Report on the manuscript entitled :

« Methods for growth experiments at ALOISA"

presented by Francesco Bruno

Francesco Bruno has performed his PhD work during the first three years of operation of the ALOISA beamline. This specific situation lead him to conduct his activity along two main lines. The first one, with a methodological coloration, aimed at clearly defining the significant ingredients to be put in data analysis, in the unique context of the ALOISA experimental station, particularly for photoelectron and X–Ray diffraction measurements. The second aspect is concerned with an original research on the Pb/Ge(001) interface, fully anchored on the main field of activity of the local group namely the physics of growth and erosion processes at surfaces and interfaces.

The manuscript is organized as follows : the first three chapters introduce the general purpose of the work (chap. I), the specific features of the ALOISA beamline and experimental station on ELETTRA (chap.II) the main interests and open questions about the two systems under study : Fe/Cu_3Au and Pb/Ge (chap.III). The fourth chapter is dedicated to the in–situ assessment of the sample surface morphology and thickness with a special emphasis on the methods and corrections factors. The fifth chapter deals with the structure determination of growing films and the last one presents the latest results obtained on the Pb/Ge(001) interface in the early stages of growth.

The main contribution of Francesco Bruno to the methodological aspect is developed in chapter IV and is concerned with the extraction of pertinent information on the sample surface morphology, thickness and structure from the raw experimental photoelectron spectroscopy and diffraction data. The physical basis of each process is clearly discussed together with the geometrical parameters specific of the ALOISA station. The output is a rational strategy for data analysis which is fully exploited in the following chapter on the example of Fe/Cu_3Au and is now extremely useful for the users of the beamline. In the same fourth chapter, Francesco Bruno gives a detailed presentation of the X–ray specular reflectivity method with the emphasis on the unique information obtained on surface roughness and growth mechanism by in–situ measurements performed in real time. Simulations of the expected reflectivity curves are presented, helping to better apprehend the influence of each parameter. Francesco Bruno has achieved a thorough literature search on the subject and demonstrated his deep understanding of the problems, he has made available the necessary tools to address properly the mechanisms of homo and heteroepitaxial growth on the ALOISA experimental station.

To go beyond mere surface morphology and have an insight on the atomic structure X–ray and(or) photoelectron diffraction methods are needed. Chapter V illustrates the potentialities of the beamline on the particular example of the Fe/Cu_3Au(001) interface. The complementarity of both methods is clearly evidenced since X–ray diffraction under grazing incidence and exit is unique to assess the in–plane lattice mismatch and orientational relationship between epilayer and substrate whereas photoelectron diffraction is particularly sensitive to the out–of–plane bond distance and angle around a given element. For these experiments, Francesco Bruno has developed a specific code for sample and detector positioning since the ALOISA « diffractometer » does not correspond to any standard setting for which codes are already available. Moreover, the easy photon energy scan ability, seldom achieved on surface diffraction beamlines, enables a reciprocal space exploration closer to the LEED scheme than to the usual crystallography angular scans and new software for data collection and correction had to be implemented. The application of this conjunction of methods to the Fe/Cu_3Au growth has provided a clear understanding of the initial fcc stage and evidence for Au surface segregation. An accurate determination of the parameters of the strained bcc structure presented by thicker deposits has been obtained.

The last chapter presents an original work on the Pb/Ge(001) interface growth pertaining to the scientific program of the beamline team. Previous results obtained by helium scattering had brought the evidence of quantum size effects in thin layers resulting in an oscillatory variation of the step height. However helium scattering can only reveal phase shifts between waves scattered by adjacent terraces and is not sensitive to the actual interlayer spacing within the thin film. On the contrary, X–ray reflectivity and diffraction rod analysis can unambiguously provide this information. A large corpus of independent reflectivity data collected at different photon energies during the growth of thin films up to an equivalent of about 20 monolayers (ML) has revealed two growth regimes : above 5ML deposited, a good fit of the experimental intensity is obtained in the assuption of Pb(111) layer by layer growth with adjustment of the interlayer diffusion coefficient and interface distance between Ge and Pb. Below 5ML, this mechanism does not hold : the film structure has been determined by fitting the Pb diffraction rod $2\bar{2}\,l$ in the assuption of a free standing (111) film adjusting each interlayer spacing and occupancy. A convincing model showing an oscillatory behaviour of the interlayer spacings is proposed for a 5ML deposit. For successive deposits, the data analysis show alternately contracted and expanded top interlayer spacings in agreement with the oscillatory step height found in the helium scattering experiment.

In conclusion, the work presented by Francesco Bruno in this manuscript shows a good balance between development of analysis methods enabling to take a full advantage of the ALOISA beamline capabilities and fundamental research on the very important topic of size effect in the ground state of solid materials. I consider this work at the required level for a PhD thesis.

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