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Report:

The aim of this project was to analyse the surface diffusion at very low temperature of *3d* metal adatoms on a surface by using X-ray Magnetic Circular Dichroism (XMCD). The surface diffusion of adatoms is a key parameter that governs important phenomena such as chemical reactions or island growth on surfaces. For example, the morphology of growing islands, on which depend many important catalytic, electronic, or magnetic properties, is essentially determined by surface diffusion kinetics.[1]

Surface diffusion is commonly driven by thermally activated processes, and the diffusion jump rate is given by an Arrhenius law. The activation energies are element and surface orientation dependent, and vary typically in the range of several tens of meV to several hundreds of meV. Consequently, at very low temperatures (below 10 K - 20 K), no atomic motion is allowed by classical diffusion theory.

However, in this temperature range, the de Broglie wavelength of a *3d* metal adatom becomes comparable to the site separation, and tunneling diffusion should become possible. In a previous project (HE-1231) we noticed that even below 10 K Cr adatom probably move on the surface in a timescale of minutes, on the basis of the XMCD signal shape time evolution.

In this project we wanted to specifically address this low temperature diffusion problem. It is probably the first time that XMCD was used to determine surface diffusion coefficients of adatoms.

A collection of single adatoms was obtained by low-temperature deposition of small amounts of matter (in the order of 0.5 % to 1% of a monolayer). Due to a high sensitivity to coordination, the XMCD spectrum of isolated adatoms can be distinguished from the one of very small clusters (several atoms only)[2].

Here we chose to deposit Cr adatoms, since from our experience it revealed to be the best candidate for such type of experiments (clearly visible differences between single atoms and clusters)

Chromium was deposited at the lowest achievable temperature (~6 K) directly in the cryo-magnet chamber on various single-crystal substrates, namely Cu(111), Au(111), Cu(100), Ag(100), prepared in a LEED-STM connected UHV chamber. Changing substrates allows us to vary the activation energies. XMCD spectra were recorded as a function of time at different temperatures from about 6 K to 150 K.

Figure 1 shows the typical evolution of the Cr XMCD spectrum at the $L_{2,3}$ edges on Cu(111). The spectrum at a given time is decomposed as a linear combination of a single adatom spectum and a cluster

pectrum. From this decomposition, we deduce the proportion of isolated adatom. Figure 2 shows the evolution of the ratio of the number of single Cr adatoms to the total number of adatoms as a function of time for two different surfaces. One sees that the evolution is faster on Cu(111) than on Cu(100).

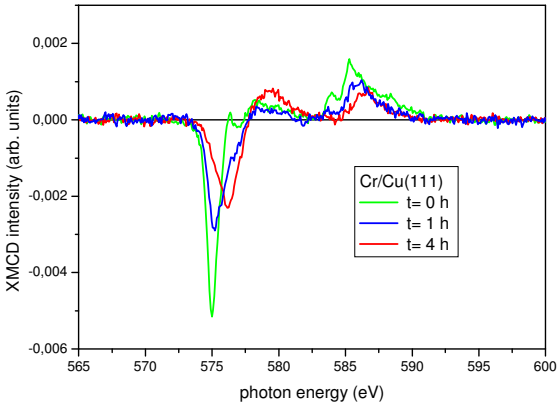

Figure 1

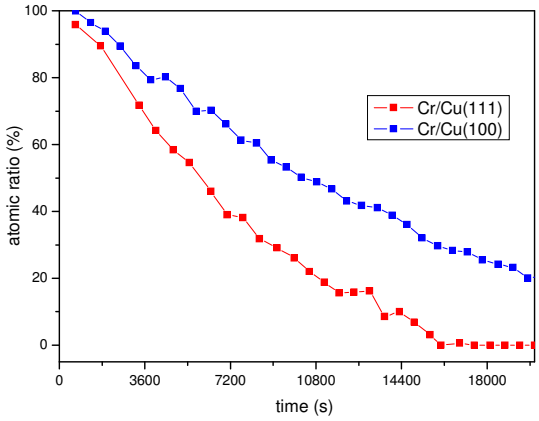

Figure 2

We tested that this evolution occurs also without beam and without magnetic field, by waiting for a while before operating the first measurement. We also tested if the XMCD signal shape evolution is produced by contamination. Therefore, we purposely contaminated the sample with CO in the 5.10^{-9} mbar to 1.10^{-8} mbar pressure range for several minutes (the base pressure was below 1.10^{-10} mbar). Figure 3 shows different CO-contaminated spectra obtained in different ways and a spectrum recorded after several hours evolution (bottom). Clear differences can be noticed. The top spectrum resembles to a Cr in a Cr_2O_3 environment, and the middle one possibly to a CrO_2 configuration, but both are different from the uncontaminated evoluted spectrum.[3,4]

The quantitative determination of diffusion coefficients remains to be done via Monte Carlo simulations, but one can say that at present time that low temperature surface diffusion exists in a temperature range where thermal activated diffusion is forbidden. Hence, we confirm quantum tunnelling adatom surface diffusion.[5]

One must emphasise that these experiments were only possible at ESRF because of the very high beam stability and the extremely efficient data collection: indeed an XMCD spectrum can be obtained in less than 5 minutes even for such small amounts of material, and this is unique in Europe.

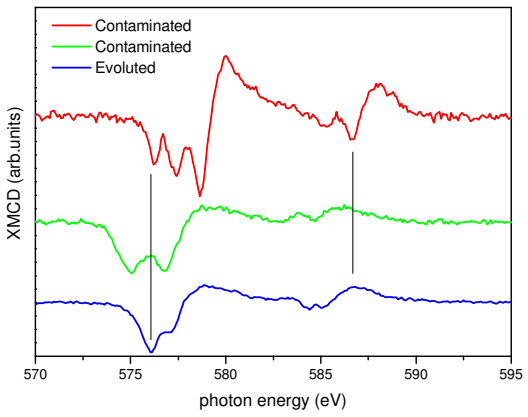

Figure 3

[1] H. Brune, Surf. Sci. Rep. **31**, 121 (1998).
[2] C. Boeglin et al. Phys. Stat Sol. (b) **242**, 1775 (2005)
[3] E. Goering et al. Appl. Phys. A **78**, 855 (2004)
[4] E. Gaudry, Ph.D. thesis of Université Pierre et Marie Curie, 2004.
[5] P. Ohresser et al. Phys. Rev. Lett. sumbmitted