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High phosphorous doped germanium: Dopant diffusion and modeling

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The *in situ* n-type doping of Ge thin films epitaxial grown on Si substrates is limited to $1 \times 10^{19}\,\mathrm{cm}^{-3}$ by the phosphorous out-diffusion during growth at 600 °C. By studying the phosphorous diffusion in Ge with different background doping, we find that the diffusion coefficient is extrinsic and is enhanced 100 times in Ge doped at $1 \times 10^{19}\,\mathrm{cm}^{-3}$ compared to intrinsic diffusivity. To achieve higher phosphorous concentration, delta-doped layers are used as a dopant source for phosphorous in-diffusion. We show that the doping level is a result of the competition between in-diffusion and dopant loss. The high diffusivity at high n-type carrier concentration leads to a uniform distribution of phosphorous in Ge with the concentration above $3 \times 10^{19}\,\mathrm{cm}^{-3}$. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4745020]

I. INTRODUCTION

High n-type doped, tensile strained Ge-on-Si has been proposed and demonstrated to be a very promising candidate for the CMOS compatible light source in silicon photonics.[1–6] Liu *et al.*[1] proposed to introduce tensile strain into Ge to shrink the direct bandgap in relation to the indirect bandgap and concurrently dope Ge with n-type dopants to fill the L valleys with electrons. Due to this indirect-valley-states filling effect and the subsequent increase in the Fermi level, Ge becomes a pseudodirect bandgap material. An optically pumped Ge laser at room temperature has been demonstrated with $50\,\mathrm{cm}^{-1}$ gain using a phosphorous concentration of $1 \times 10^{19}\,\mathrm{cm}^{-3}$.[6] Since theoretical calculations predict that the Ge material gain increases with doping concentration,[7] higher doping concentrations are advantageous for electrical pumping to overcome additional losses due to free carrier absorption in highly doped contact layers and at metal interfaces.

Current *in situ* n-type doping using ultra high vacuum chemical vapor deposition (UHVCVD) epitaxy is limited to about $10^{19}\,\mathrm{cm}^{-3}$ active carrier concentration due to a delicate balance between dopant incorporation and dopant out-diffusion during growth.[8] There are several approaches to increase the n-type doping level. Molecular beam epitaxy (MBE) has been shown to reach activated n-type dopant concentrations of $10^{20}\,\mathrm{cm}^{-3}$.[9] Phosphorous delta-doped layers formed by encapsulating sheet of phosphorous atoms with i-Ge by MBE also demonstrate an active concentration of $10^{20}\,\mathrm{cm}^{-3}$. However, only 20% of phosphorous is activated because of the heavily doping above solid-solubility limit.[10] Ion implantation is routinely used to introduce dopants in CMOS devices.[11] Due to the lattice damage, a temperature annealing step is always needed to remove the damage and activate the dopants. In Ge, the highest achieved active concentration is $(5–6) \times 10^{19}\,\mathrm{cm}^{-3}$ after rapid thermal annealing (RTA) at 600 °C.[12–14] However, implant damage is difficult to remove by annealing due to the lack of undamaged Ge as a regrowth substrate when implanting into thin films and, thus, resulting in increase of optical loss in the Ge. Long time furnace annealing can remove most implantation damages but, due to out-diffusion, the average doping concentration is reduced to $1 \times 10^{19}\,\mathrm{cm}^{-3}$.[15] Laser annealing has been demonstrated to remove most of the implantation damage with limited dopant loss.[15] However, we are unable to reproduce these results in high n-type doped Ge. Unlike the study of diffusion in Ge for next generation MOSFETs, which requires shallow n + p junction,[12] high phosphorous diffusivity is a desirable property to help diffusion in Ge laser devices to achieve high doping level in a short annealing time. In regard to the phosphorous diffusion mechanism in Ge, a general agreement on a vacancy assisted mechanism exists, and the diffusion depends on the square of carrier concentration in the extrinsic diffusion region. However, the quantitative values of the phosphorous diffusion coefficient reported in previous studies are for implanted bulk Ge.[15] The material for Ge laser device is 0.2% tensile strained thin film Ge with $1 \times 10^{19}\,\mathrm{cm}^{-3}$ *in situ* doping level. Therefore, the phosphorous diffusion in this material system may be affected by the background doping and the Ge/Si interface which has a high misfit dislocation density.

In this letter, to avoid implantation damage introduction, we utilize delta doping to increase the n-type doping concentration to above $3 \times 10^{19}\,\mathrm{cm}^{-3}$ in single crystal Ge. Phosphorous concentration as high as $10^{21}\,\mathrm{cm}^{-3}$ can be achieved in Si[16] or Ge[10] by delta doping method, but they are confined into an ultranarrow several-nm-wide layer without a full electrical activation. The encapsulated dopants in our delta doping method are treated as a dopant reservoir. By RTA, the dopants are driven into the underlying single crystal Ge layer. We investigate the phosphorous diffusion in the Ge thin film taking into account the phosphorous out-diffusion from the surface and the phosphorous pileup at the Ge/Si interface. Considering that the Ge layer underneath the dopant reservoir has a background doping level of

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$1 \times 10^{19}\,cm^{-3}$, diffusion is in the extrinsic region with a quadratic dependence on carrier concentration.

II. MATERIALS PREPARATION

The Ge films were epitaxially grown on Si (100) substrate using a hot-wall UHVCVD reactor. By controlling growth temperature and GeH_4/PH_3 gas ratio, the desired active concentration (up to $1 \times 10^{19}\,cm^{-3}$) of the n-type donor phosphorus can be incorporated into the Ge layer. A 60 nm Ge buffer layer was first directly grown on Si at 360 °C to kinetically suppress island formation. Then a 300 nm thick Ge layer with *in situ* activated phosphorous at a doping level of $1 \times 10^{19}\,cm^{-3}$ was grown at 600 °C.[17] After the two-step growth, the wafer was exposed to PH_3 at 400 °C to deposit approximately a monolayer of phosphorous atoms onto the Ge surface while at the same time desorbing the hydrogen from the surface. Subsequently, a 60 nm thick i-Ge film was then deposited at 400 °C. Several cycles of P saturation and i-Ge growth were performed in the reactor to encapsulate multiple layers of phosphorous. We then deposited 100 nm SiO_2 on the Ge film as a cap layer to prevent out-diffusion during annealing. The films were annealed by RTA at temperatures ranging from 600 °C to 750 °C using different annealing times. More details on the growth process can be found in Ref. 18.

Secondary ion mass spectroscopy (SIMS) measurements were performed on the as-grown sample to determine the phosphorous depth profile (Fig. 1). The high phosphorous concentration up to $10^{20}\,cm^{-3}$ within the depth of ~335 nm shows a successful encapsulation of phosphorous atoms in delta-doped layers. Because the deposition temperature of i-Ge is at 400 °C, phosphorous diffusion occurs and, thus, the phosphorous concentration peaks at the delta-doped positions are not very distinct. Similarly, phosphorous also diffuses into the *in situ* doped Ge layer from the delta doped layers, which can be clearly seen in Fig. 1. The knock-on artifact, i.e., ions pushing phosphorous further into the underneath layer during the SIMS process, can be ruled out since this is effective within 10–20 nm only.[16] The arising phosphorous concentration at the interface of the Ge buffer and Si substrate is due to phosphorous accumulation in the undoped Ge buffer layer due to the high dislocation density.

TABLE I. Average phosphorous concentration from SIMS measurements and activated carrier concentration from Hall effect measurements.

RTA Temperature (°C)	RTA Time (s)	Average P concentration by SIMS (cm^{-3})	Activated carrier concentration by Hall (cm^{-3})
	30	3.3×10^{19}	$(2.9 \pm 0.3) \times 10^{19}$
	60	3.0×10^{19}	$(2.6 \pm 0.3) \times 10^{19}$
600	180	2.5×10^{19}	$(2.8 \pm 0.3) \times 10^{19}$
700	60	2.5×10^{19}	$(2.6 \pm 0.3) \times 10^{19}$

Hall effect measurements were performed on an as-grown Ge sample finding an active carrier concentration of $1.5 \times 10^{19}\,cm^{-3}$. Comparing to the average phosphorous concentration of $4.4 \times 10^{19}\,cm^{-3}$ from the SIMS profile, we can conclude that the phosphorous is not completely electrically activated. Hence, annealing is required to activate the phosphorous, which also causes phosphorous diffusion into the *in situ* doped Ge layer to achieve higher doping concentration with single crystalline quality. RTA at various conditions was performed on the as-grown samples and SIMS and Hall effect measurements were carried out (Table I and Fig. 2). The standard measurement error for the Hall effect measurement setup is ± 10%. The comparable data between average phosphorous concentration from SIMS and active carrier concentration from Hall effect measurements show that the phosphorous dopants are completely activated after annealing. During annealing, dopant loss by out-diffusion is observed, which can be concluded from the decrease of average phosphorous concentration with longer annealing times. Driven by the concentration gradient, phosphorous diffuses deeper into the Ge layer from the source with longer annealing time. High carrier concentration and an even distribution profile are the criteria to determine the best annealing conditions. We conclude that RTA at 600 °C for 3 min is a good

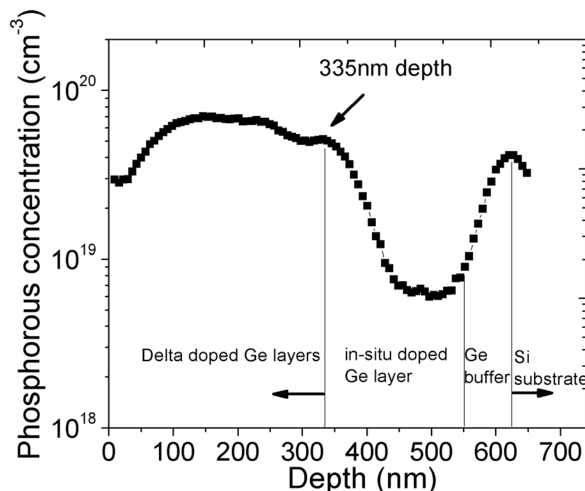

FIG. 1. Phosphorous depth profile of the as-grown Ge-on-Si thin film determined by SIMS.

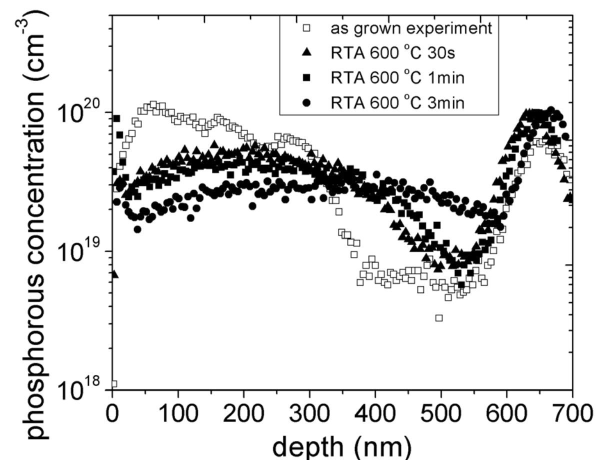

FIG. 2. SIMS depth profile of phosphorous in Ge for an as grown sample (open squares) and after RTA at 600 °C for 30 s (full triangles), 1 min (full squares), and 3 min (full circles).

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annealing condition for dopant diffusion. In this case, an evenly distributed carrier concentration of $2.5 \times 10^{19}\,\text{cm}^{-3}$ is achieved in the single crystalline Ge.

III. ANALYSIS AND DISCUSSIONS

Theoretical calculations predict that the vacancy formation energy in Ge (2 eV) is significantly lower than that in Si (3.5 eV), which implies that vacancies play a more important role in Ge than in Si.[19] Previous results by Werner *et al.*[20] demonstrate that Ge self-diffuses by a vacancy mechanism in which vacancies act as acceptors and all n-type dopants (P, As, and Sb) have higher diffusivity in Ge than Ge itself.[21] This fact reveals that an attractive interaction between n-type dopants and vacancies helps the formation of dopant-vacancy pairs (DV^-). In the following, we present a model using the vacancy mechanism for the phosphorous diffusion in Ge. The charge state of the vacancy is assumed to be doubly negative which is consistent with other assumptions of the phosphorous diffusion mechanism.[22] The extrinsic diffusivity $D_{(DV)}{}^-$ is quadratic carrier concentration dependent and can be expressed as follows:

$$D_n = D_{(DV)^-} = D_0\left(\frac{n}{n_i}\right)^2, \tag{1}$$

where D_0 is the intrinsic diffusivity, n is the equilibrium carrier concentration in the Ge, and n_i is the intrinsic carrier concentration in tensile strained Ge at the diffusion temperature. Due to the reduction in the bandgap and the splitting of the valence bands, the intrinsic carrier concentration in tensile strained Ge is given by[23]

$$n_i = \sqrt{N_c\left(N_{lh} + N_{hh}\exp\left(\frac{-\Delta E}{kT}\right)\right)}\exp\left(-\frac{E_g}{2kT}\right), \tag{2}$$

where N_c is the effective density of states of electrons in the conduction band, N_{lh} is the effective density of states of light holes in the valence band, N_{hh} is the effective density of states of heavy hole in the valence band, and ΔE is the splitting energy of light and heavy holes at the Γ valley. The basic Fick's second law is used to model the diffusion,

$$\frac{\partial n(x)}{\partial t} = \frac{\partial}{\partial x}\left(D_{(DV)^-}(n)\frac{\partial n}{\partial x}\right). \tag{3}$$

The boundary condition across the Ge/SiO_2 interface is characterized by the surface loss flux in the following expression:[24]

$$\Phi = -f_P(n_{P(Ge)} - n_{p(SiO_2)}), \tag{4}$$

where f_P is the loss rate, $n_{P(Ge)}$ is the phosphorous concentration on the surface of Ge, and $n_{P(SiO_2)}$ is the phosphorous concentration on the surface of SiO_2. For the short RTA time, we assume f_P is constant.

Since Ge is grown on Si using a 60 nm undoped Ge buffer, we have to consider the buffer layer with a high dislocation density as a sink for phosphorous diffusing into this layer. A pile-up model is described by Fahey[24] and Normand[25] based on the McNabb and Foster model. This model is used by Tsouroutas *et al.*[15] to simulate the phosphorous pile-up phenomenon close to the Ge surface during the diffusion. Using a similar approach, Fick's second law has to be modified,[15]

$$\frac{\partial n(x,t)}{\partial t} + P(x)\frac{\partial \Phi(x,t)}{\partial t} = \frac{\partial}{\partial x}\left(D_{(DV)^-}(n)\frac{\partial n(x,t)}{\partial x}\right), \tag{5}$$

$$\frac{\partial \Phi(x,t)}{\partial t} = kn(x,t)(1-\Phi(x,t)), \tag{6}$$

where $P(x,t)$ is the concentration of the traps related to dislocations and $\Phi(x,t)$ is the fraction of traps that is occupied with dopants at time t. Equation (6) holds that the trapping is permanent and k is a measure of the trapping rate. Since the diffusion coefficient is carrier concentration dependent in the extrinsic diffusion region, a simple erf function, which assumes a constant diffusivity, is not applicable for the modeling. The simulations of phosphorous diffusion in Ge were performed using the finite difference time domain (FDTD) method.

A box-shaped profile of dopant distribution normally appears in bulk Ge with increasing doping level,[22] however, in thin Ge films the profiles (Fig. 3) tend to have an even distribution of dopants. The good fit between simulation and experiment shows that the dopant diffusion coefficient $D_{(DV)}{}^-$ increases quadratically with the carrier concentration, n, in the extrinsic diffusion region. To study the doping effect on the phosphorous diffusion, we prepared Ge samples with different background doping levels. As shown in Fig. 4, two as grown films have the same delta doped layers as the diffusion source but one film (full squares) has an intrinsic Ge layer with phosphorous concentration below $2 \times 10^{17}\,\text{cm}^{-3}$ and the other film (full circles) has an *in situ* doped Ge layer with phosphorous concentration of $\sim 1 \times 10^{19}\,\text{cm}^{-3}$. Under the same thermal treatment, it is obvious that phosphorous in *in situ* doped Ge (open circles) diffuses faster than that in intrinsic Ge (open squares).

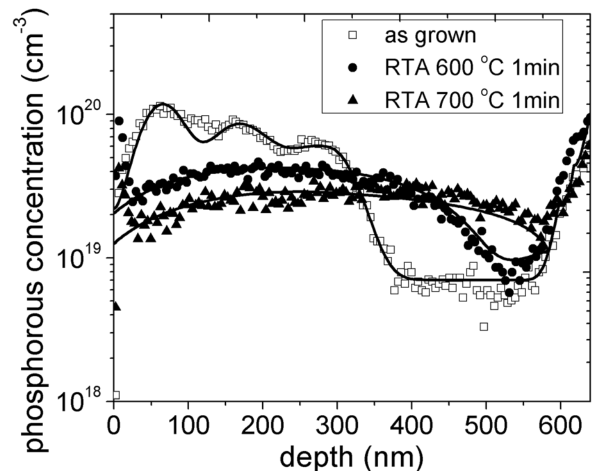

FIG. 3. SIMS depth profile of phosphorous in Ge for an as grown sample (open squares) and after RTA at 600 °C (full circles), and 700 °C (full triangles) for 1 min. The solid lines represent best fits from FDTD simulations.

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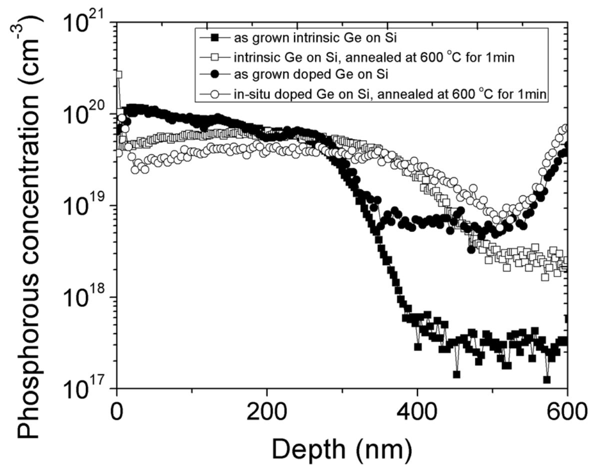

FIG. 4. SIMS depth profiles of phosphorous from different Ge-on-Si films. Phosphorous in intrinsic Ge with the delta doped layer on top as grown (full squares) and annealed at 600 °C for 1 min (open squares). Phosphorous in *in situ* doped Ge with the delta doped layer on top as grown (full circles) and annealed at 600 °C for 1 min (open circles).

Considering that the diffusion source and boundary conditions are the same, we conclude that the enhanced diffusion is due to the background doping level.

The intrinsic diffusion coefficients are illustrated in Fig. 5, deduced from the best fit for annealing temperatures of 600 °C, 650 °C, and 700 °C against the diffusivity extracted for the same temperature range from recent publications of other groups. As shown in Fig. 5, the intrinsic diffusivities from our experiments have comparable values to the results from other groups. Generally, the diffusivity versus temperature shows an Arrhenius behavior and hence, we use $D = D_0^* e^{-E_a/kT}$ to fit the three data points. The activation energy is $E_a = 1.98$ eV and the pre-exponential coefficient is $D_0^* = 2.2 \times 10^{-4}$ cm^2/s, similar to the literature values. The intrinsic carrier concentration in tensile strained Ge is 2.09×10^{17} cm^{-3} at 600 °C and 3.88×10^{17} cm^{-3} at 700 °C. Due to the *in situ* doping during growth, the carrier concentration in the Ge layer before annealing is increased to 1×10^{19} cm^{-3}. Therefore, the extrinsic diffusivities in the *in situ* doped Ge region, shown as the open squares in Fig. 5, are about 2 orders of magnitude higher than the diffusivities in intrinsic Ge.

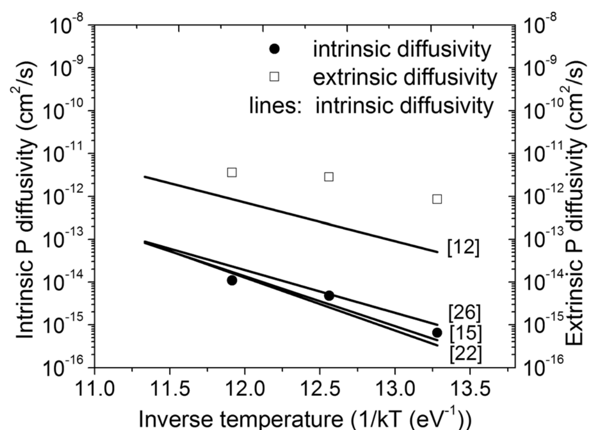

FIG. 5. Full circles represent the intrinsic diffusivity of phosphorous in Ge from this work. Open squares represent the extrinsic diffusivity of phosphorous in Ge from this work. Intrinsic diffusivities from other groups[12,15,22,26] for temperature ranging from 600 °C to 750 °C are plotted by lines as comparisons. The estimated standard error for the extracted diffusivity is ± 5%.

IV. CONCLUSIONS

We have investigated the background doping effect on the phosphorous diffusion in Ge. Phosphorous diffusion is enhanced by a factor of 100 when background doping is 1×10^{19} cm^{-3} in Ge. We built an FDTD modeling to simulate phosphorous diffusion which fits the depth profile measured by SIMS very well; we find that the diffusivity varies quadratic with carrier concentration. We show that we can achieve a uniform phosphorous distribution with the concentrations above 3×10^{19} cm^{-3} by utilizing phosphorous delta-doped layers created in a UHVCVD growth process as a dopant reservoir and then annealing. The dopant enhanced in-diffusion defeats the net loss by out-diffusion from the Ge surface and the pileup at Ge/Si interface. This concentration is significantly higher than that can be achieved in a CVD growth process, while preserving high quality Ge thin films that can be used as gain medium for CMOS compatible Ge lasers.

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