Modeling of the hysteretic phenomena in RHEED intensity variation versus temperature for GaAs and InAs surfaces

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This paper describes a study of the reflection high energy electron diffraction intensity change against temperature for GaAs and InAs surfaces. The reflection high energy electron difraction intensity variation against temperature shows different hysteretic characters for the two materials. To date, the explanations for these phenomena were also different for the two substances. Here, we put forward an explanation for these hysteretic phenomena in general terms, applicable to both materials by using the hyperbolic model of hysteresis for coupled systems. Experimental results presented in the paper are in good agreement with the model predictions, supporting the proposed common explanation.

1. Introduction and experimental preliminaries

The compound semiconductor structures grown by MBE (Molecular Beam Epitaxial) technology play an important role in the consitruction of semiconductor devices. Out of the compound semiconductors the most important group is the one $A^{III}B^{V}$ based. In this techology, the polar surfaces with (001) orientation are fundamentally significant. For the growth of low-dimensional nanostructures it is vital to know the growth kinetics of the process. The MBE growth can be observed in-situ by monitoring the surface condition of the crystal (i. e. the surface reconstruction) by RHEED (Reflection High Energy Electron Diffraction), which helps significantly the understanding of the growth kinetics. In case of the exact and the near layer–to–layer growth, the mostly observed surface reconstruction is (2×4), although other surface reconstructions are also present.

In the following we are going to investigate the changes in the behaviour of GaAs and InAs (001) surfaces against temperature variation, based on the experiments of Yamaguhi and Horikoshi [1]. This intensity variation against temperature shows hysteretic properties. During the experiment the change in temperature was slow, so every point could be regarded as being in the state of thermal equilibtium. Shown by the experimental results the temperature dependence of the intensity of the specular spot depends neither on of incident azimuth angles nor on the energy of the electron beam. We can say, therefore, that the intensity change of the specular spot is not the result of diffraction. The results show that at lower temperature the specular spot intensity in high. As the temperature rises the intensity gradually diminishes. Both in the case of GaAs and InAs the surface at lower temperature shows arsenic–terminated (2×4) surface reconstruction. At higher temperature, however, In/Ga terminated (4×2) surface reconstruction can be observed. The change of the direction of the temperature shows, that the process is subject of hysteresis. In the case both semiconductors the observed hysteresis loops fall within approximately a 50°C temperature range. In case of InAs we had two distinct hysteresis loops. At lower temperature the observed wide loop is the indication of smaller intensity variation. At higher temperature however we have seen a narrower loop with large and sudden intensity change. In case of GaAs there is only one real loop at lower temperature and a supposed degenerate pseudo loop at higher temperature. In the second loop the ascending and the descending branches seemingly overlap. We will apply the general description of the phenomenon to both materials. In our present work we will consider two loops in both cases, giving a qualitative explanation for this inverse spin–valve like, coupled loop structure. For the quantitative investigation we applied the $T(x)$ hyperbolic hysteresis model, developed for a general description of hysteretic phenomena [2].

2. Discussion

2.1. Dependence of the specular spot intensity

In the case of diffraction the specular spot intensity would depend on one hand on the surface morphology on the other hand on the surface construction. The reflection from a perfect crystal surface is good, therefore the specular spot intensity in high. A surface with any imperfection disperses the electrons, therefore the specular spot intensity is reduced. The increase in temperature causes primarily arsenic to leave the surface, therefore a perfect surface with high reflectivity is rich in arsenic. Any surface, rich in metallic components, has low reflectivity, disperses the electrons and droplets can also form on it. Both the InAs and the GaAs crystallize in face–centered–cubic structure, with covalent binding, where four identical binding can form due to sp^3-hybridisation. The four

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[¶] E-mail: nemcsics.akos@kvk.uni-obuda.hu equivalent bindings represent identical electron distribution in different directions. At the surface the symmetry brakes and the surface relaxes. In case of the arsenic terminated surface, the probability of the presence of the electrons is higher, due to the distorted electron distribution, because of the missing binding, perpendicular to the surface. When the surface is Ge terminated, then the effect is the opposite. The arsenic terminated surface becomes marginally negative, repelling the electrons, making the specular spot intensity higher. When however the surface is In/Ga terminated then the electrons of the incident beam are neutralized and the specular spot intensity is reduced. The composition and also the morphology of the surface is linked to the surface reconstruction and this reconstruction is the function of the temperature as well as the arsenic pressure. The various reconstructions are periodically roughing up the surface.

2.2. Hysteretic behaviour of the specular spot intensity

As we said before, the spot intensity change versus temperature shows hysteretic properties. The change in stoichiometry is the result of the ongoing absorption and desorption processes and these processes are generally regarded as hysteretic. The surface reconstruction is non-uniform and a number of similar processes could be acting simultaneously, forming domain like structures on the surface. Any exchange between them in shapes and sizes can also be the cause of hysteresis. The increasing temperature shall start the migration of the crystal constituents, leading to increased roughness of the surface. Because the sticking coefficients of the metallic components are close to unity, the migration of arsenic is expected when the temperature is increased. The arsenic incorporation takes three stages. The first step is the physisorption of the arsenic species, followed by the dimeralization of the arsenic to be finally chemisorbed in this form to the surface. The dimers will split at this stage and the arsenic atom will finally be incorporated in the lattice. These processes represent three different energy levels. In our experimental temperature range we only have to deal with the last two processes [3,4]. The process of incorporation is more complex than the process when the arsenic leaving the substance. Before the arsenic is incorporated, it has to be dimeralized and it also has to find two neighbouring vacant locations on the surface for the dimmer to enable the arsenic atom to set in the surface. When arsenic is leaving the surface the process does not need to follow these conditions, therefore the process becomes simpler. As a result the two processes follow two different paths.

The surface behaviour is different from that of the bulk material, because due to the energy minimalization, during relaxation, it forms various surface reconstructions [5–9]. These reconstructions, depending on the temperature and the flux of the components present, result in very complicated phase–diagrams. Each of the $I(T)$ graphs is composed of two hystetesis loops (in case of the GaAs we assume that the second loop is degenerate).

We can conclude from the model, that each of the loops describe one of the separate processes and that these processes are coupled. An inverse spin-valve shows similar character. The loop at lower temperature belongs to higher intensity, therefore it describes a process associated with arsenic–rich surfaces. In both cases the RHEED indicates (2×4) surface reconstruction in this region. On the surfaces without reconstruction the dangling bonds are pointing in $[\bar{1}10]$ direction, that forces the lines of the arsenic–rich reconstructions running in the same directions. This surface symmetry can be formed by a number of reconstructions such as: $a(2\times4)$, $a2(2\times)4$, $b(2\times4)$, $b2(2\times4)$, $b3(2\times4)$, $g(2\times4)$. These domain transformations could also cause hysteresis. The energy levels of these domains are very close to one another. At lower temperature first the arsenic-rich $b2(2\times4)$ reconstruction will take place, followed by the formations less rich in arsenic such as $a(2\times4)$ and $a2(2\times4)$. The $b2(2\times4)$ reconstructions are more stable, than the ones listed above, therefore, they will last longer. They will only transform at higher temperature and than at a faster rate. This represents the upper part of the hysteresis loop. With lowering the temperature, first the reconstructions, of less arsenic content will form. Domains of $b2(2\times4)$ reconstruction will start forming at an appropriate temperature, but they will rapidly dominate the surface due to their favourable construction. The intensity curve therefore will follow another path, which forms the lower hystetesis loop. We can conclude that the transformations are governed not only by the change in the temperature but also the phase transitions and the delay in their excitations.

The upper parts of Figs 1, *a* and *b* depict the coupled nature of the process and the complex functional relationship between the main and the constituent loops for InAs and GaAs respectively. In also shows the relative contributions of the various surface effects to the specular spot intensity variations.

The various surface reconstructions are associated with different stoichiometry, which are linked to the RHEED intensity [10]. Although a large volume of information is available on the subject of surface construction, the temperature effect and growth, the comprehensive and consistent explanation is still awaited for. It is obvious, from what we said before, that the connection between the surface roughness and the RHEED intensity is only part of the truth. The surface, producing maximum intensity is far from perfect due to the process of relaxation, although the surface stoichiometry is changing continuously. The arsenic-rich crystal surface produces higher RHEED specular spot intensity against the lower intensity surface, rich in In/Ga. This assumption is supported by the phenomenon observed at droplet epitaxial formation as well, which shows that the arsenic-rich stripy RHEED picture becomes diffused, when the atoms in the Ga beam combine with the arsenic atoms

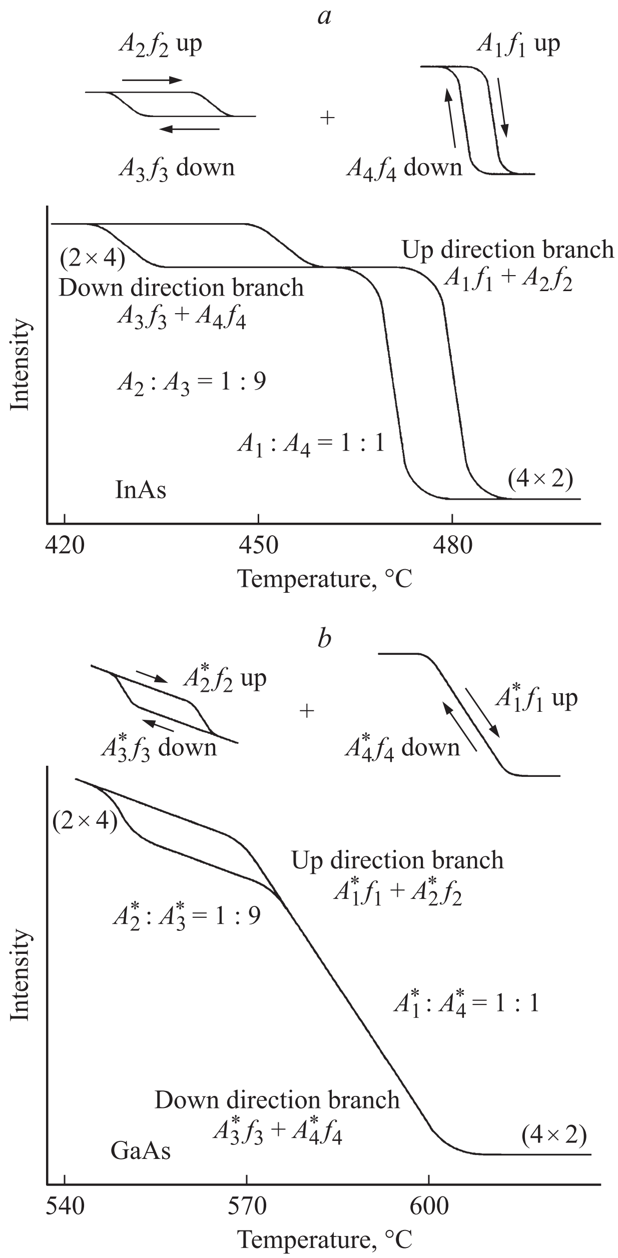

Fig. 1. The two constituent hysteresis loops of $I(T)$ curve for InAs (a) and GaAs (b).

and by absorbing them the result is the presence of Ga atoms on the surface [11,12].

2.3. Modeling the hysteretic phenomenon

The phenomenon of hysteresis in many fields of science is well known and well documented in the literature. The spin-valve and later the inverse spin-valve effect, as part of the hysteretic processes have only been discovered less than ten years ago. Although the phenomenon described in this paper is far removed from the physical mechanism of spin-valves, its general character is strikingly similar. In general terms, the spin-valve effect involves two coupled hysteretic processes and characterized by two hysteresis loops flowing into each other as the excitation varies periodically. Although there are a number of known models for describing hysteresis, so far there is only one, the $T(x)$ hyperbolic model, which can describe this rather complex phenomenon of two coupled hysteretic processes, like in a spin-valve [13]. The model is based on the Langevin's theory of ferromagnetism. The independent constituent components are identified and formulated by their separate hyperbolic functions ($A_n f_n$, $n = 4$, A_n — amplitude, f_n — same functions with different numerical parameters) and linearly superimposed by using Maxwell's superposition principle [14]. The loop, predicted by the model, gives a good fit to the measured specular spot intensity versus temperature $I(T)$ curve as shown in Figs 2, a and b for GaAs and InAs respectively. Although the model predicts the presence of a very narrow loop in the InAs intensity plot at higher temperature as well, its width is probably within the experimental error. It is interesting to note, that while the physical parameters used in modeling are different for the two substances the approximate ratios between the amplitudes (A_n) remained nearly the same. This is a good indication that with the changing temperature the surface

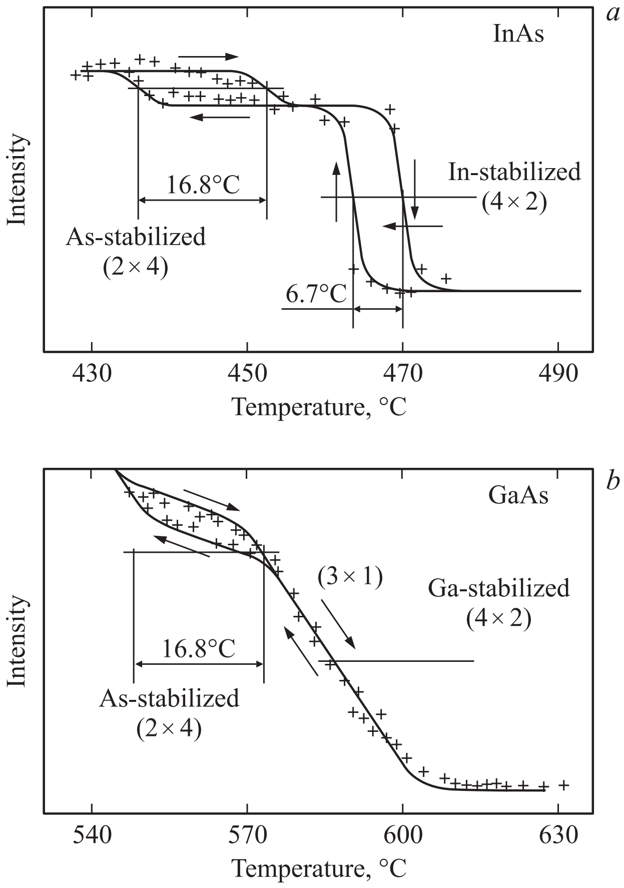

Fig. 2. RHEED specular spot intensity versus temperature for InAs (001) surface (a) and GaAs(001) surface (b). Solid line reperesents experimental results, crosses are predicted by the model.

reaction is the same for the two experimental substances as we have initially assumed.

This first successful modeling of the phenomenon has far reaching implications. The model predicts that, there are two separable, simultaneous coupled processes taking place on the surface of both the GaAs and InAs crystals, at a given time. In one temperature cycle, both cases the up and down processes can be represented by two single simultaneous physical processes. The two simultaneous coupled processes are surface morphology and surface stoichiometry. Each of the four processes is represented by one separate function in the model. In the phase of up-going temperature the dominant process is stoichiometric, representing approximately 90% of the RHEED intensity changes. The rest 10% is due to the morphologic changes. When the temperature is decreasing however, the contribution of the two processes are equal (50%–50%). This finding fortifies the physical explanation given before and the results of the RHEED studies on GaAs and InAs. While, at higher temperature, the As can leave the surface with relative ease, at lower temperature the surface reconstruction with the As capture, involves complex timely processes, as described earlier. The model also leads to the calculation of the activation energy involved in the absorption/desorption processes. Following Boltzman's relations [15,16] the ε activation energy involved in each leg of the hysteresis loop can be calculated as $\varepsilon = \alpha t k T$, where α is the inclination of the leg of the loop in K^{-1}, representative of the speed of the process or reaction at t temperature (in K), k is the Boltzman constant and T is the mid temperature of the processes in K [1,17]. Detailed model interpretation and mathematical formulation of this surface phenomenon is outside the scope of this paper, because its limited size, that will be pulished elsewhere in full [18].

3. Conclusion

In the former literature the RHEED intensity variation in InAs as a GaAs has a completely different interpretation, attributed to different physical processes [1]. The one in InAs is described as a first order phase transition, whilst the one in GaAs is regarded as a second order phase transition. We propose in this paper, that driving force in this kind of surface phenomenon is the same for both of the investigated substances with only different emphasis on its constituent components. The model predicted the presence of two hysteresis loops in both cases, showing the two processes involved. (At higher temperature, in the case of GaAs the second loop is present but narrow or degenerate). The model describes both cases with identical formulation using different physical parameters. This shows a logical approach and points towards a unified way of describing the phenomena without introducing different interpretations in each individual case. We also pointed out that the double hysteresis loop is due to the absorption-desorption process and the changing, domain structured surface morphology (surface reconstruction). The explanations, given for the intensity change, are the stoichiometry, polarization and surface roughness. We can distinguish between a soft and a hard loop, which also characterizes the spin-valve constructions. The constituent processes causing the double hysteresis loop (absorption–desorption and the changing reconstructions) affect the phenomenon in a different way when the temperature increases or decreases. The model showed, that whilst the two processes play equal role (approximately 1 : 1) when the temperature decreases, this ratio is substantially different (approximately 1 : 9) when the temperature is on the increase, due to the kinetics of As incorporation.

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