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Demixing a melange: Transient interface sharpening in miscible alloys

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Diffusion belongs to our everydays' life experience. When you carefully pour milk on your black coffee that you get a coffee and a milk layer and wait long enough, the two liquids will mix „automatically" by diffusion and you obtain a nice medium brown melange. It is now cold and to enjoy your melange you will probably have to heat it again. Diffusion processes always take place when two (or more) substances – even solids – share an interface. In solids these processes are not a major problem as long as the material layers are big and bulky but with continuously shrinking structures in microdevices such as microprocessors and read-write-heads atomic interdiffusion can cause serious problems, e.g. destruction of the device and reduced life-time.

Computer simulations have shown a new diffusion behaviour on nanoscale [1, 2]. For strongly composition-dependent diffusion coefficients, an initially diffuse A/B interface can sharpen even in ideal systems with complete mutual solubility. This sharpening is surprising at first sight because the direction of diffusion is always opposite to the direction of the composition gradient: $J = -D\,\mathrm{grad}\,c$, with J the atomic flux, D the diffusion coefficient and c the concentration – that is why you never get your black coffee back. Indeed, for constant D, the composition profile will gradually decay and a flattening of the (sharp or diffuse) interface is expected (Fig. 1a). On the other hand, when the diffusion coefficient strongly depends on the local composition, the flux distribution can lead to a sharpening of the interface (Fig. 1b, Fig. 2). The sharpening can be qualitatively predicted from the classical Fick I law although it is not able to provide correct kinetics on nanoscale [1, 4].

We studied Mo/V multilayers because they are completely miscible. Structures of 20 Mo/V bilayers with a modulation length ≈5-6 nm were produced by magnetron sputtering. The layers were separated by a roughly 1 nm thick diffuse interface with a constant composition gradient (Fig. 3). In order to follow the change of the composition profiles in situ during heat treatments we used X-ray diffraction measurements at the KMC2 beamline. The samples were placed on a heater inside a hemispherical Be window under high-vacuum conditions. Diffraction experiments were carried out at temperatures that gradually increased from 293 K up to about 973 K in 10 steps. At each temperature consecutive symmetrical scans between 53° and 66° of the scattering angle 2Θ were performed, measuring the scattering intensity around the (002) Bragg reflection of the Mo/V multilayer structure. The sample was kept at every temperature until no change in the diffraction pattern could be observed.
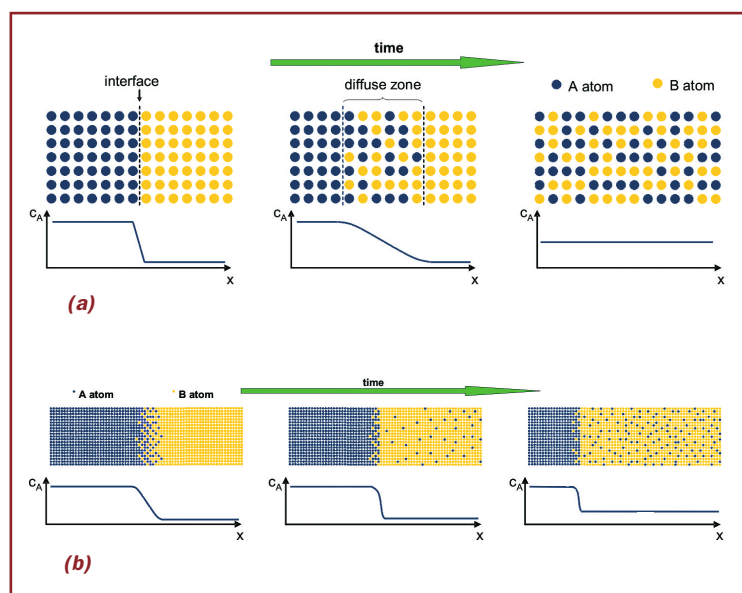
References:

[1] Z. Erdélyi et al., Phys. Rev. Lett. **89**, 165901 (2002)

[2] Z. Erdélyi et al., Phys. Rev. B **68**, 092102 (2003)

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Fig. 1: Scheme of the intermixing in case of composition (a) independent and (b) dependent D.
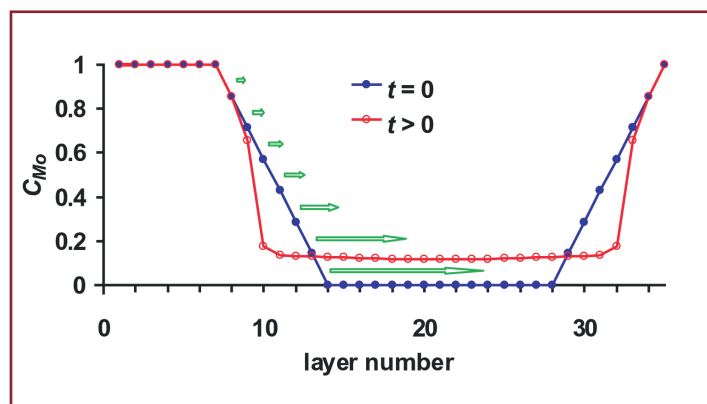
From the reconstruction of the diffraction patterns (Fig. 4) we found an interface sharpening during the heat treatment. This conclusion is also supported by the analysis of changes of the intensity ratios during heat treatment. A further proof of sharpening is due to the fact, that at a fixed temperature no more changes were observed after a certain time, i.e. a gradually increasing temperature was necessary. Since the interface is sharper, the Mo atoms are bound more strongly in the interface (the interface becomes more and more Mo-rich), consequently their diffusion into the V is slower (Fig. 2). Thus, in order to counterbalance this effect, we had to increase the temperature slightly (the diffusivity has exponential dependence on both the temperature and the composition).
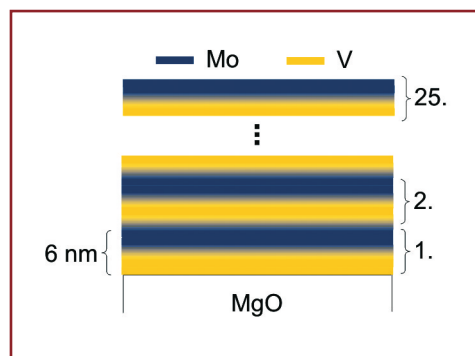
The thickness of the Mo layers did not change apart from a tiny increase caused by thermal expansion. In contrast, the V-rich layers became much thicker, which cannot be explained solely by thermal expansion. The interface thicknesses decreased by about a factor of two (from 1.7 and 1.4 nm, respectively, to 0.78 nm), confirming the sharpening effect.

Our studies provide the first experimental evidence for the predicted interface sharpening. This behaviour is not only interesting to fundamental research. It could provide a useful tool for the improvement of interfaces and offer a way for fabrication of better X-ray or neutron mirrors, microelectronic devices or multilayers with giant magnetic resistance.


Fig. 2:
Composition distribution during intermixing in one period of a Mo/V multilayer calculated from an atomistic model, as in [1]. The arrows represent schematically the ‚flux distribution', i.e. their lengths are proportional to the absolute value of the atomic flux.

Fig. 3:
Scheme of the multilayer sample.

Fig. 4:
Diffraction patterns measured (circles) during the heat treatments and their reconstructions by the modified Stearns model (solid line): (a) room temperature, (b) 903 K, and (c) 953 K.
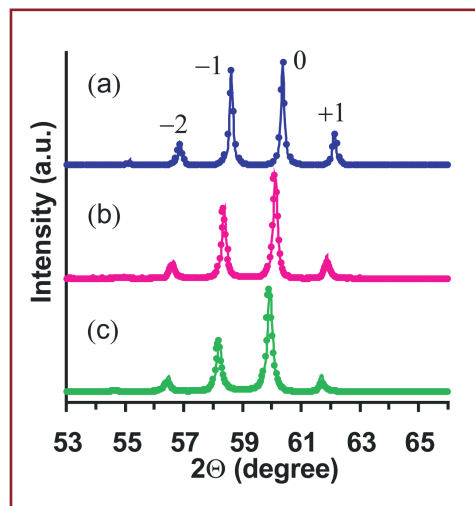
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