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Ge/GaAs(100) Thin Films: Large Effect of Film Growth Rate and Thicknesses on Surface Morphology, Intrinsic Stresses and Electrical Properties

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We found out and studied a profound effect of film growth rate on the electrical properties, intrinsic stresses and surface morphology of thin Ge films grown on GaAs(100). This effect is essential and has to be accounted for when developing and producing devices based on the Ge/GaAs heterostructure. All the Ge films under investigation were single-crystalline and epitaxially-grown on the GaAs(100) substrates. However, the transport phenomena in Ge films grown at low and high deposition rate differed drastically. Those obtained at low deposition rate were p-type and high resistant. They had a low concentration of free charge carriers and thermally activated conductivity, which is characteristic of heavily doped and strongly (in the limiting case, fully) compensated semiconductors. Although such films were single-crystalline, their conductivity was percolation-type. The Ge films obtained at high deposition rate were n-type and low resistant. They had high concentration of free charge carriers. The temperature dependence of conductivity in such films was weak or practically absent, which is characteristic of degenerate heavily doped semiconductors. Besides, the surface morphology cardinally differed for films obtained at low and high deposition rate. At low film growth rates, surfaces with developed relief were observed whose valleys and ridges formed grains of irregular shape with pronounced substructure. As the film thickness grew, the surface relief became essentially pronounced. At rather high film deposition rates, contrary to the above, the Ge film surface was fine-grained and smooth; the surface relief practically did not depend on the film thickness. As the deposition rate went down, the intrinsic stresses in films essentially decreased. The results obtained were analyzed from the viewpoint of formation of compositional and morphological inhomogeneities, and fluctuations of electrostatic potential at low growth rates. Such potential fluctuations modulate Ge energy bands leading to appearance of potential relief and deep tails of density of states in the Ge bandgap.

Keywords: Ge/GaAs, Electrical properties, Intrinsic stresses, Surface morphology.

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1. INTRODUCTION

The Ge/GaAs is promising for a number of practical applications and an excellent model system for IV/III-V heterostructures in general. This heterostructure is characterized by close values of lattice constants of both materials (Ge and GaAs) as well as practical coincidence of their coefficients of thermal expansion. The distinguishing feature of Ge films on GaAs is also the strong dependence of their structure, electrical and optical properties on the technological conditions and method of preparation.

In our previous [1, 2] and recent work [3–5] we show that deposition rate drastically affects the electrical and optical properties as well as surface morphology of thin (~100 nm) Ge films on GaAs. In particular we show that efficient control of the film properties can be achieved by varying their deposition rate.

This work is a continuation of the studies presented in Ref. [5], with focus on the effect of deposition rate on the intrinsic stresses and morphology in Ge films. We have found that at low deposition rates films surface morphology strongly depends on the film thickness as well as the underlying film deposition rate. These effects are essential and should be taken into account when developing and producing devices based on the Ge/GaAs heterostructure.

2. EXPERIMENT

2.1 Preparation Conditions and Measurement Procedure

The Ge films were deposited using thermal evaporation of Ge in vacuum onto substrates made of semi-insulating GaAs(100). The temperature of GaAs substrate in the course of Ge film deposition was maintained constant (500°C). The film deposition rate was also maintained constant in the course of deposition; it varied from 0.02 up to 0.35 nm/s for different specimens. The deposition rate was set by the crucible current that determined the Ge evaporation temperature. The film thickness varied from 1 nm up to 250 nm for different films.

The morphology of Ge film surface was studied with a scanning atomic force microscope (AFM) NanoScope IIIa.

The value and sign of intrinsic stresses σ in films related to distinction between the Ge and GaAs lattice constants were determined from the radius of curvature of specimen surface with a profilograph using the Stoney's formula [6, 7] $\sigma = Ed^2/[6(1-\nu)Rt]$. Here E, t and ν are Young's modulus, thickness and Poisson ratio of the substrate, respectively, d is the film thickness and R is the heterosystem bending radius.

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Table 1 – Dependencies of root-mean-square (RMS) roughness and surface area increasing (SAI) on thickness for the Ge films of different thicknesses grown at 0.02 nm/s and 0.35 nm/s rates

d (nm)	**RMS** (nm) 0.02 nm/s	**RMS** (nm) 0.35 nm/s	**SAI** (%) 0.02 nm/s	**SAI** (%) 0.35 nm/s
1	0.45	0.67	0.1	0.2
10	1.3	1.76	2.5	2.4
20	2.9	2.75	4.1	3.0
50	7.5	0.94	8.0	0.3
100	14.6	0.49	12.5	0.15
250	27.0	0.7	25.0	0.15

The electrical properties of the Ge films were determined by Hall effect, magnetoresistance and conductivity measurements.

2.2 Surface Morphology

The films prepared with low (0.02 nm/s) deposition rate had surface with developed relief whose ridges and valleys formed grains of irregular shape with pronounced substructure (Fig. 1). The root-mean-square (RMS) roughness and surface area increasing (SAI) (i.e., the difference of the surface area and its projection area divided by the projection area) of Ge films monotonically grow: at a film thickness of 250 nm, they are 27 nm and 25%, respectively (Table 1) . At a film thickness of about 1 nm, a nucleating layer with characteristic grain diameter of 12 nm is formed. The character of surface grain distribution changes from log-normal to normal and bimodal in a 1 nm, 10 nm and 20 nm thick layers, respectively. At Ge layer thickness over 50 nm, the grain size increases abruptly, with essential broadening of the grain diameter histogram.

Contrary to the above, a practically smooth surface is formed for films obtained at high (0.35 nm/s) deposition rate (Fig. 2 and Table 1). The only exception is the initial growth stages. The growth begins from a nanograin nucleating layer (at a thickness of 1 nm) with the same log-normal size distribution as at low growth rate. At thickness of about 1 nm, a nucleating layer (with typical grain diameter of 9 nm) is formed that is thereafter growing with pits. The density and depth of pits decrease as film thickness increases; the pits are practically not observed at film thicknesses over 50 nm. RMS and SAI peaks at epitaxial layer thickness $d = 20$ nm which is related to presence of the above-mentioned pits. At $d \geq 50$ nm, RMS and SAI are 0.7 nm and 0.15%, respectively, and do not depend on the film thickness.

2.3 Intrinsic Stresses

The investigations of radius of curvature of the Ge/GaAs heterostructure showed that the Ge films were compressed. The intrinsic stresses in them calculated using the Stoney's formula depended on the film growth rate: they decreased from 6.7×10^{8} Pa down to 2×10^{8} Pa (i.e., by more than three times) as the deposition rate decreased from 0.35 nm/s down to 0.02 nm/s (Table 2).

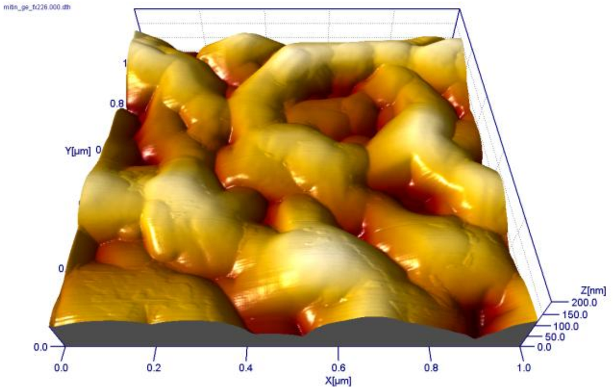

Fig. 1 – 3D AFM images from (1×1) µm area of Ge films on GaAs for film thicknesses of 250 nm. Ge growth deposition rates used is 0.02 nm/s

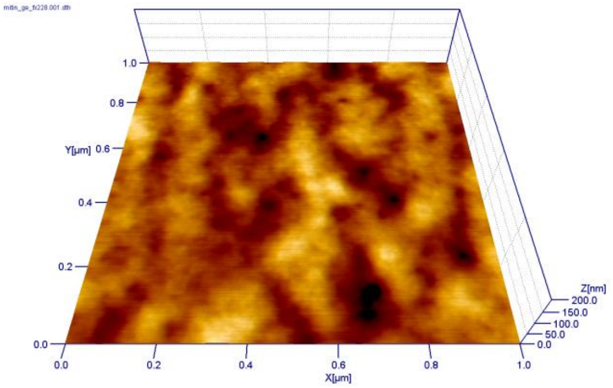

Fig. 2 – 3D AFM images from (1×1) µm area of Ge films on GaAs for film thicknesses of 250 nm. Ge growth deposition rates used is 0.35 nm/s.

2.4 Electrical Transport Properties

The transport phenomena also differed drastically in films obtained at high and low deposition rate (Table 2).

Our investigations showed that Ge films with thickness of about 130 nm obtained at low deposition rate are *p*-type and high-resistant (10-120 Ω·cm), with low concentration of free charge carriers (10^{15}–10^{16} cm^{-3}) at room temperature. Their conductivity is thermally activated and is described by the expression $\rho(T) = \rho_1 \exp(\varepsilon_1 / kT)$ with activation energy ε_1 close or equal to $E_g/2$ for Ge (Table 2). In the most high-resistant (compensated) Ge films Hall coefficient reverses sign as temperature grows. The transverse magnetoresistance greatly increases with temperature (Fig. 3).

Contrary to the above, the Ge films obtained at low deposition rate are *n*-type and low-resistant (0.1–0.01 Ω/cm), with high concentration of free charge carriers (10^{18}–10^{19} cm^{-3}) (Table 2). Thermal dependence of their conductivity, Hall coefficient and magnetoresistance is weak or practically absent, which is characteristic of heavily doped degenerate semiconductors (Fig. 3).

Table 2 – Dependencies of intrinsic stresses (σ), resistivity (ρ), activation energy (ε_1), conductivity type (CT) and root-mean-square (RMS) roughness for the Ge films of 130 ± 30 nm thicknesses grown at different deposition rates *V*

V (nm/s)	**σ** (Pa)	**ρ** (Ω/cm)	ε_1 (eV)	**CT**	**RMS** (nm)
0.02	-	123	0.35	p	-
0.025	2×10^8	108	0.3	p	10.7
0.033	2.11×10^8	21	0.25	p	9.0
0.04	2.2×10^8	4	0.19	p	-
0.075	2.68×10^8	0.19	0.05	n	6.4
0.118	3.38×10^8	0.21	0.04	n	3.4
0.37	6.7×10^8	0.018	0	n	3.0

3. DISCUSSION OF RESULTS

Let us discuss the main results obtained. Our investigations showed that all Ge films under consideration had single-crystalline structure and coherent dislocation-free film–substrate interface. However, the deposition rate strongly affected the electrical properties, intrinsic stresses and surface morphology of Ge films on GaAs. Besides, the film surface morphology strongly depended on the deposition rate as well as on film thickness.

The early stages of film growth (up to thickness of 20 nm) at low and high deposition rates are similar. The growth begins from a nanograin nucleating layer. Then, at high deposition rate, pits appear. They are healed thereafter as film thickness increases and practically disappear at film thicknesses over 50 nm; in this case, surface roughness is below 1 nm. Appearance of pits at high deposition rates may be explained by the effect of phase decomposition and formation of As atom clusters that prevent from acquiring of Ge adatoms. Such a situation has been observed and discussed in [8, 9]. This correlates with the electrical investigations that showed that such films were *n*-type.

At low deposition rate, the pits do not appear while a surface is obtained with a developed relief. Its ridges and valleys form grains of irregular shape. Both the film and grains on it have a single-crystalline relief. This can be clearly seen at film thicknesses over 50 nm. Most likely a developed surface leads to reduction (relaxation) of intrinsic stresses in films. It was noted in [10] that appearance of compositional and morphological inhomogeneities in stressed dislocation-free single-crystalline films may be one of the main ways of structural relaxation. Thus, one may assume that mechanism of intrinsic stresses relaxation in Ge films on GaAs grown at low deposition rate is surface corrugation of the epitaxial layer. In addition, compositional inhomogeneities in Ge films also can reduce the elastic strain energy in heterosystem.

It is also known that for layers grown on mismatched substrates, there is a coupling between the local composition and the surface morphology, so that a rough film will always decompose, while a flat layer will always be homogeneous [9, 10].

Let us discuss the transport phenomena in Ge films. Presence of compositional inhomogeneities in the Ge films is confirmed by electrical measurements.

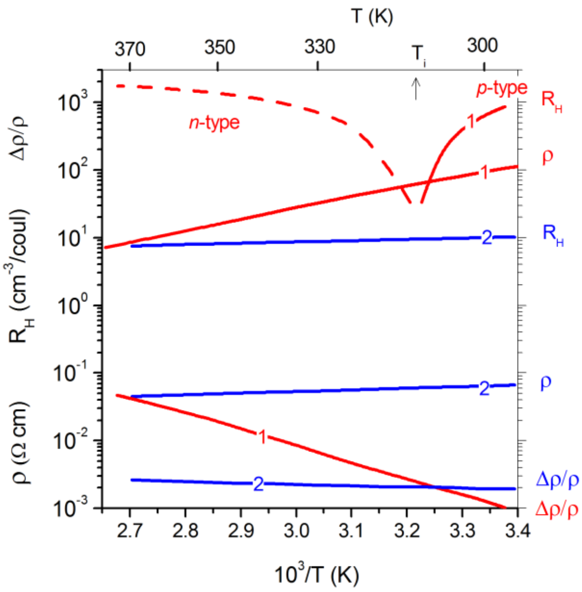

Fig. 3 – Typical dependencies of resistivity (ρ), Hall coefficient (R_H) and magnetoresistance (Δρ/ρ) on inverse temperature for 100 nm Ge films obtained at deposition rate of 0.02 nm/s (1) and 0.35 nm/s (2).

Indeed, the films grown at low deposition rate have higher resistivity as well as low both concentration and mobility of free charge carriers. To give consistent explanation for the electrical and optical properties of such films (see [4, 5]), we applied the theory [11] that assumes presence in semiconductor of large-scale fluctuations of electrostatic potential related to randomly nonuniform distribution of impurities and compensation, i.e., compositional disorder. Such potential fluctuations modulate semiconductor energy bands leading to appearance of potential relief and deep tails of density of states in the semiconductor bandgap. The main characteristics of potential relief are amplitude γ and spatial scale r that depend on the characteristics of random electrostatic field. In heavily doped and strongly compensated semiconductors γ and r depend on the type of spatial distribution of impurities (correlated or uncorrelated), doping level and degree of compensation [11]. Potential relief leads to appearance of regions with free charge carriers in the potential relief valleys and dielectric regions containing no charge carriers. In this case, conductivity is percolation-type and is characterized by percolation level ε_p [11].

At high temperatures ($T > 250$ K for Ge films under investigation [3, 4]) conductivity of Ge films (that is realized by thermionic emission of charge carriers from the Fermi level ε_F to the percolation level ε_p) is characterized by activation energy $\varepsilon_1 = \left|\varepsilon_p - \varepsilon_F\right|$.

For thin films (thickness of about 100 nm) a situation may be realized that typical (characteristic) size of potential relief is more than or equal to the film thickness. In this case, the potential relief is two-dimensional (2D), and 2D percolation is observed [3–5]. In such films (that are fully compensated) temperature dependence of conductivity is exponential, with activation energy close or equal to $E_g/2$ [1–5].

We observed $p \rightarrow n$ inversion of Hall coefficient R_H as temperature increased in the Ge films with extremely high degree of compensation (Fig. 3). The inversion temperature T_i was about 310 K. The sign of R_H corresponded to n-type (p-type) conductivity at $T > T_i$ ($T < T_i$). Such inversion indicates strong compensation and bipolar conductivity in semiconductor. Indeed, high degree of compensation (the Fermi level is in the midgap) and fluctuations of the electrostatic potential lead to two independent conductivity channels. At relatively high temperatures, conductivity is realized by both electrons and holes thermally transferred to their percolation levels that are spaced by potential barriers, i.e., conductivity is bipolar. In this case, one should take into account contribution to kinetic effects from electrons as well as holes. It is known that different charge carriers contribute differently to the kinetic coefficients.

Inversion of Hall coefficient in Ge films indicates predominance of electrons over holes (for the Hall effect) as temperature increases, i.e., transition to intrinsic conductivity. Low inversion temperature ($T_i = 310$ K) indicates strong compensation of the films under investigation. A relatively high concentration of intrinsic charge carriers n_i (10^{15}–10^{16} cm^{-3}) as compared with that in pure intrinsic i-Ge ($n_i = 10^{14}$ cm^{-3} [12]) results from presence of deep tails of the density of states in the semiconductor bandgap which leads to increase of the number of intrinsic charge carriers in the semiconductor. This may cause effects uncommon from the viewpoint of the theory of homogeneous semiconductors.

It should be also noted that transverse magnetoresistance in Ge films increases with temperature. It is known that the magnetoresistance effect reflects the behavior of drift mobility of free charge carriers. Increase of the drift mobility of free charge carriers with temperature may be explained by reduction of their scattering on fluctuations of random electrostatic potential as well as by reduction of probability of their capture to localized states near the conduction (valence) band edge. This also follows from presence of a potential relief and compositional disorder in the single-crystalline Ga films under investigation.

As to Ge films grown at high deposition rate, there is practically no compositional disorder in them, the amplitude of electrostatic potential fluctuations is low ($\gamma < kT$) and the films are heavily doped and slightly compensated. Therefore, their conductivity practically does not depend on temperature.

4. CONCLUSION

In conclusion, we showed that growth rate strongly affects the properties of Ge films on GaAs. This effect is essential and should be taken into account when developing and producing devices based on the Ge/GaAs heterostructure.

All Ge films have single-crystalline structure and coherent dislocation-free film–substrate interface.

At low growth rate, film surface with a developed relief is obtained whose ridges and valleys form grains of irregular shape with a pronounced substructure. The surface relief in Ge films becomes more pronounced as film thickness increases.

Contrary to this, at rather high deposition rate, the Ge film surface is smooth, and surface relief does not depend on film thickness.

The intrinsic stresses also strongly depend on the rate of Ge films growth on GaAs: they decrease as deposition rate goes down. This is explained by formation of compositional and morphological inhomogeneities at low growth rate that are one of the main ways for structural relaxation in stressed dislocation-free single-crystalline films.

The transport phenomena in Ge films obtained at low and high deposition rate differ drastically. The films obtained at low deposition rate are p-type, high-resistant, heavily doped and strongly (in the limiting case, fully) compensated. Their conductivity is thermally activated. Although such films are single-crystalline, their conductivity is percolation-type.

The Ge films obtained at high deposition rate are n-type, low-resistant, heavily doped and slightly compensated. Temperature dependence of conductivity in these films is weak or practically absent.

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