Study of atomic motion in single-crystal metals

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The motion of atoms in single-crystal metals is studied by the observation of the Doppler-broadened line-structure formed when the nuclei decay while recoiling. The recoil is imparted to the nucleus by the emission of gamma rays following thermal neutron-capture, the so-called (n,γ) reaction. The high-resolution gamma-ray spectrometer GAMS4 and molecular dynamics simulation of the slowing down of atoms in oriented crystals provide information on interatomic potentials. Three transition metals (Ni, Fe, Cr) were studied and two nuclear levels in each element were analysed.

Numerous interatomic potentials exist in the literature and there are many ways to elaborate them. Potentials for metals can be constructed from equilibrium properties using the embedded atom method [1], where they give fairly good results. These potentials are defined by two terms: an embedded function which is correlated to the force needed to embed an atom in the electron density caused by the neighbouring atoms, and a core repulsive term which describes the behaviour of atoms at close separations. Besides the embedded atom method potentials other theories like the BM [2], defined by an exponential function, or the ZBL [3], based on a coulomb screened function, are calculated for very small interatomic separation.

The Crystal-GRID [4] technique based on the observation of Doppler shifts produced by the motion of radioactive atoms in the solid state fills in the missing energy gap. This technique measures the Doppler energy of the second gamma-ray emission in a two gamma cascade. After neutron capture, the newly formed isotope decays down to the ground state by emission of a sequence of gamma rays. The emission of the first gamma ray, induces a recoil to the atom and the later will start to move inside the crystal with a kinetic energy depending on the difference between the capture state and the energy of the nuclear state populated. When the atom is still moving while it emits a second gamma ray, the observed gamma-ray energy will be Doppler shifted. Interatomic potentials, which govern the slowing down, can now be tested in the recoil energy domain by comparing the line shape measured with the double-flat crystal-spectrometer [5] and the one simulated with different interatomic potentials. The Doppler line-structure depends on three terms: i) the lifetime of the nuclear state populated, which gives the probability emission of the second gamma ray, ii) the orientation of the crystal, because blocking and channelling of the recoiling atom due the ordered structure of the crystal influence the trajectories and velocities, and finally iii) the interatomic potential which determines the slowing down and the motion of the atom recoiling inside the crystal.

For each transition metal, almost 10 different potentials were tested with 2 different crystal orientations. Figure 1 shows the crystalline structure of the metals and the directions of observation of the second gamma-rays. The slowing down theories are always simulated by molecular-dynamics simulation which calculate the trajectories and velocities of the recoiling atom in a molecular-dynamics cell of about 1000 atoms by solving the Newtonian equations of motion for each individual atom. From the stored value of the trajectories and velocities, a line shape can be reconstructed assuming a lifetime value. The simulated line structure is then fitted to the measured one leaving the nuclear state lifetime as a free parameter. Figure 2 shows the simulated trajectories calculated by molecular dynamics simulations [6] and the fitted and measured line shape obtained for chromium in the two different orientations. The analysis results in two parameters. One is the lifetime of the nuclear state under study, and the other is a χ^2 per degree of freeedom which gives information on the agreement between the simulated and measured line structure. A perfect match should return a value of 1. We found that among the listed potentials, very few were able to reproduce the measured line structure, and the lifetime of the nuclear state. This demonstrates that many of the tested potentials are not good candidates for the slowing down of atoms in metals at higher energies. Figure 3 shows for the best potentials the values of the lifetime fitted as well as the range in which the different nuclear state lifetimes should be according to the known value found in the literature. The best potential which

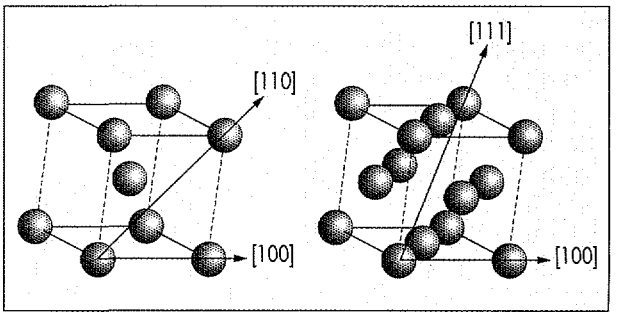

Figure 1: *Direction of observation for the 2 crystalline structures used for metals. Nickel has a fcc structure shown on the right, Fe and Cr have a bcc structure.*

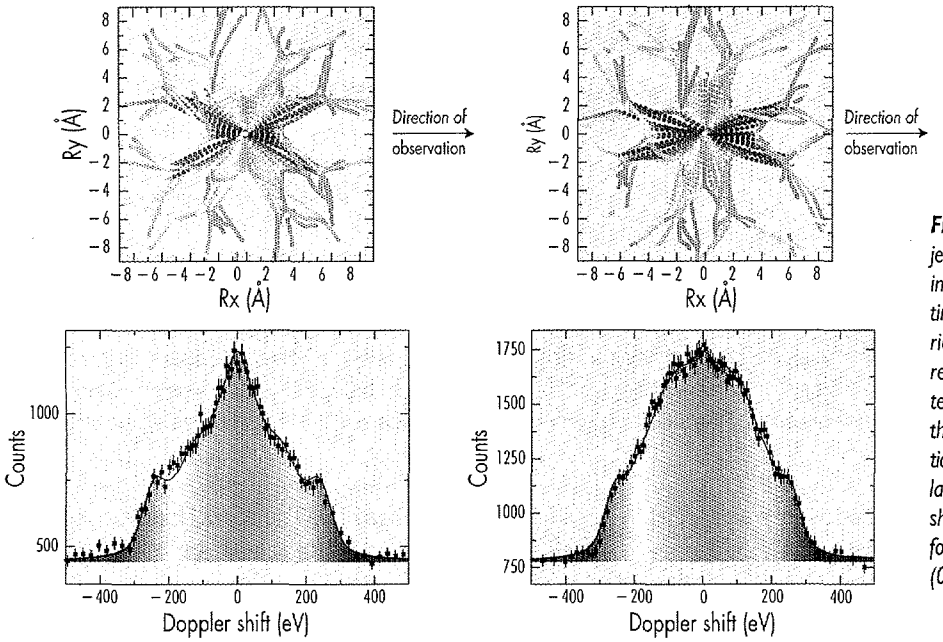

Figure 2: *Molecular dynamics simulated trajectories of 150 Cr atoms (^{24}Cr, bcc crystals) in the [100] (left) and [110] (right) plane. The time difference between dots of the trajectories represents 1 fs. The colour represents the red/blue energy shift of the gamma ray emitted, obtained by the scalar product between the velocity of the recoiling atom and the direction of observation. The bottom shows the simulated and measured Doppler-broadened lineshape obtained with the EAMWB potential for the two orientations for a lifetime of 14.72 (0.53) fs.*

is able to reproduce the known lifetime and which gives the smallest χ^2 per degree of freedom is the embedded atom method potential developped by Voter and Chen (EAMVC) [10] for nickel and iron. For chromium, the best slowing down theory is the one derived by Wang and Boerker (EAMWB) [13] using the embedded atom method. Another nuclear state level with a higher lifetime value was also analysed for each transition metals and the same behaviour occurs. The crystal-GRID measurements permit also the construction of new potentials. Some of the parameters of the ZBL potential [3] were modified and fitted in order to have the best χ^2 value by comparing the simulated and measured line shape. The new potentials have the same shape as the best potentials which were selected by the first analysis. This confirms our choice in the selection of the best possible potential. The present experiment opens the way to a systematic study of interatomic potentials in alloys following a building up principle. This study will be pursued in the next years and might lead in combination with molecular dynamics simulations to a better understanding of these technological important materials.

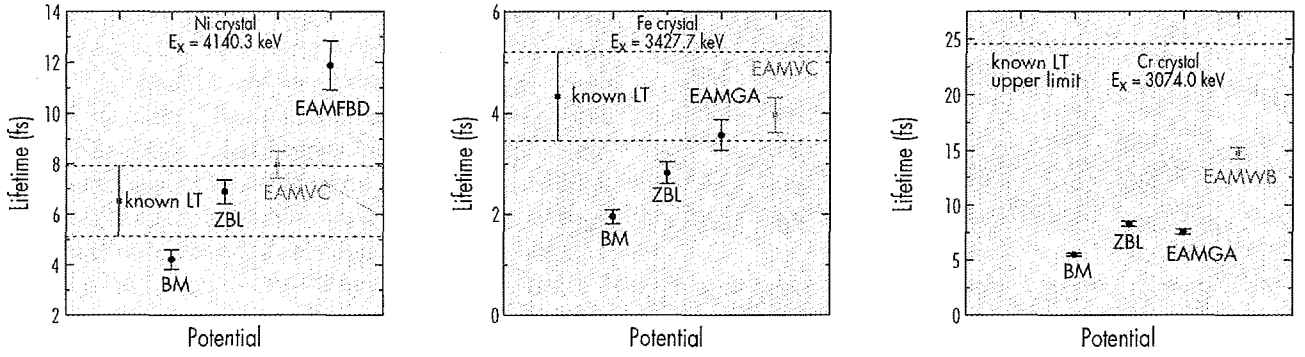

Figure 3: *Fitted lifetime (LT) for the three transitions metals. Also shown is the known lifetime taken from the literature [7], [8] and [9]. For the name of the potential see the references: BM [2], ZBL [3], EAMVC [10], EAMFBD [11], EAMGA [12], EAMWB [13].*

References

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