ESRF	**Experiment title:** Investigating the 3D Compton Profile and the role of electron-electron correlation in H2	**Experiment number**: CH-6396
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Report:

After performing two highly successful beamtimes before the ESRF upgrade shutdown in 2018 (see experimental reports on CH-5345, Kircher, period 4/2018 and CH-5524, Kircher, period 8/2018) with four publications in Physical Review Letters, we followed the path of our long-term program for atomic and molecular physics utilizing Compton scattering. We brought a COLd Target Recoil Ion Momentum Spectroscopy (COLTRIMS) reaction microscope to beamline ID31 at ESRF and performed gas-phase studies of Compton scattering at nitrogen and neon. We successfully measured multiple Compton scattering and photoionization reaction channels on an event-by-event basis. Data analysis is still ongoing, however, some preliminary results are presented below.

With the COLTRIMS technique, an internally cooled supersonic gas jet is crossed at 90° angle with the synchrotron light. After Compton scattering at a single isolated atom or molecule, the resulting fragment ion(s) and electron(s) are guided by electric and magnetic fields toward two time- and position-sensitive detectors. By measuring the times-of-flight and positions-of-impact, one can deduce the 3D momenta of the ion(s) and electron(s) at the instant of the photoreaction in coincidence.

Our first goal was to measure the directional Compton profile of a molecular orbital in an isolated molecule in the gas phase. Even 100 years after Compton's discovery, Compton scattering could never been scrutinized in this level of detail because of the random orientation of free molecules. Initially, we planned to use an H_2 sample. Due to technical reasons, we performed the measurement studying N_2 instead. For the following considerations, however, H_2 and N_2 are analogous.

We measured the following Compton reaction:

$$\gamma + \mathrm{N}_2 \rightarrow \gamma' + \mathrm{N}^0 + \mathrm{N}^+ + e^-$$

The photon scatters at an electron bound in a molecular orbital of N_2, ionizing the system. With some probability, the intermediate N_2^+ molecular ion breaks up. The resulting N^0 and N^+ fragments possess back-to-back momenta, which we measure. For each event, we infer the orbital from which the electron was ejected from the ion energy and the orientation of the molecule from the direction of the N^+ fragment. By measuring the final

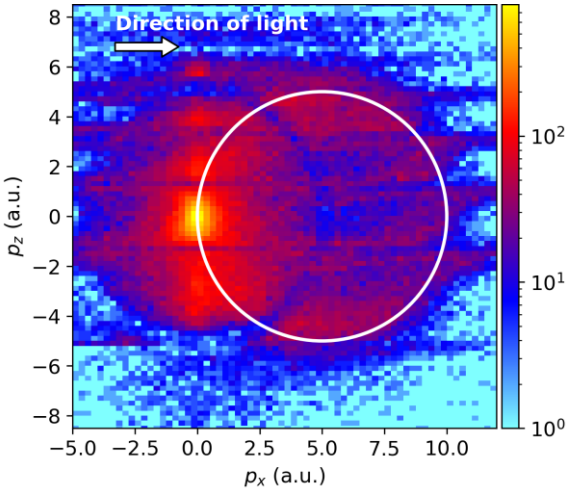

Fig. 1: Preliminary results for electron momenta resulting from ionization of N_2 by valence-shell Compton scattering. The direction of light is indicated in the upper left corner. The periodic features along the upward direction are an artifact of the detection method and require further analysis to be removed.

electron momentum (see Fig. 1), one gains insight into the Compton profile: for large scattering angles where the impulse approximation (IA) is applicable, the final electron momentum is simply the momentum transfer $\vec{Q} = \vec{p}_\gamma - \vec{p}_{\gamma'}$ plus the electron's initial momentum in the orbital. (Within the IA, the binding of the electron is only considered insofar that the electron's momentum distribution is given by the initial bound-state momentum distribution.) The width of the ring visible in Fig. 1 (emphasized by the white circle) correponds to the Compton profile. Since we have access to the molecular orientation at the reaction time, we can investigate the Compton profile for fixed-in-space molecular orientations. This requires further data analysis.

The initial photon can also scatter at the inner shell of N_2 instead of a valence shell. After ionization by Compton scattering, the intermediate $N_2^+(1s^{-1})$ state undergoes Auger decay, resulting in two N^+ ions and two electrons. This gives access to interesting dynamics of the two electrons and the molecular fragments. For a benchmark measurement of a less complicated (i.e., atomic) system, we investigated *K*-shell Compton scattering at neon. There, we are able to detect all charged particles of the reaction

$$\gamma + \mathrm{Ne} \rightarrow \gamma' + \mathrm{Ne}^{2+} + e^-_{\mathrm{Compton}} + e^-_{\mathrm{Auger}}$$

Therefore, the momentum of the final photon can be calculated by exploiting momentum conservation, which gives direct access to the momentum transfer onto the Ne system. Then, one can acquire the Compton electron momenta in a fixed-momentum-transfer reference frame, as is shown in Fig. 2. There, the momentum transfer always points upward. Interestingly, we find that while the majority of electrons follow the direction of the momentum transfer (i.e., upward), a significant amount with opposite direction exist (i.e., downward). The downward lobe is a result of the interaction of the electron with the remaining ion. As can be seen in panels B and C, the relative intensity of upward- to downward-pointing electrons depends on the magnitude of the momentun transfer. For small magnitudes (panel B), the upward and downward lobe are of almost equal intensity. For large magnitudes (panel C), an overall majority of electrons points in the direction of $\vec{Q}$. This shows a transition from a case where the IA is invalid (Q small) to a case where it is mostly applicable (Q large).

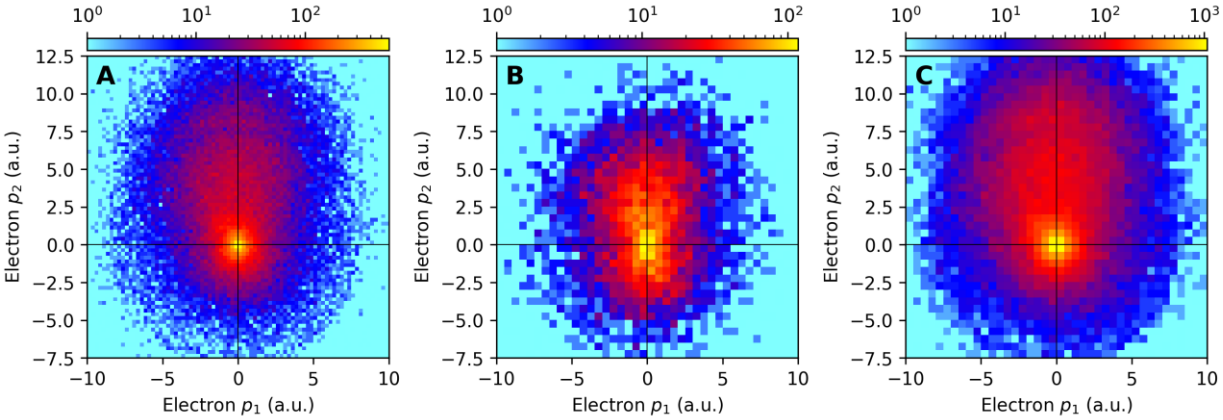

Fig. 2: Preliminary results for ionization of Ne by *K*-shell Compton scattering. (A)-(C): Electron momenta in a fixed-momentum-transfer reference frame. The momentum transfer points upward. (A): Integrated over all magnitudes Q. (B): 1.5 $a.u. < Q <$ 4.5 a.u. (C): 8 $a.u. < Q <$ 10 a.u.

Expected outcome: >2 publications after finalization of data analysis.
Required additional beamtime: Depending on our N_2 final results, one more beamtime on a diatomic molecule is necessary.