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Diagnostics of films and layers of nanometer thickness using middle energy ion scattering technique

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The paper presents a short description of thin film parameter study technique based on the analysis of energy and angular spectra of ions, back scattered from the film, with initial energies in 50−250 keV range as well as on the registration of characteristic X-ray radiation induced in the film under study by the probing ion beam. The technique is nondestructive one and makes it possible to determine geometrical parameters of the film, element composition and its change with depth, crystal structure quality of the film as a whole and of sublattices of definite elements. Some examples are given on this technique application for the study of multicomponent film structures at different stages of their growth.

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1. Introduction

Among ion beam diagnostics of solid film materials, the very universal one is light ion Rutherford back scattering (RBS). The traditional He^+ ion beam energy of 1−4 MeV is convenient for the investigation of films (layers) having the thicknesses from parts of microns up to several microns. The development of modern microelectronics tends to the use of more and more thin films and layers up to a few of nanometers in thickness. For the investigation of such objects, the application of ion beams with energies from tens to hundreds keV seems to be preferable.

Middle Energy Ion Scattering (MEIS), being the RBS modification, has both advantages and shortcomings when compared with MeV energy RBS. The advantages are large back scattering cross section values which depend on beam energy E as $1/E^2$ making it possible to provide two orders back scattered ion flux increase, as well as smaller energy spread (straggling) values, which decreases the lowering of resolution with depth (thickness) growth. The main technical advantage of MEIS is the possibility to use high-aperture electrostatic analyzers (ESA) for back scattered ion energy analysis which makes it possible to achieve the depth resolution better then one nanometer in estimation of geometric parameters, element composition and defects of the sample under investigation [1–3].

The main MEIS shortcoming is mass resolution decrease, which can be partly overcome by means of heavier probing ions (N^+) use, but, according to the existing experience, the thicknesses in this case should be limited to 10−20 nm.

In MEIS, as well as in RBS, the factor of vital importance is the computer simulation of back scattered ions energy spectra and their comparison with experimentally obtained ones, but, in contrast to RBS, MEIS requires taking into consideration some factors, that in RBS can be neglected. These factors are: the charge state distribution in the flux of back scattered particles (for ESA measurements), the deviation of back scattering cross section from the Rutherford one, energy dependence of straggling. Taking into account of these factors requires the creation of special database and, in cases of data absence or their insufficient accuracy, the experimental obtaining of necessary data.

At the use of MEIS for investigations of film samples having the crystalline structure the back scattered ion energy spectra can be measured in two regimes: „random“ orientation, when the angle between probing ion beam direction and nearest low-index crystalline direction θ exceeds the critical channeling angle θ_0 (usually $\theta_0 \approx 2{-}3^\circ$), and aligned orientation when these two directions coincide. The comparison of these two spectrum types together with dependences of back scattered particles yield on θ (so called angular scans, AS) makes it possible to draw a conclusion about the quantity and type of such crystalline structure defects as point defects, texture, inclusion of polycrystalline or amorphous phases [4]. Thus, in case of point defects, the dependence of backscattered ions yield in aligned regime would differ from the case of presence of continuous defects, such as texture or polycrystalline phase inclusions. For point defects the distribution of their concentration on depth can be obtained by the procedure described in [5]. The comparison of width (on half depth) of angular scan with that of ideal single crystal makes it possible to distinguish between the texture and polycrystalline phase inclusions.

In fact the defect diagnostics in MEIS is similar to that in RBS, but the advantage of MEIS over RBS in the investigation of textured samples is that the range of block axes misorientation angles which can be registered is several times wider. For instance, single crystalline YBaCuO films with mean block axes misorientation angle of $\sim 2^\circ$ in RBS would be registered as amorphous or polycrystalline, whereas MEIS gives the opportunity to detect the presence of crystalline structure and to determine the misorientation angle value.

In the process of sample probing by middle energy ion beam, besides the back scattered particles, there appear the characteristic X-ray radiation quanta. The simultaneous measurement of characteristic X-ray spectra (Particle Induced X-ray Emission — PIXE) gives the additional

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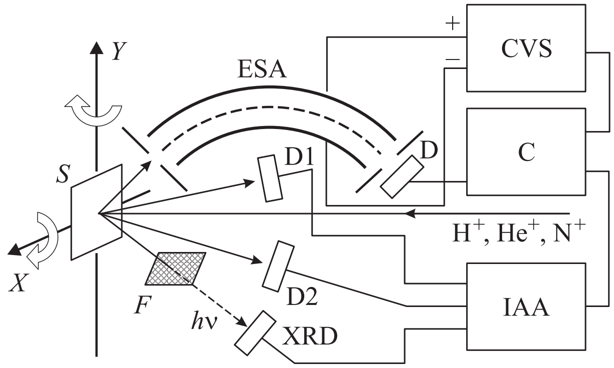

Figure 1. The scheme of MEIS installation. S — sample under investigation, X,Y — axes of sample rotation, ESA — electrostatic analyzer, D, D1, D2 — spectrometer semiconductor detectors, F — 3 μm beryllium foil filter, XRD — X-ray detector, CVS — controlled voltage supply, IAA — impulse amplitude analyzer, C — control computer.

possibilities to specify the information about the sample under investigation. The significant advantage of PIXE measurements in case when middle energy ion beams (e. g. 250 keV protons) are used is practically full absence of bremsstrahlung.

For X-ray quanta the angular scans can also be measured what gives the opportunity to make conclusions on the quality of sublattices of certain elements in complex crystalline compounds [4].

2. MEIS technique

The MEIS installation consists of the accelerator forming well collimated probing ion beam with adjustable ion energy in the range 50−250 keV and of equipment complex for the analysis and registration of back scattered ions and X-ray radiation quanta (Fig. 1). The ion beam has the energy spread less than 10^{-4} of ion energy, energy instability during the measurement cycle also about 10^{-4}, beam angular divergence in sample surface dropping point less then 0.05°. The enumerated above parameters ensure high energy resolution; the worsening of depth resolution due to these beam parameters is less than 0.1 nm. The cross section of ion beam dropping onto the surface of the sample is of order of 1 mm^2.

The back scattered ions are registered by means of two spectrometric semiconductor detectors and ESA, designed and manufactured at the Ioffe Physicotechnical Institute.

The particle detectors have energy resolution of ~ 3.5 keV (as a comparison — for RBS the analogous detectors have the resolution of order of 10 keV) and register the ions scattered to the angles of 120° and 170°. The comparison of spectra measured at these two scattering angles provides the information about the surface porosity of sample under investigation.

The high energy (depth) resolution channel includes high-aperture electrostatic spherical deflector having adjustable relative resolution $E/\Delta E$ from 200 to 500 (choice of which is made according to the optimization of exposition time and resolution relation). For example, for He$^+$ beam and silicon sample it is possible to reach (for near-surface region) the resolution of 0.5 nm (the utmost value is 0.3 nm).

The constituent part of the installation is X-ray quanta (PIXE) registration channel. Its peculiar feature is the absence of vacuum-tight beryllium window as a component of spectrometric quanta detector (nitrogen-cooled ultra pure silicon detector), because the detector is placed directly into the high vacuum chamber and defended from back scattered ions by means of beryllium foil having the thickness of only 3 μm, which makes possible to register quanta with energies beginning from 500 eV, i. e. the K-line of oxygen.

For the processing of experimentally obtained information the original code was created. This code takes into account the factors listed in section 1 and is aimed at the modeling of MEIS spectra of films consisting of separate isles and films with thickness inhomogeneity [3].

3. Some examples of MEIS application

3.1. Investigation of parameters of ultra thin layers in multilayer structures

MEIS spectra of the crystalline multilayer structure consisting of several Si layers of ~ 30 nm thickness divided by Ge layers are shown in Fig. 2. Two experimental spectra — aligned and „random“ — are presented in figure, as well as the result of MEIS spectrum computer simulation. In the inset the relative Ge concentration as a function of depth, obtained from MEIS spectra data processing, is presented. The amount of Ge in each Ge layer approximately corresponded to that of monolayer. As it was estimated, the thickness of the nearest to the surface Ge layer was $7 \cdot 10^{14}$ atoms/cm^2, the thickness of the next layer — $8 \cdot 10^{14}$ atoms/cm^2.

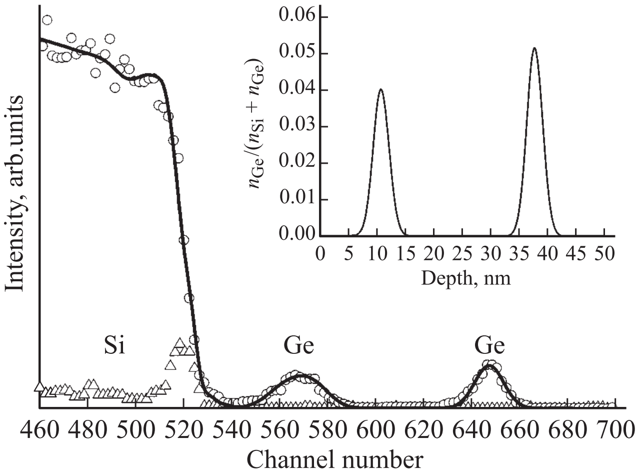

Figure 2. MEIS spectra of the structure Si/Ge/Si/Ge/Si/... (He$^+$, 230 keV, ESA). Triangles — aligned regime, circles — „random“, solid curve — computer simulation. Inset — the dependence of Ge concentration on depth.

The minimum relative yield of back scattered ions just after Si surface peak (channel 500), being near 2.5%, evidences that the sample has crystalline quality similar to that of the best silicon single crystals. The broadening of peaks on the dependence of Ge relative concentration on depth, as well as low magnitudes of peaks (4–5%) are the evidence of absence of continuous uniform Ge layers.

3.2. Diagnostics of nanostructures containing clusters

Fig. 3 depicts the MEIS spectra of structure consisting of InAs clusters deposited onto Si substrate, as well as the result of computer simulation of corresponding MEIS spectrum. The following parameters were deduced as a result of spectra processing: substrate coverage by InAs clusters (5%), medium cluster thickness X_m ($11.2 \cdot 10^{15}$ InAs molecules per cm^2), thickness spread σ ($4.8 \cdot 10^{15}$ InAs molecules per cm^2), integrated amount of matter in clusters ($5.6 \cdot 10^{14}$ InAs molecules per cm^2), the amount of surplus As ($2.6 \cdot 10^{14}$ As atoms per cm^2). The presence of „surplus“ amorphous As becomes apparent, firstly, in the abnormal In and As peak magnitudes ratio (it differs from that corresponding to InAs stoichiometry) and, secondly, in the difference in back scattered ions relative yield (aligned to „random“ signals ratio) in In and As peaks — the rise of yield in As peak as compared to the yield in In peak can be explained only by the presence of not-channeling amorphous As phase. The large value of cluster thickness spread ($\sigma/X_m \approx 0.43$) may be the evidence of the pyramidal form of these clusters, what agrees with the data obtained by means of Reflection High Energy Electron Diffraction (RHEED).

Fig. 4 demonstrates the evolution of MEIS spectra of described above structure in the process of covering Si layer depositing. With its thickness increase, the fore (high energy) In peak front shifts to lower energies. Knowledge of this shift value makes it possible to evaluate the thickness of coverage with high accuracy. The larger fore front extension in comparison with back (low energy) front of curves *2* and *3* proves that the covering layer surface is flat, as it is shown in the right part of Fig. 4. In case of repeating the surface relief by this layer, the relation between the front extensions would be opposite. The presence of a signal, corresponding to the parts of clusters jutting out the surface of coverage (part of the fore front, outlined by rectangle on curve *2*), confirms this conclusion.

3.3. MEIS–PIXE investigation of certain element sublattices in crystalline compounds

Fig. 5 depicts the spectrum of characteristic X-ray radiation emitting by $NdBa_2Cu_3O_7$ sample under 230 keV proton beam bombardment. The appearance of peak with energy near 1480 keV evidences the presence of Al, impurity with concentration of approximately 3% relative to the total concentration of atoms of other sample components. This value was evaluated from corresponding peak intensity. The

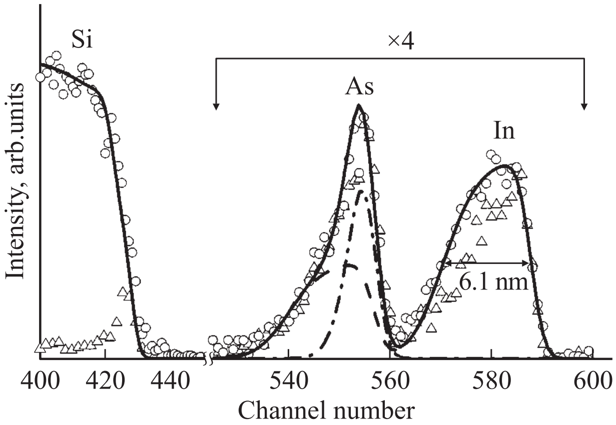

Figure 3. MEIS spectra of InAs clusters deposited onto Si substrate (He^+, 190 keV, ESA). Trianlges — aligned regime, circles — „random“, solid curve — computer simulation.

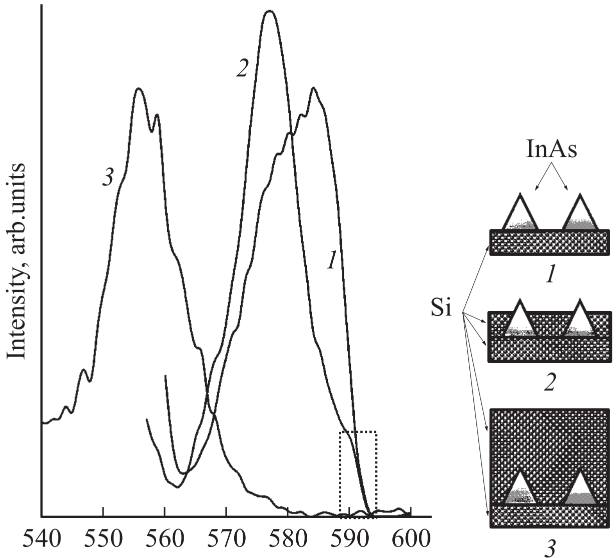

Figure 4. The evolution of MEIS spectrum (In peak) in the process of covering Si layer depositing of the system shown in Fig.3. Curves *1, 2* and *3* correspond to coverage thicknesses of 0, 3 and 13 nm respectively.

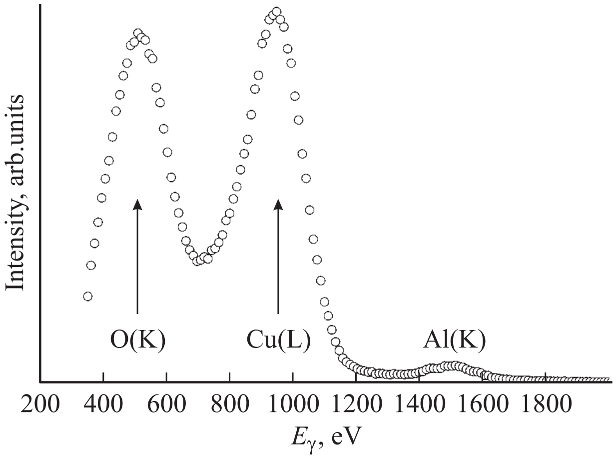

Figure 5. PIXE spectrum of $NdBa_2Cu_3O_7$ with presence of Al impurity.

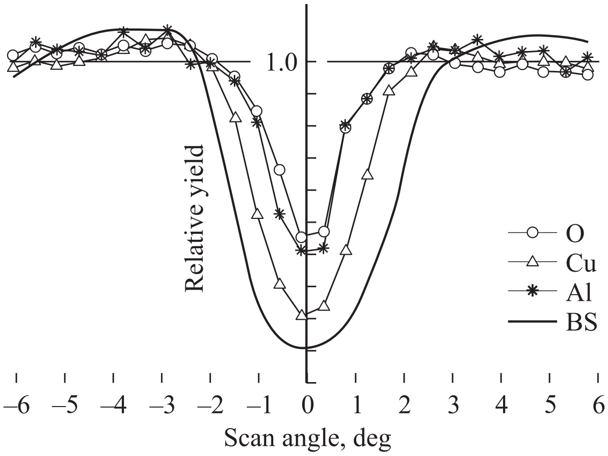

Figure 6. Angular scans of the same as in Fig. 5 system. The angle dependences of O, Cu and Al X-ray emission lines, as well as AS of total flux of back scattered ions (BS) are shown.

dependences of radiation intensity of each line on the angle between probing beam direction and nearest low-index crystalline axis (Angular Scan, AS) are shown in Fig. 6. The AS of back scattered particles, which form is mainly determined by heaviest components (Nd and Ba) sublattice quality, is also presented in Fig. 6. The coincidence of AS for Al and O evidences that Al impurity atoms are built into oxygen rows in the sample.

4. Conclusion

The above presented data show that Middle Energy Ion Scattering — MEIS, combined with Particle Induced X-ray Emission — PIXE, is powerful and rather promising method of solid film and layer diagnostics and is able to ensure the measurements of several important and various parameters of the samples under investigation.

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