INSTRUMENT DEVELOPMENT

HAXPES or easy photoemission

Using the HAXPES (hard X-ray photoemission spectroscopy) station on the GALAXIES beamline, it is possible to carry out high kinetic energy photoemission experiments on samples in the solid, liquid or gas phase (ref.1). The ability to analyze kinetic energies ranging from 10 eV to more than 10 keV makes this a unique experimental station at SOLEIL.

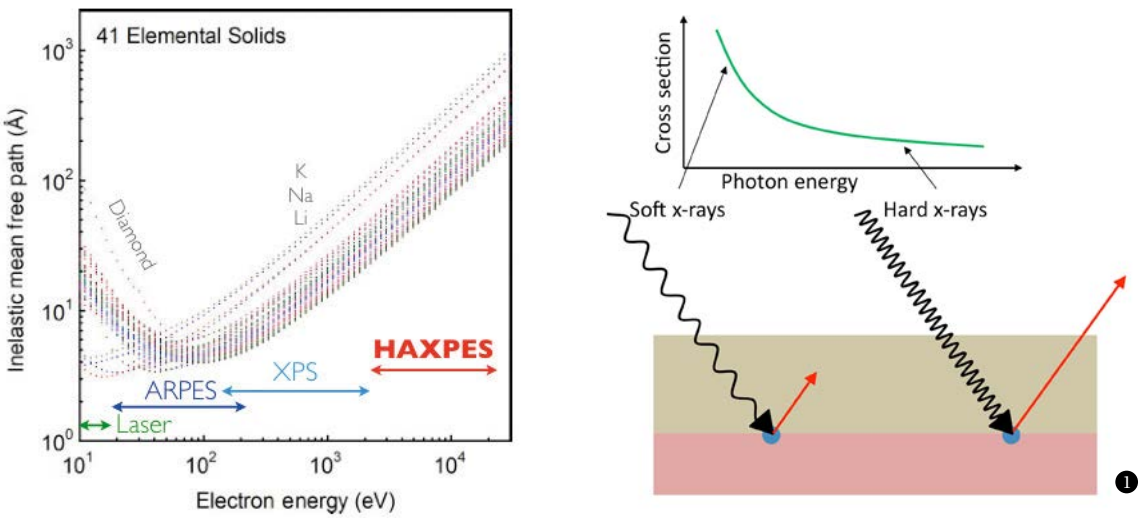


Figure 1:
On the left, the electron inelastic mean free path as a function of its kinetic energy. The high sensitivity of HAXPES in depth is illustrated on the right for a (red) buried interface. With soft X-rays, the electron excited at the interface (red arrow) cannot exit from the material because of insufficient kinetic energy. In contrast, at high photon energy, the electron has a high energy and is able to exit the material and be analyzed. The downside is lower ionization efficiency (green curve).

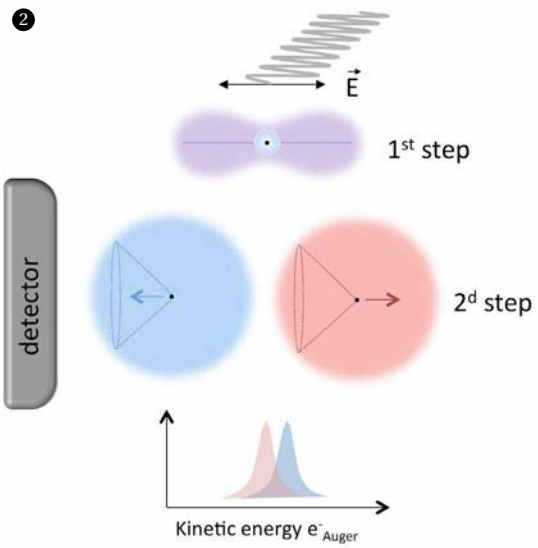
There are several advantages in the use of high energy. First, the energy analysis of the emitted electrons gives specific information about the ionized element and its chemical environment. At high energies, it is possible to reach deeper electronic shells which relaxation is very fast ($t < 10^{-15}$ s). On the other hand, due to the high kinetic energy of the electrons, it is possible to probe about 10 times deeper into the material than with standard photoemission (VUV and soft X-rays), which opens the way to the study of interfaces, buried layers or ion mobility in liquids. HAXPES is generally much less sensitive to the surface condition of materials than standard photoemission techniques and requires no special treatment of the surface. It is particularly well suited for applied materials (multi-layers, buried interfaces and functional materials).

Seeing the movement of atoms

In the case of an isolated atom, ejecting an electron by giving it high kinetic energy leads to nuclear displacement, i.e. a recoil effect. This phenomenon is difficult to identify because of the large mass difference between the electron and the nucleus. To induce a strong effect, the photoelectron must have a high kinetic energy during ejection. At high energies, however, the spectral resolution decreases as well as the effective ionization cross sections, making it difficult to measure. An alternative way is to measure the recoil effect not directly on the photoelectron spectrum, but on the Auger spectrum, which does not depend on the incident energy bandwidth. On the GALAXIES beamline, the high photon flux and large angular acceptance of the electron spectrometer can compensate for the reduction in effective ionization cross-section. These parameters allowed us to make measurements of Auger spectra of ionized neon 1s shells and show that the recoil can be big enough for a separation of Auger lines to be possible. The observed process, equivalent to a Doppler effect, showed that the dependence of the Auger effect on the photon energy was possible because of the nuclear movement (ref.2).

HARPES or bulk band structure

The properties of materials are largely based on their electronic structure. The method often used to visu-



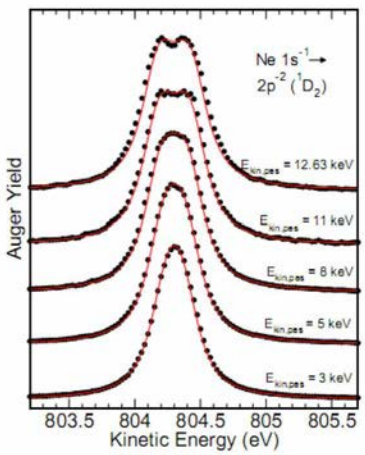


Figure 2: In the first step, an electron from the Neon 1s shell is ejected with a cos² angular distribution with respect to the polarization vector (shown in purple). The Ne⁺ ion recoils in the opposite direction. Auger relaxation occurs in a second step as a result of this ion motion. Auger electrons are emitted (1) isotropically (blue and red clouds) and (2) collected necessarily towards the detector (cone in the figure). Those emitted from the ion whose pulse is directed at the detector (blue arrow) are measured as being faster than the electrons emitted in the opposite direction (red arrow). The experimental results (dots) and numerical simulations (red) are shown on the right for different kinetic energies of Ne1s photoelectron (ref.2).

alize this structure is called ARPES (angle-resolved photoemission spectroscopy). This combines the kinetic energy detection of photo-emitted electrons with emission direction measurements (the electron momentum). When both kinds of information are combined, it is possible to reconstruct the electronic structure.

The development of ARPES is important for the study of fundamental physical phenomena such as electronic correlations, superconductivity or topological insulators. Measurements are traditionally performed with low energy photons to benefit from the high energy and high time resolution and greater cross section. However, this requires having perfectly clean surfaces which limits the method to mostly two-dimensional samples. Moreover, even in ideal cases, ARPES ultimately only provides a picture of the surface electronic properties due to the very short mean free path and not of the bulk state.

Recent studies have shown that it is possible to perform these measurements with high kinetic energy, paving the way for the characterization of the bulk electronic structure of materials, using HARPES (hard x-ray angle-resolved photoemission spectroscopy), based on the traditional technique. The first measurements on the GALAXIES beamline (see Figure 3) show the feasibility of the method and pave the way to new applications, such as the study of the electronic structure of buried layers or interfaces.

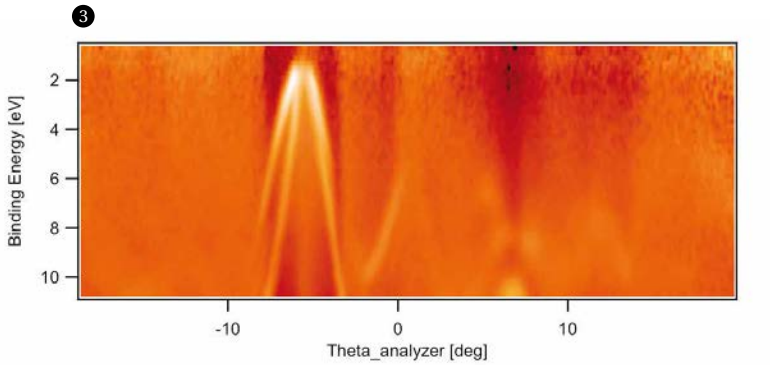


Figure 3: High kinetic energy band structure or HARPES in doped diamond obtained on the GALAXIES beamline. The measurements were performed at 15 K with incident photons of 2.5 keV (ref.3).

Conclusion

The HAXPES station on the GALAXIES beamline has extended the photoemission field to high kinetic energy. This method allows materials to be probed in depth, providing so far unexplored photoemission opportunities to study materials and diluted (gaseous, liquid) phases. The reduced surface sensitivity makes HAXPES measurements easy to perform.

→ **Contacts :**
denis.ceolin@synchrotron-soleil.fr ;
jean-pascal.rueff@synchrotron-soleil.fr ;
marc.simon@upmc.fr

References:
[1] D. Céolin et al. J. Electr. Spectr. and Relat. Phenom. (accepted)
[2] M. Simon et al. Phys. Rev. Lett. (submitted)
[3] C. Fadley et al., in preparation