Diffusion in Mo/Si based multilayers, a story from 77K to 770K

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Abstract:

Diffusion behaviour at the interfaces of multilayered thin film structures generally leads to intermixing and subsequent compound formation. For multilayer systems based on the Mo/Si material combination, it is well known that diffusion induced silicide formation leads to a loss of optical contrast as well as a change in the reflected wavelength. Often, diffusion barriers are applied to reduce diffusion speeds, slowing down, but not stopping the deterioration of these systems at elevated temperatures. This paper intends to provide an overview of in-depth diffusion analysis in several state-of-the-art multilayer systems.

We report here on a method of increasing the optical contrast between thin films, as applied to the Mo/Si system. Using physical vapour deposition, multilayer structures were grown on Si substrates mounted on an actively (liquid-N_2) cooled substrate holder. The low energy of the deposited atoms, combined with the reduced substrate temperature, leads to a significantly reduced interaction between Mo and Si. The first experiments reported here show a reduction of the total silicide interlayer at both interfaces by 0.9 nm. This reduced interface width was maintained upon subsequent warming up of the samples to room temperature, suggesting that the atoms deposited in the interaction zones are “frozen in” during subsequent deposition of atoms. This meta-stable structure is destroyed at elevated temperatures, where diffusion speeds up and a strong increase in the silicide interface width is observed.

To study the diffusion kinetics at elevated temperatures in more detail, we developed a novel method based on Grazing Incidence X-ray Reflectometry (GIXR). Using an *in-situ* analysis of the GIXR data during thermal annealing, it is possible to determine multilayer period changes with a sub-pm accuracy. This method enables determination of the diffusion constants in temperature ranges that are inaccessible by means of traditional diffusion studies. The data show that already at 100°C, there is a small but observable diffusion induced change in the Mo/Si multilayer period. An Arrhenius-type plot of the obtained diffusion constants shows that diffusion in the 100°C-275°C temperature range can be described with a single proportionality factor and a single activation energy, signifying a single physical process to be at the base of interface diffusion.

Finally, well above 300°C and depending on the exact structure and application of diffusion barriers, a strong acceleration of the diffusion is generally observed. Using low energy ion scattering (LEIS), we investigated diffusion of Mo and Si atoms through a diffusion barrier at 500°C. The LEIS data show several distinct diffusion regions and reveal an instantaneous acceleration of the diffusion rate by an order of magnitude. X-ray photoemission spectroscopy and transmission electron microscopy analysis clearly show that it is not the “chemical breakdown” of the applied diffusion barrier, but crystallization of the interface region that leads to the observed dramatic increase in diffusion rate.

The research presented here is particularly relevant for coatings applied at high fluency photon sources such as next-generation pulsed EUV sources and X-ray Free Electron Lasers.