Report on the outcomes of a Short-Term Scientific Mission[1]

Action number: CA20129

Grantee name: Rebecca Ingle

Details of the STSM

Title: **First time observation of KLM Auger-Meitner process in organic chloro-compounds**

Start and end date: 10/10/2023 to 16/10/2023

Description of the work carried out during the STSM

Description of the activities carried out during the STSM. Any deviations from the initial working plan shall also be described in this section.

(max. 500 words)

During the experiment, we recorded a number of KLM, KMM and KLL Auger 2D maps across the pre-edge and to near-threshold region. We selected three samples CH_2Cl_2, $C_2H_4Cl_2$ and trans-CHCl=CHCl that had Cl in a variety of chemical environments. All samples were successfully characterised. Besides the 2D KLM maps, we XPS at both K-edges (Cl and C edges), and the Cl 2p.

The employed energies were in the range 3100-3500 eV for the X-ray characterisation and ~3200 eV for the 2D maps. The 2D KLM map example for dicholoroethane is provided in Figure 1.

[1] This report is submitted by the grantee to the Action MC for approval and for claiming payment of the awarded grant. The Grant Awarding Coordinator coordinates the evaluation of this report on behalf of the Action MC and instructs the GH for payment of the Grant.

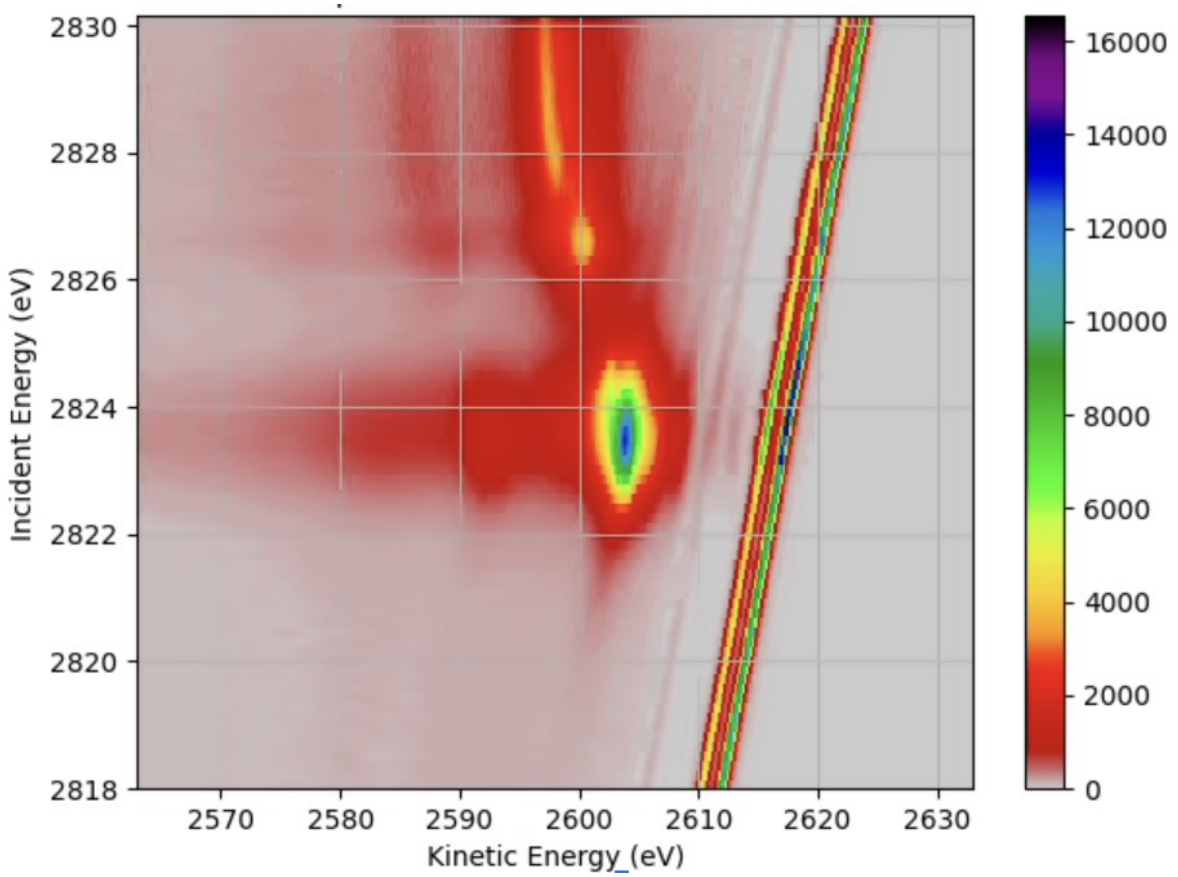

Figure 1: KLM 2D map of trans-dichloroethene. The intense region with an onset of 2823 corresponds to the main 1s->sigma* resonance.

We have observed varying peaks in the sample set showing molecular-like characteristics of the KLM decay which is due to differences in the valence band of the molecules (the M-shells are different).

Owing to the high flux of the beamline, it was also possible to record KMM maps for the three samples. This is a challenging experiment as the probabilities of the KMM transitions are only 10 % that of the KLM (which is already significantly weaker than other Auger processes. The KMM maps, however, exhibit a significant overlap with the valence band direct photoionization peaks and the signal subtraction may not be trivial. An example of the KMM 2D map for tdichloroethane is presented in Figure 2.

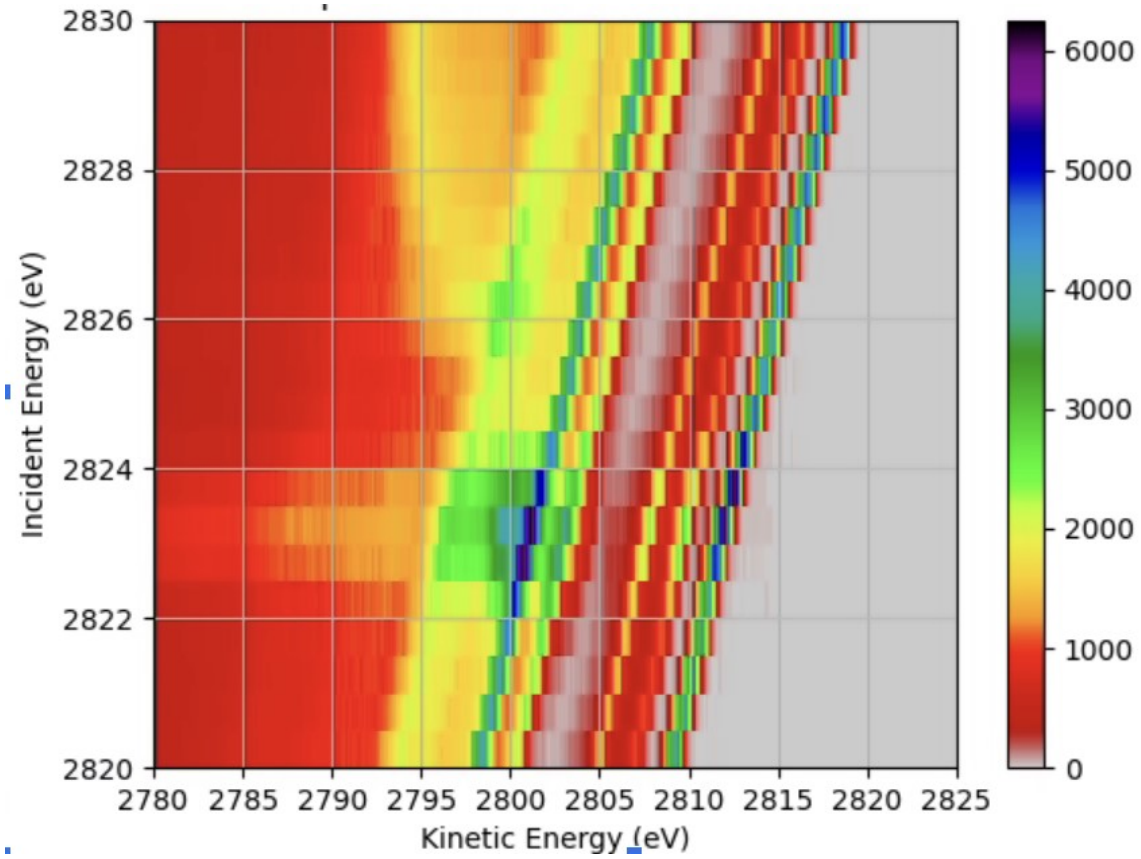

Figure 2: The KMM 2D map of dichloroethane across the main pre-edge absorption feature and into the ionisation continuum.

The obtained data require theoretical interpretation which will be performed by the applicant. Modelling of the KLM processes is challenging from different perspectives and will certainly require method some method development because the theoretical tools for core/excited and decaying states are still limited compared to the UV-Vis domain and in this project the decay involving both L and M-shells must be addressed. The theoretical activities performed by the applicant will involve X-ray absorption calculations, analysis and interpretation of the core-excited transitions, assessment of individual peaks in the KLM spectra. The measured spectra show additional complexities related to post collision interaction or possible induced ultrafast dynamics. During the beamtime, we also discussed the results within the team and planed for the interpretation of the results.

Description of the STSM main achievements and planned follow-up activities

Description and assessment of whether the STSM achieved its planned goals and expected outcomes, including specific contribution to Action objective and deliverables, or publications resulting from the STSM. Agreed plans for future follow-up collaborations shall also be described in this section.

(max. 500 words)

The main aims of the STSM were achieved, during the beamtime the full X-ray spectral characterization and KLM 2D maps have been recorded for the first time. The experiments were successfully conducted and we managed to measure 3 samples (instead of anticipated 2). We have obtained spectroscopic fingerprints of valence-band in the KLM. The resonances formed during the experiment will be furthermore theoretically modelled in terms of the Fano-Feschbach theory, the decay widths will be modelled most probably via on one-centre atomic radial Auger integrals using the multistate restricted-active-space perturbation theory.

The aims of the STSM fully comply with the action aims to acquire a fundamental-level understanding of Irradiation-Driven Chemistry (IDC). The full X-ray characterization and mainly information encoded in

resonant Auger spectra now recorded at unprecedented Cl 1s edge (only energetically accessible at very few beamlines worldwide) can be used to understand how the molecular chemical environment changes atomic-like. The STSM has significantly helped further the collaboration between Dr Eva Muchova and myself, with the group of Professor Mark Simon and Dr Denis Ceolin at the Galaxies beamline. For my professional development, it was an invaluable opportunity to get experience working in the tender X-ray regime, at a new beamline and work alongside experts in atomic physics to learn more about the irradiation driven processes in chemical systems following excitation in this range. Using the Auger processes to explore chemical systems also provides invaluable insights into the timescales of ionisation and electron rearrangement processes, as well as the mechanisms for the formation of highly charged ions, which is well aligned with the objectives or WP1.

The follow-up activities will involve post-processing of the obtained spectral data and their experimental characterization. The applicant will perform *ab initio* modelling and interpretation of the spectra using the theoretical toolbox, a further development is also anticipated. The KLM maps have proven to contain an the “chemical aspect and can be useful for further development of the method for spectroscopic characterization of dynamics in the excited or ionized states. Following on from this scientific exchange, we will hold regular meetings between myself and Eva Muchova for the project management as well as meetings for discussion with the wider collaboration and are in the process of planning an additional proposal application based on the results of this experiment.