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Diffusion and electrophysical properties of rare earth elements of erbium, thulium, samarium and ytterbium in silicon

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ABSTRACT: Carried out by complex methods of labeled atoms, autoradiography, measurement of electrical conductivity and the Hall effect, isothermal relaxation of capacitance and current, diffusion, solubility and electrophysical properties of rare-earth elements: erbium, thulium, samarium and ytterbium in silicon were studied in various annealing media and for a wide temperature range (1100 - 1250 ^0C). Diffusion parameters, solubility were established, and a small acceptor nature was revealed on the surface and in the bulk of *n*-type silicon, of the studied rare-earth impurities of erbium, thulium, samarium, and ytterbium in silicon.

KEYWORDS: silicon, electrophisical properties, rare earth elements, silicon stability, diffusion, solubility of impurities, radioactivity isotope, treatment, diffusion coefficient diffusion, temperature dependence of the diffusion.

I. INTRODUCTION

It is known that silicon doped with rare-earth elements (*REE*) is attracting increasing attention of researchers as a promising material for optoelectronics. This is due to the prospect of using *Si<REE>* structures in silicon optoelectronics as light sources, for example, for *Si<Er>* m, which corresponds to the minimum loss and dispersion of fiber-optic communication lines, and from the point of view of structural features electronic shells of this group of elements [1-4] at a wavelength of 1.54 μm. The effectiveness of REE impurities in silicon, the manifestation of the optical properties of structures depends on the spectrum of optically and electrically active centers containing REEs, the total concentration and diffusion mechanism of rare-earth elements.

II. DESCRIPTION AND RESULTS OF THE RESEARCH

The aim of this work was a comprehensive study of the diffusion, solubility, and electrical properties of the impurities of erbium, thulium, samarium, and ytterbium in silicon. A metal layer of the radioactive isotope of erbium - ^{169}Er, thulium - ^{170}Tm, samarium - ^{153}Sm and ytterbium - ^{175}Yb was sprayed onto the surface of *n*-type silicon samples with specific resistance ρ = 15• Ω ·cm, or a layer of erbium, thulium, samarium and ytterbium - labeled chloride was deposited from the solution the radioactive isotope of erbium is ^{169}Er, thulium is ^{170}Tm, samarium is ^{153}Sm and ytterbium is ^{175}Yb, also in other batches of silicon samples (sample area ~ 1.5÷2.5 cm^2, thickness ~ 350 ÷ 380 μm).

For electrical studies, diffusants were stable isotopes of metallic erbium, thulium, samarium, and ytterbium, or salts of trivalent chloride of erbium, thulium, samarium, and ytterbium. Diffusion annealing was carried out in air, in evacuated ampoules (~ 10^{-5} ÷ 10^{-7} mm Hg), as well as in evacuated ampoules in an argon atmosphere in the temperature range 1100 ÷ 1250 ^{0}C. The duration of diffusion annealing varied depending on the diffusion temperature from 5 to 48 hours.

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After diffusion annealing, the samples were repeatedly washed in hydrofluoric acid, aqua regia, and also in a boiling mixture of H_2O_2 : *HCl*. After that, the edges of the sample were smoothed to a depth (~ 100 μm), significantly exceeding the diffusion depth (~ 10 μm). The profile of diffusants was determined by the method of partitioning - etching of thin layers (in a HF : HNO_3 solution = 1 : 50 with washing in a H_2O_2 : *HCl* mixture) and measuring the residual activity of the sample. The residual activity of the sample was measured using a small background setup UMF-1500M with a SBT-11 counter. The identification of the energy spectra of *REE* impurities: ^{169}Er, ^{170}Tm, ^{153}Sm, and ^{175}Yb was also carried out by measurements on the AI-1024 pulse analyzer (Physico-Technical Institute named after A.F. Ioffe RAS, St. Petersburg). The thickness of the removed layers (0.05 ÷ 0.5 μm) was determined by weighing the sample.

Autoradiograms obtained before and after annealing, as well as during the removal of the layers, indicated a uniform distribution of impurities ^{169}Er, ^{170}Tm, ^{153}Sm and ^{175}Yb, over the cross section of the sample and the absence of inclusions. Assuming that Fick's law is observed and that the surface concentration of the impurity does not change with time (diffusion from a constant source into a semi-bounded body), the diffusion coefficient (D) of impurities ^{169}Er, ^{170}Tm, ^{153}Sm, and ^{175}Yb in silicon is determined. To this end, the experimental curve of the residual amount of impurities was approximated by a theoretical curve for diffusion from a constant source:

$$Q(x)=2C_0\sqrt{Dt}\ \text{ierfc}(x/2\sqrt{Dt}) \quad (1)$$

here, C_0 is the surface concentration (1), x is the total thickness of the removed layers, t is the diffusion time. The concentration profile of charge carriers was determined by measuring the conductivity and the Hall effect, isothermal relaxation of the capacitance and current of diffusion studies, etching thin layers and measuring the conductivity and the Hall effect. The concentration of charge carriers $p(x)$ was determined by the formula:

$$p(x)=\frac{1}{e}\left(\frac{d\sigma_S}{dx}\right)^2\cdot\left[\frac{d}{dx}\left(R_S\sigma_S^2\right)\right]^{-1} \quad (2)$$

Here R_s - is the measured (effective) Hall coefficient; σ_S is surface conductivity; e - is the electron charge. Note that this formula (2) is given in [5] in a more general form, taking into account the difference between the Hall and drift mobilities:

$$p(x)=\frac{1}{e}\left(\frac{d\sigma_S}{dx}\right)^2\cdot\left[\frac{d}{dx}\left(R_S\sigma_S^2\right)\right]^{-1} \quad (3)$$

The analysis of the obtained data was carried out under the assumption that Fick's law is satisfied and the surface concentration of the impurity does not change with time (diffusion from a constant source: $c(0,t)$ = const). Under these assumptions and taking into account the fact that the thickness of the sample far exceeds the diffusion depth, the curve $c(x)$ is described by the function *erfc*.

As the results show, these distributions for: erbium [6], thulium [7], samarium [8] and ytterbium [9] in silicon are a sharply declining curve, satisfactorily approximated by the erfc function corresponding to the solution of the Fick equation for diffusion from a constant source into a semi-limited body.

The concentration of charge carriers $p(x)$ in silicon doped with erbium, thulium, samarium, and ytterbium was determined taking into account the mobility of charge carriers. In this case, the complete ionization of *Er*, *Tm*, *Sm*, and *Yb* impurities in silicon was assumed, i.e. it was believed that the concentration of *Er*, *Tm*, *Sm*, and *Yb* $c(x)$ is equal to the concentration of charge carriers $p(x)$: $c(x) = p(x)$. The thickness of the removed layers was determined by weighing the sample on the balance.

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As in previous studies of impurities of europium [10], praseodymium [11] and scandium in silicon [12], electrical measurements taken at several points on the surface indicated a uniform distribution of the impurity over the sample cross section and the absence of second phase inclusions. The results obtained, the form of the distribution for *Er*, *Tm*, *Sm*, and *Yb* in silicon, are a sharply declining curve that is satisfactorily approximated by the erfc function corresponding to the solution of the Fick equation for diffusion from a constant source into a semi-bounded body. The concentration of charge carriers *p(x)* in silicon doped with *Er*, *Tm*, *Sm*, and *Yb* was determined taking into account the mobility of charge carriers [5].

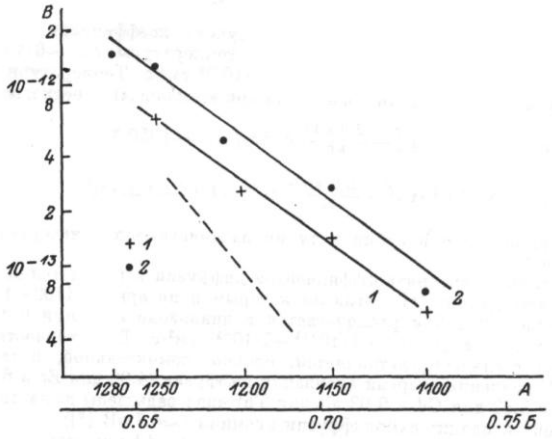

Fig. 1. Temperature dependence of the diffusion coefficient of erbium (1) and thulium (2) in silicon. A - temperature, ^{0}C; B - 10^3□ T, K^{-1}; B – *D*, cm^2/c.

Also, when determining diffusion parameters, along with doped samples, control samples of *n-Si* (SEP -15) were also studied. In the control samples, changes in the type of conductivity and conductivity values are not observed. This confirms that during annealing, it is precisely the *Er*, *Tm*, *Sm*, and *Yb* impurities that diffuse into silicon. The temperature dependence of the diffusion coefficient for the corresponding *Er*, *Tm*, *Gd*, and *Yb* impurities in silicon is of an Arrhenius character and can be described by the following relationships:

$D_{Er} = 2\cdot10^{-3}\exp(-2.9\ eV/kT)$ cm^{2}s^{-1};
$D_{Tm} = 8\cdot10^{-2}\exp(-3.0\ eV/kT)$ cm^{2}s^{-1};
$D_{Sm} = 4\cdot10^{-3}\exp(-2{,}8\ eV/kT)$ cm^{2}s^{-1};
$D_{Yb} = 3\cdot10^{-2}\exp(-3{,}0\ eV/kT)$ cm^{2}s^{-1}.

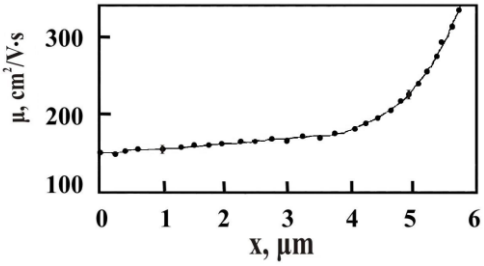

Fig. 2. Carrier mobility distribution profile in *Si*<*Sm*>. T = 1150^0 C, t= 10 h.

The penetration depth of *Er*, *Tm*, *Sm*, and *Yb* impurities in silicon, for the evaluation of which we used a value of $2\sqrt{Dt}$, does not exceed several micrometers in the entire temperature range studied. The surface concentrations of REE impurities: *Er*, *Tm*, *Sm*, and *Yb* in silicon in this case are ~ 10^{18}-10^{19} cm$^{-3.}$

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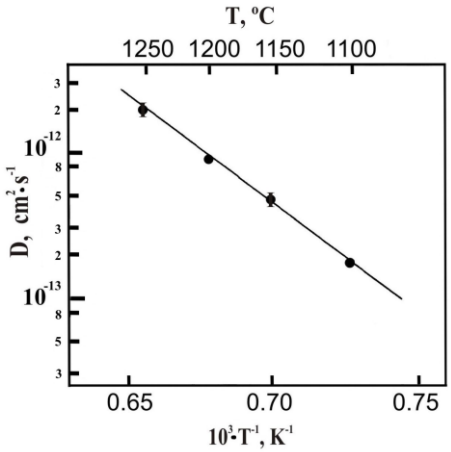

Fig. 3. Temperature dependence of the diffusion coefficient of samarium in silicon.

As the analysis of the data shows, the diffusion coefficients depending on the temperature and activation energy of *Er*, *Tm*, *Sm*, and *Yb* impurities in silicon are in the range of values characteristic of the diffusion of typical elements of group III *B* of the periodic system, diffusing along the lattice sites. A comparison of our data on the diffusion of *Er*, *Tm*, *Sm*, and *Yb* impurities in silicon with the early results on the diffusion of other *REE*s in silicon obtained using radioactive and other methods used above shows that the diffusant deposition method and diffusion medium do not significantly affect diffusion parameters of rare earth elements in silicon.

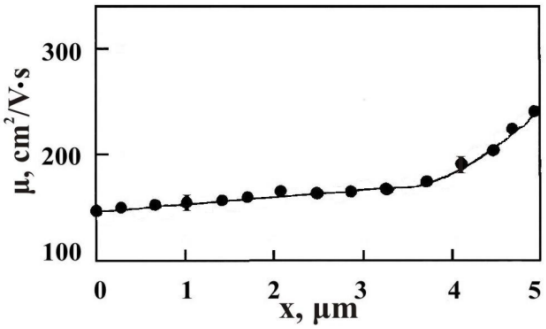

Fig. 4. The profile of the distribution of carrier mobility in *Si<Yb>*. $T = 1150^0$ *C*, $t = 10$ *h*.

III. CONCLUSION

It was found that *Er*, *Tm*, *Sm*, and *Yb* impurities, like the other rare-earth elements studied by us, exhibit a small acceptor nature on the surface and in the bulk of *n*-type silicon after diffusion annealing. Using the Hall effect, we determined the mobility of charge carriers — holes in diffusion-doped *Er*, *Tm*, *Sm*, and *Yb* impurities in silicon layers. The mobility of charge carriers in the diffusion layers *Si<Er>*, *Si<Tm>*, *Si<Sm>* and *Si<Yb>* ~ 140 ÷ 240 $cm^2/V \cdot s$ is and decreases with increasing concentration of *Er*, *Tm*, *Sm*, and *Yb* impurities in silicon.

According to the temperature dependence of the Hall coefficient - the concentration of charge carriers, as well as by the method of isothermal relaxation of the capacitance and current, no deep levels in the band gap of silicon characteristic of *Er*, *Tm*, *Sm*, and *Yb* in silicon were detected. The concentration distribution of charge carriers in silicon layers doped with *Er*, *Tm*, *Sm*, and *Yb* impurities, determined by electrical measurements, can be described by the relations:

$$C \approx (2 \div 4) \cdot 10^{17} \, erfc(x / 2\sqrt{Dt}), \ cm^{-3}.$$

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In this case, the surface concentration of *Er*, *Tm*, *Sm*, and *Yb* atoms in silicon, determined using the method of labeled atoms, is ~ $10^{18} \div 10^{19}$ cm^{-3}. Thus, the concentration of electrically active *REE*s of *Er*, *Tm*, *Sm*, and *Yb* impurities in silicon is less than 10% of the total concentration of *Er*, *Tm*, *Sm*, and *Yb* impurities in silicon.

As an analysis of the data shows, the diffusion coefficients and activation energies of *Er*, *Tm*, *Sm*, and *Yb* impurities in silicon are in the range of values characteristic of the diffusion of typical elements of group III, as well as for other *REE*s that are substitutional impurities and diffuse along the lattice sites. This suggests that the *REE* of *Er*, *Tm*, *Sm*, and *Yb* impurities of the same group is also an impurity of substitution and diffuses like other *REE*s along the sites of the silicon lattice (tabl. 1).

Table 1. Diffusion coefficients of *Er*, *Tm*, *Gd*, and *Yb* in silicon.

Diffusion temperature, ^0C	Diffusion coefficients of in silicon; D, $cm^2 \cdot c^{-1}$			
	Erbium (*Er*)	Thulium (*Tm*)	Samarium (*Sm*)	Ytterbium (*Yb*)
1100	$5{,}9 \cdot 10^{-14}$	$6.4 \cdot 10^{-14}$	$2 \cdot 10^{-13}$	$3 \cdot 10^{-13}$
1150	$1.2 \cdot 10^{-13}$	$2.8 \cdot 10^{-13}$	$4{,}9 \cdot 10^{-13}$	$7{,}2 \cdot 10^{-13}$
1200	$2.6 \cdot 10^{-13}$	$4.9 \cdot 10^{-13}$	$9{,}6 \cdot 10^{-13}$	$1{,}6 \cdot 10^{-12}$
1250	$7.1 \cdot 10^{-13}$	$1.4 \cdot 10^{-13}$	$2 \cdot 10^{-12}$	$4 \cdot 10^{-12}$

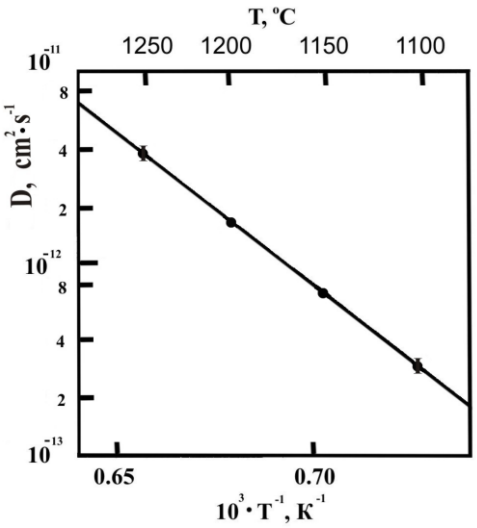

Fig. 5. Temperature dependence of the diffusion coefficient of ytterbium in silicon.

Comparison of the obtained data with the early results on the diffusion of REEs in silicon, obtained using radioactive and other methods, shows that the method of applying a diffusant and the diffusion medium do not significantly affect the diffusion parameters of *REE*s in silicon. Thus, we can assume that the *REE* impurities *Er*, *Tm*, *Sm*, and *Yb*, elements of the third group of the periodic system that we studied, are also substitutional impurities and diffuse along the lattice sites (vacancy diffusion mechanism) of silicon.

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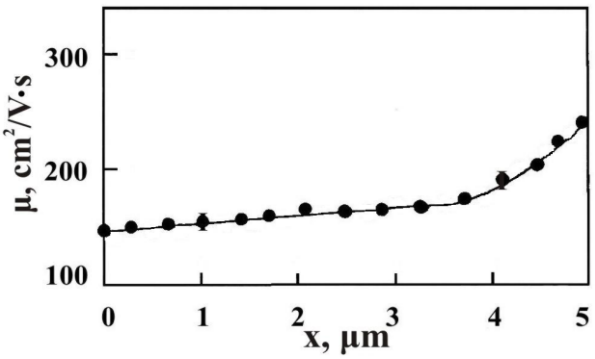

Fig. 6. The profile of the distribution of carrier mobility in *Si<Yb>*. $T = 1150^0 C$, $t = 10$ *h*.

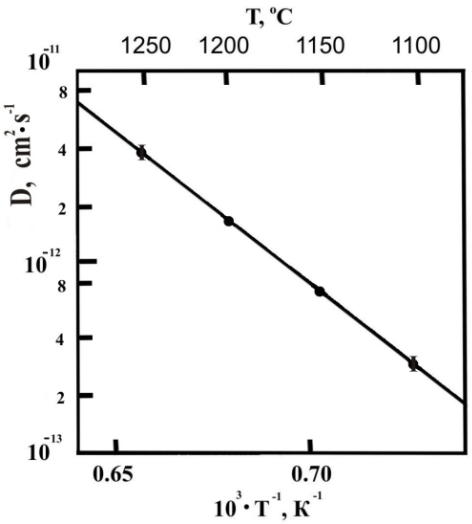

Fig. 7. Temperature dependence of the diffusion coefficient of ytterbium in silicon.

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