Effect of neutron irradiation and doping level on defect structure formation in gallium arsenide crystals

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Abstract. Using the technique of diffuse x-ray scattering at the “tails” of diffraction reflection curves, we analyze the effect of irradiation of dislocation GaAs crystals with high-energy neutrons on evolution of radiation clusters as a function of fluence and doping level. The effect of doping level on size of the above defects is shown to be considerable at low fluences. We advance a model for disordered regions in an irradiated crystal based on the results of x-ray experiments.

Keywords: diffuse scattering, neutron, radiation defect, disordered region, nonstoichiometry parameter.

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1. Introduction

Irradiation of crystals with heavy high-energy particles results in point defect (PD) production, as well as formation of disordered regions (DR). It is known that change of principal properties of semiconductor crystals exposed to irradiation with neutrons, protons and other particles is determined, to a great extent, by spectra of complex defects [1-3]. The form of such defects (radiation defect clusters) is ellipsoidal or near-spherical [3]. However, despite rather intense investigation of the nature of such crystal imperfections, up to now there is no consensus concerning their origin and parameters. True enough, some authors [4, 5] state with confidence that a core of such defect aggregation of divacancies. In [6, 7] an analysis of diffuse x-ray scattering was used to study the effect of neutron irradiation on Cu crystals.

The objective of this paper was further investigation of defect structure transformation in GaAs crystals irradiated with fast neutrons, at various fluences and doping levels, using an analysis of diffuse x-ray scattering.

2. Procedure and subject of investigation

The procedure of investigation of crystal defect structure is based on analysis of behavior of the diffuse component of x-ray scattering on the tails of diffraction reflection curves. The diffuse component of x-ray scattering carries principal information on PDs and Coulomb-type defects (clusters and dislocation loops). We used a double-crystal spectrometer in the symmetric diffraction mode (reflections of 400-type, CuKα-line). The mean defect radii were calculated from the expressions given in [6, 7]. The heart of this approach is that for different reciprocal space regions (that correspond to different distances from defect core in ordinary space) the scattering intensity obeys different laws. Far from defect, where crystal lattice distortions are weak (the so-called Huang region), the x-ray intensity I changes at crystal departure from the exact Bragg position as I ∝ q−2. (Here q = 2π/λ cos θ = √ε/r, r is the effective defect size.) At strong crystal distortions (i.e., closer to a defect) the x-ray intensity drops as I ∝ q−4 (Stokes-Wilson region). From position of the point where character of x-ray scattering changes one can determine microdefect size and power (i.e., a parameter that involves product of defect size and concentration). The nonstoichiometry parameter (CA−CA) (here C is relative atomic concentration) was checked with the procedure described in [8]. This procedure involves use of the so-called quasi-forbidden reflections for which the x-ray scattering intensity is proportional to the difference between form-factors of the crystal components.

We investigated GaAs crystals doped with various elements. The initial dislocation density was 10⁴ cm⁻², n-GaAs crystals differed in concentrations of the main donor impurity (TeGa). At the temperature T = 300 K equilibrium electron concentrations in these crystals was from 2×10¹⁶ up to 2×10¹⁸ cm⁻³. The samples were irradiated in the force-cooled reactor channel with neutrons whose energy was 2 MeV; the fluences were 10¹⁵–10¹⁷ n/cm². The sample temperature did not exceed 60 °C. We studied how the defect structure depended on fluence, as well as doping level and dopant nature. It is known [9] that con-
centrations of point defects, \( N_S \) and DR, \( N_D \), induced by neutron irradiation are proportional to fluence \( \Phi_n \) and are, respectively, \( N_S \propto 50 \Phi_n \) and \( N_D \propto 0.21 \Phi_n \).

3. Results and discussion

The principal results of our x-ray experiments are given in Fig. 1. We present dependence of intensity changes on crystal departure from the exact Bragg position on the log-log scale. The sizes of clusters induced by neutron irradiation were determined from the position of the point at which the Huang region of intensity changes transforms into the Stokes-Wilson one. The principal defect parameters are given in Table 1. The third column contains the greatest possible cluster size values, \( r_{\text{max}} \), calculated from the above expressions in the case when there is still no overlapping. An amount of defects per cluster is as big as about 10000. Additional information was obtained from the antisymmetric part of diffuse scattering intensity, \( I_{\text{as}} = [I_{\text{diff}}(+)-I_{\text{diff}}(-)] \). If this antisymmetric part is positive at \( qh < 0 \), then vacancies predominate in the crystal; if it is positive at \( qh > 0 \), then interstitial atoms are predominant. (Here \( q \) is the wave vector and \( h \) is the strain vector.) In our case interstitials atoms are predominant for all the crystals studied, whatever their doping levels are.

Now let us analyze the obtained dependencies and defect parameters starting from the DR model that has been advanced in recent years. One can distinguish the following four stages of DR formation: the cascade, postcascade, quasichemical and accommodation ones. At the first and second stages a DR core (5–15 nm in size) is being formed during a very short period (\( 10^{-14}–10^{-13} \) and \( 10^{-11}–10^{-10} \) s, respectively). At the quasichemical stage the PDs (predominantly fast interstitial atoms) that have been driven out of the cascade intensively diffuse to drains in the crystal matrix. There they either recombine or form composite defect-impurity complexes [10]. The growing defects (dislocations), as well as PDs induced in the crystal matrix, can serve as drains. So the DR size will be determined by the dimension of the defect-impurity envelope that has been formed around the DR core. As a result of diffusion and recombination processes, the secondary defects are produced that are stable at this temperature. It is the distribution of these stable secondary defects that determines the effect of neutron irradiation on the crystal properties.

One can see from the results presented in Table 1 that the doping level effect on defect size is essential at low \( (\Phi_n = 10^{15} \text{ n/cm}^2) \) fluence. When fluence increases, then the defect size dependence on the dopant concentration becomes less pronounced, and at high \( (\Phi_n > 10^{16} \text{ n/cm}^2) \) fluences this dependence is absent. Such character of the parameter \( r \) changing with fluence evidences that there are some principal factors that determine its value. At low fluences production of PDs in the GaAs matrix almost does not change the matrix properties. In particular, for all values of the doping level (i.e., at any \( N_{\text{Te}} \) value) the relation \( N_S < N_{\text{Te}} \) holds. Besides, at low DR concentration the mean spacing between these regions is bigger than \( r \) (in other words, the DRs do not overlap). Growth of \( r \) value with doping level comes from intense interaction between PDs driven out of the cascade and Te\(_{\text{Ga}}\) atoms; as a result, defect-impurity complexes are formed. From an analysis of Fig. 2 it follows that content of As and Te atoms in DRs increases. At further growth of fluence, concentration of DRs grows, and for some \( \Phi_n \) values DRs should not be treated as being isolated, because their defect-impurity envelopes become to overlap. At a result, one will observe drop in the \( r \) value. On the other hand, growth of \( \Phi_n \) value leads to increase of PD concentration. This means that at a given dopant concentration the relation \( N_S > N_{\text{Te}} \) is valid, and therefore the defect sizes can increase due to decoration of PDs produced in the matrix. One can see from Table 1 that at the highest value of dopant concentration (when the isolated DR size is maximal) fluence growth leads to DRs overlapping and decrease of the \( r \) value. At the lowest value of dopant concentration (and, correspondingly, the

### Table 1: Dependence of defect size in GaAs crystals on dopant concentration and neutron fluence.

<table>
<thead>
<tr>
<th>Fluence, ( \text{n/cm}^2 )</th>
<th>Dopant concentration, ( \text{cm}^{-3} )</th>
<th>Maximal cluster size ( r_{\text{max}}, \text{nm} )</th>
<th>Effective defect size ( r, \text{nm} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( 10^{15} )</td>
<td>( 5 \times 10^5 )</td>
<td>96.5</td>
<td>36.1</td>
</tr>
<tr>
<td>( 10^{15} )</td>
<td>( 5 \times 10^6 )</td>
<td>44.7</td>
<td>62.5</td>
</tr>
<tr>
<td>( 10^{15} )</td>
<td>( 5 \times 10^7 )</td>
<td>67.8</td>
<td>64.7</td>
</tr>
<tr>
<td>( 10^{16} )</td>
<td>( 5 \times 10^5 )</td>
<td>20.9</td>
<td>29.3</td>
</tr>
<tr>
<td>( 10^{16} )</td>
<td>( 5 \times 10^6 )</td>
<td>28.5</td>
<td>28.5</td>
</tr>
<tr>
<td>( 10^{16} )</td>
<td>( 5 \times 10^7 )</td>
<td>28.3</td>
<td>28.3</td>
</tr>
</tbody>
</table>

Fig. 1. Intensity of diffracted x-rays as function of GaAs crystal rotation angle on the log-log scale at different fluences.
minimal size of an isolated DR) fluence growth results in increase of the r value due to accommodation processes, until DRs still remain isolated. At \( \Phi_n > 10^{16} \text{ n/cm}^2 \) DRs overlapping becomes the predominant factor; for all the samples studied, whatever the dopant concentration, a decrease in the effective DR size is observed.

4. Conclusion

From analysis of the experimental results it follows that dopant atoms serve as efficient drains for PDs during DR formation. The most probable model for DR formation in our case is complexes of initial matrix defects and technological (doping) impurities in the DRs.

References