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Brilliant, Pulsed Coherent X-Rays - Manifold Ways to Follow Moving Atoms

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Synchrotron radiation has many intriguing virtues, some of them paving the way for new approaches to diffusion.

The brilliance of 3rd generation synchrotrons is many orders of magnitude higher than that of the world's best X-ray facilities, therefore even the extreme narrowness of nuclear levels poses no hindrance for resonant absorption. Different from the well-known Mössbauer effect we can observe the re-emission directly in time, because the synchrotron beam is pulsed with a pulse length of about hundred picoseconds. The method is called nuclear resonance scattering of synchrotron radiation (NRS) [1]. It permits studying the atomisticity of diffusion directly in time (see *e.g.* [2]). The high brilliance of the beam allows even grazing incidence measurements of the dynamics of surface monolayers [3,4]. The method is until now restricted to iron diffusion.

We aimed at studying the structure of an iron monolayer on W(110) by nuclear resonance scattering of X-rays. We succeeded to grow a nearly perfect iron monolayer as proven by LEED and by NRS spectra, both taken at room temperature. When increasing the temperature an increasing fraction of the iron atoms feels the appearance of defects, *i.e.*, an asymmetry of the neighbourhood. The defects cause an additional electric field gradient (in addition to the electric field gradient due to the two-dimensional nature of the monolayer) leading to a beat structure of the NRS spectra (Fig. 1). The beat increases in amplitude with increasing temperature due to the creation of lattice defects, probably vacancies, and the beat structure gets smeared due to the incipient motion of the defects. The defects must be thermally activated since they disappear when returning to room temperature.

From the amplitude of the beat we deduce the vacancy concentration, and from its temperature dependence the vacancy formation energy 0.17(5) eV. From the broadening of the beat, with less accuracy, we deduce for the vacancy migration energy 0.16(6) eV, resulting in 0.33(11) eV for the activation energy of diffusion.

The coherence of synchrotron radiation – though until now only partial – permits still another access to studying diffusion: X-ray photon correlation spectroscopy (XPCS) [5,6] permits following the diffusion of nearly any atom and at very low diffusivities [7,8]. We have shown how by monitoring the spatial and temporal variations of the scattered coherent X-ray intensity the diffusion of single atoms can be studied. This was demonstrated for the intermetallic alloy $Cu_{90}Au_{10}$. By measuring along several directions in reciprocal space, we can elucidate the dynamical behaviour of single atoms as a function of their neighbourhood. This method does not rely on specific atomic species or isotopes and

can thus be applied to almost any system. Thus, given the advent of the next-generation X-ray sources, XPCS has the potential to become the main method for quantitatively understanding diffusion on the atomic scale.

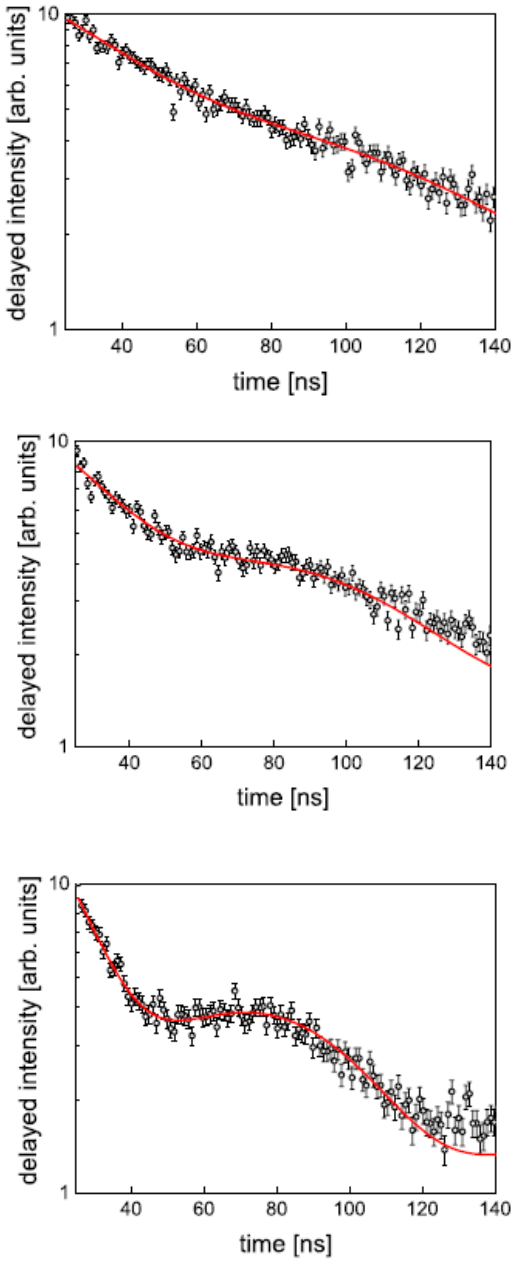

Fig. 1 Nuclear resonance scattering at an iron monolayer on W(110) at three different temperatures: room temperature, 570K and 770K.

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