment due to the very small difference of longitudinal recoil momentum ($\Delta p_x = 0.09$ a.u.). We estimate the excitation fraction α to be 25% from the formula given by Townsend *et al.* [23].

In our case of cylindrical scattering symmetry around the x axis, the 1D projection of the scattering pattern onto the z axis is given by the Abel transform of the differential cross section, $\sigma(\theta)$. By inverting this transform, we are therefore able to extract $\sigma(\theta)$ from p_z alone [24], the momentum component where we have the best resolution. The zero point of the p_z axis was found as the symmetry axis in the transverse momentum spectrum.

Figure 3 shows such a pattern at a beam velocity of 0.2 a.u. The angular resolution derived from our momentum resolution is varying inversely proportional to the beam velocity and is 0.003° at 0.2 a.u. The ring pattern

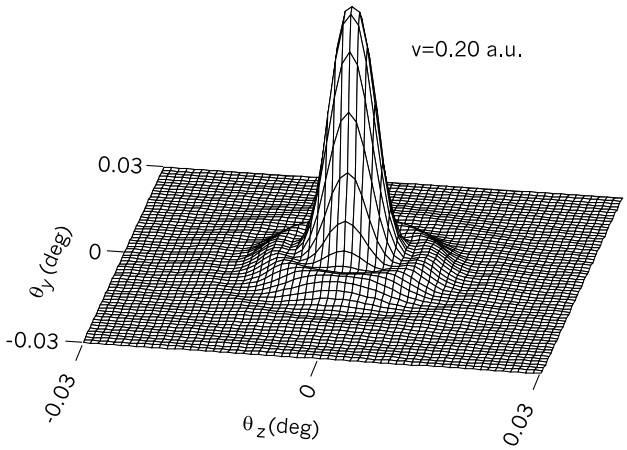

FIG. 3. Example of the Li angular scattering pattern $\sigma(\theta)$ in $Li^+ + Na$ electron transfer collisions deduced from experiment.

is clearly visible, with a spacing $\Delta\theta$ close to 0.008°, as estimated by the standard formula.

Figure 4 shows the quantity $\sigma(\theta)\sin(\theta)$ for collision velocities $v = 0.135$, 0.15, and 0.20 a.u., respectively. The experimental results are compared with theoretical predictions based on recent close-coupling calculations using 26 state AO [9,11] and 28 state MO [10] basis sets. Earlier

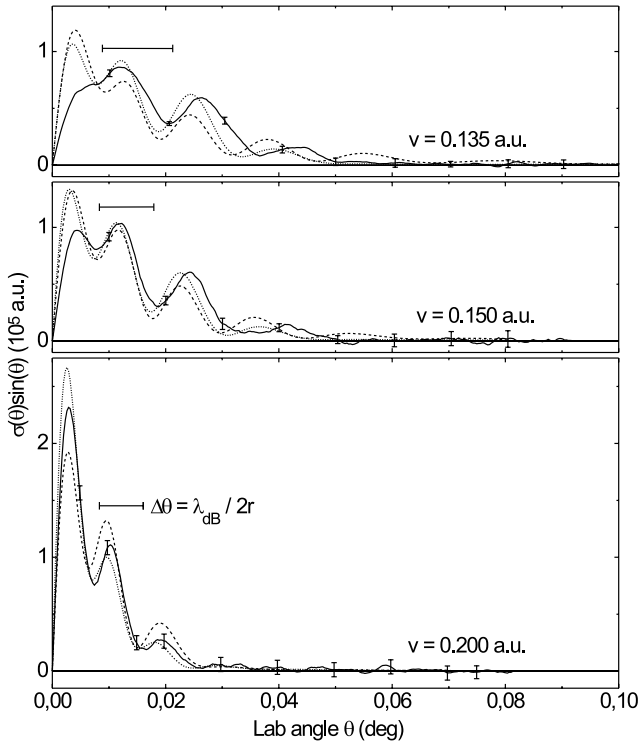

FIG. 4. $\sigma(\theta)\sin(\theta)$ for $Li^+ + Na$ electron transfer. The solid line is the experimental result; the dashed and dotted lines are predictions of AO and MO calculations, respectively. In the theory curves the target is assumed to be 25% isotropically excited to the Na($3p$) level. All curves are normalized to the AO total cross section and the theory has been convoluted with an apparatus function with a FWHM of 0.12 a.u. The error bars indicate statistical uncertainty off the raw data at selected angles.

experimental tests of these theories gave good agreement at the level of integrated [25] and differential cross sections [7]. In the latter case, however, much of the predicted angular structure was washed out in the experiment. Also indicated in the figure are the expected angular spacings $\Delta\theta$ (calculated for a critical radius of $r = 10a_0$), which vary inversely proportional to the impact velocity. The predicted positions and heights of the oscillations are close to the measured values, in particular, at the highest velocity. For the lowest velocity and the largest scattering angles, the predicted and observed oscillations tend to come out of phase. Detailed discussion of the results and comparisons between experiment and the theoretical predictions will be presented in a forthcoming paper [11].

In conclusion we have succeeded in achieving high resolution 3D recoil ion momentum spectra with a laser cooled target. Fraunhofer-type rings in the angular scattering pattern have been clearly resolved. Our measurements show good agreement with theoretical predictions using AO or MO basis sets. Furthermore, the present study is a crucial step towards the more delicate problem of addressing alignment and orientation effects for an optically prepared Na($3p$) target near velocity matching with dramatically improved angular resolution compared to earlier studies [8]. We also note the more tantalizing prospect of studying alignment and orientation effects in kinematically complete experiments of the ionization channel $Li^+ + Na(3p) \rightarrow Li^+ + Na^+ + e^-$.

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