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Magnetic Susceptibility of $Si_{0,97}Ge_{0,03}$ Filamentous Crystals Irradiated by Protons

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The article deals with the filamentous $Si_{0,97}Ge_{0,03}$ crystals with transverse dimensions of 40 ± 2 μm grown by the method of chemical transport reactions in the closed bromide system using gold as a growth initiator. The focus of research was the influence of proton irradiation with doses up to $1\cdot10^{17}$ p^+/cm^{-2} and the following thermal treatments at temperatures of 200 - 500°C on the magnetic susceptibility of these crystals. The dependence of the magnetic susceptibility on the intensity of the magnetic field of the proton irradiated filamentous $Si_{0,97}Ge_{0,03}$ crystals is described within the framework of the Langevin atom paramagnetism model and explained by the formation of defects of the vacancy type. The revealed increase in the radiation stability of $Si_{0,97}Ge_{0,03}$ crystals followed the combined effect of radiation and subsequent thermal treatments.

Keywords: silicon-germanium, filamentous crystals, proton irradiation, thermal annealing, magnetic susceptibility.

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Introduction

Filamentous crystals (FC) are widely used in various fields of human practice (aviation, rocket and space, medical, transport, telecommunications, etc.) due to their unique shape, size, perfect structure, high elasticity and mechanical strength.

Studying the behavior of crystals under the influence of radiation, proton irradiation in particular, is interesting in terms of the creation of radiation-resistant sensors [1, 2]. On the other hand, high perfection of the structure of filamentous crystals (FC) allows us to simulate the defects formed in the crystals in the process of irradiation [3-5].

The purpose of this work was to study the effects of proton irradiation with doses up to $1\cdot10^{17}$ p^+/cm^2 and annealing at temperatures of 200 - 500ºC on the magnetic susceptibility of filamentous $Si_{1-x}Ge_x$ (x = 0.03). Samples of this very composition were chosen for the experiment since they are characterized by high perfection of structure and high micromechanical characteristics [6, 7].

I. Experiment methodology

The growth of $Si_{1-x}Ge_x$ FC was carried out by the method of chemical transport reactions in a closed bromide system using gold as the initiator of growth [8]. A quartz ampoule was loaded with growth material (silicon, germanium), alloying admixtures (boron) and halogen (bromine), which was used as a transport agent. The ampoule was pumped to a pressure of 10^{-5} mm Hg and placed in a tubular furnace with a temperature gradient with the temperature of the source zone reaching 900°C and the crystallization zone ranging from 550-750°C. The content of germanium in a solid solution of $Si_{1-x}Ge_x$ was determined by the method of microprobe analysis on the CAMEBAX installation and amounted to x = 0.03 molar percent. For research, we selected FC with a diameter of 40 ± 2 μm and a length of 4-5 mm. These crystals are of p-type conductivity with a specific impedance ρ = 0.018 Ohm·cm.

Crystals were irradiated with 6 MeV protons at doses of $5\cdot10^{13}$ p^+/cm^{-2}, 10^{15} p^+/cm^{-2} and $1\cdot10^{17}$ p^+/cm^{-2} at 40°C on a cyclotron U-120 of the Institute of Nuclear Research of the National Academy of Sciences of Ukraine.

Magnetic susceptibility (MS) of filamentous crystals was studied by the Faraday method in the range of magnetic fields of 0.2-5 kV.

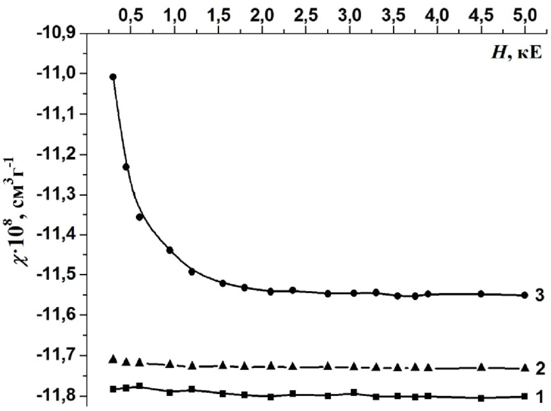

Fig. 1. The dependence of the magnetic susceptibility on the intensity of the magnetic field of the filamentous crystals $Si_{0,97}Ge_{0,03}$: 1 – the original sample, 2 – irradiated with a dose of $5\cdot10^{15}$ p^+/cm^2, 3 – irradiated with a dose of $1\cdot10^{17}$ p^+/cm^2.

II. Experimental results and their discussion

Fig.1 shows the obtained experimental results. Irradiation with the smallest dose did not lead to the change of magnetic susceptibility. An increase in the radiation dose to $5\cdot10^{15}$ p^+/cm^2 leads to the appearance of the paramagnetic component of the MS (Fig. 1, curve 2). According to its linear nature, it can be concluded that in the process of FC irradiation there form dispersed paramagnetic centers between which there is no interaction. At a radiation dose of $1\cdot10^{17}$ p^+/cm^2 in addition to the paramagnetic component there appears nonlinearity of the dependence of magnetic susceptibility on the intensity of the magnetic field. This proves that in this case, along with the dispersed paramagnetic centers, some of their clusters are formed in crystals. They behave like Langevin paramagnetism of atoms which possess a magnetic moment. The only difference is that their magnetic moments are 10^3-10^5 times bigger than the magnetic moment of individual atoms. As is known, irradiation with protons does not only lead to the formation of various kind of point defects in the material but also to the so-called areas of disordering, which, obviously, can serve as the centers of origin of magnetic nanoclusters.

For the analysis of the experimental dependences of magnetic susceptibility from the magnetic field intensity (Fig. 1, curve 3) we used the following theoretical model [9].

$$c(H)=N_C M_C\cdot\left(\frac{M_C}{kT}\cdot\left(1-\mathrm{cth}^2\left(\frac{M_C}{kT}\cdot H\right)\right)+\frac{kT}{M_C\cdot H^2}\right)+ \quad (1)$$

$$+c_{par}+c_{cell}$$

where N_C – concentration of magnetically ordered clusters; k – Boltzmann constant, T – temperature; $M_C = N_0 M_B g\sqrt{s(s+1)}$ – cluster magnetic moment; N_0 – number of paramagnetic centers in one magnetic cluster, M_B – the Bohr magneton, g – g-factor (we accept g = 2), s – spin of the paramagnetic center of which the cluster consists (we accept s = 1/2); c_{par} – paramagnetic component; c_{cell} – lattice susceptibility.

Approximating the experimental dependences $c(H)$ by theoretical expression (1), the corresponding values are estimated. The results are presented in Table 1.

In this model, we assumed in the first approximation that the magnetic moments of the clusters are identical. However, obviously, there can be a certain distribution of clusters according to the magnitude of their magnetic moments [10]. The theoretical expression that describes the magnetically-ordered component of the experimental dependence of χ (H) (Fig. 1, curve 3) can be represented as:

$$c^{\mathrm{teor}}=\int_0^{\infty} M_C\, f(M_C)\left(\frac{M_C}{kT}\left(1-\mathrm{cth}^2\left(\frac{M_C H}{kT}\right)\right)+\frac{kT}{M_C H^2}\right)dM \quad (2)$$

where $f(M)$ – magnetic moment distribution; $f(M)dM$ – concentration of particles with a magnetic moment from M to $M + dM$.

In most cases function $f(M)$ is taken as logarithmically normalized [11, 12], that is

$$f(M_C)=\frac{n}{\sqrt{2p}\,s}\frac{1}{M_C}\exp\left[-\frac{\ln^2(M_C/\langle M_C\rangle)}{2s^2}\right]. \quad (3)$$

The distribution function $f(M_C)$ is determined by the three parameters n, s, $\langle M_C\rangle$. Therefore, the construction of the distribution was reduced to finding these parameters. One way to determine them is to use the least squares method, which is to minimize the expression

$$\Delta_c=\sum_{i=1}^{N}\left(c^{\mathrm{teor}}(H_i)-c^{\exp}(H_i)\right)^2, \quad (4)$$

Table 1

Calculated parameters based on the results of approximation of experimental dependencies χ (H) by the theoretical expression (1)

№	Sample $Si_{0,97}Ge_{0,03}$	c_{par}, $cm^3\cdot g^{-1}$	N_0, 1/cluster	N_C, cm^{-3}	D, nm
1	Output	–	–	–	–
2	F = $5\cdot10^{15}$ p^+/cm^2	$0{,}1\cdot10^{-8}$	–	–	–
3	F = $1\cdot10^{17}$ p^+/cm^2	$0{,}24\cdot10^{-8}$	$1{,}62\cdot10^4$	$1{,}85\cdot10^9$	6

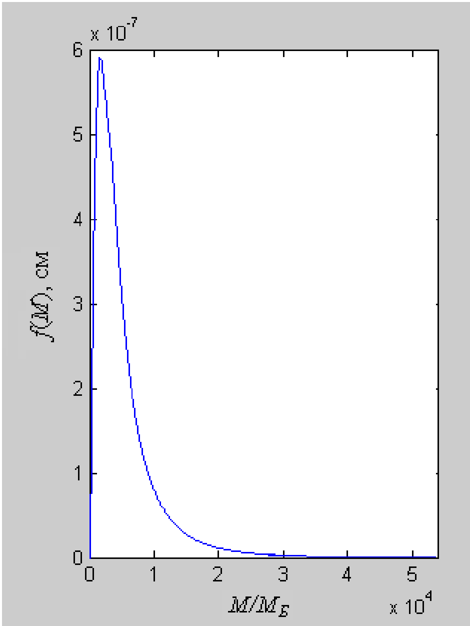

Fig. 2. Construction of the clusters distribution functions by the magnitude of their magnetic moments.

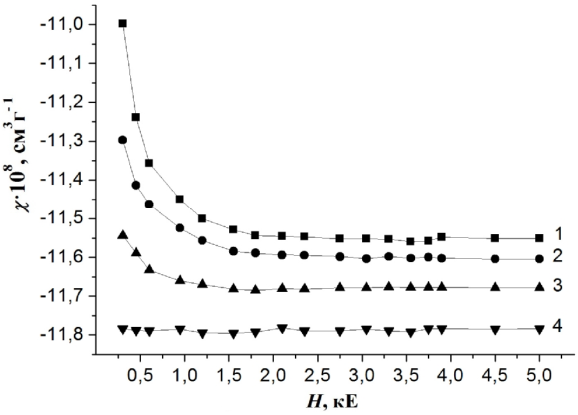

Fig. 3. Dependence of magnetic susceptibility of filamentous $Si_{0,97}Ge_{0,03}$: 1 – irradiated 6.8 MeV protons with a dose of $1{\cdot}10^{17}$ p^+/cm^2; 2 – after annealing at 200°C; 3 – after annealing at 300°C; 4 –after annealing at 500°C.

where $c^{\text{teor}}(H_i)$ – is determined from (2), $c^{\exp}(H_i)$ – experimental values of magnetic susceptibility, N – number of experimental points.

From the construction of the distribution function of clusters by their magnitude (Fig. 2) we determine the most probable cluster sizes (D, nm). The results of calculations are given in Table 1.

As is well-known [13], the main types of defects in monocrystalline silicon are: vacancy-oxygen complex (A-center), divacancy, donor-vacancy complex, boron-vacancy complex. At proton irradiation, not only point radiation defects in the silicon samples are formed but also, the so-called, divacancy type disorder areas [14]. The authors [15] observed the three stages of the annealing of the divacancy in the temperature interval of 100-200°C with activation energy of 1.0 eV in the study of the silicon divacancy annealing; 200-300°C – 1.3 eV and 300-500°C – 1.5 eV. This is connected with the different divacancy positions: in the cluster core, in the spatial charge cluster area and in the conducting silicon matrix.

We had a sample of irradiated dose of $1{\cdot}10^{17}$ p^+/cm^2 annealed at 100°C, 200°C and 500°C lasting five hours each. Heat treatment of the sample was carried out in a tubular furnace on air. We believe that the airborne annealing does not affect the formation of bulky defects in Si, since, as shown by the authors [16], the oxidation of the surface of the samples can lead to the additional generation of interstitial silicon atoms from the boundary between the separation of silicon and silicon in the crystal volume only with two-stage heat treatment with repeated annealing at temperatures above 1100°C for a duration of more than ten hours.

The obtained experimental results are presented in Fig.3 and parameters N_0 and N_C determined by formula (1) after each stage of annealing – in Table 2. As we can see, thermal treatment of protons irradiated with protons leads to the annealing of radiation defects. After heat treatment at a temperature of 500°C, the value of the magnetic susceptibility of the irradiated sample (Fig.3, line 4) practically coincides with the original sample (Fig.1, line 1).

Thus, the peculiarities of the magnetic susceptibility of $Si_{0,97}Ge_{0,03}$ FC irradiated by protons can be explained by the formation of the vacancy type defects.

It is interesting to note that with repeated irradiation of samples $Si_{0,97}Ge_{0,03}$, which passed all stages of annealing in the range of 300-500°C, there is significantly less influence of irradiation on their magnetic susceptibility (Fig. 4, Table 3). This indicates an increase in the radiation resistance of the crystals, thus opening new prospects for the practical use of these materials in electronic technology, micro-sensors, etc. The reason for this is the presence of oxygen-containing complexes in these samples (deep levels of divacancy and trivacancy) [17, 18], which apparently did not completely evaporate at a temperature of 500°C and

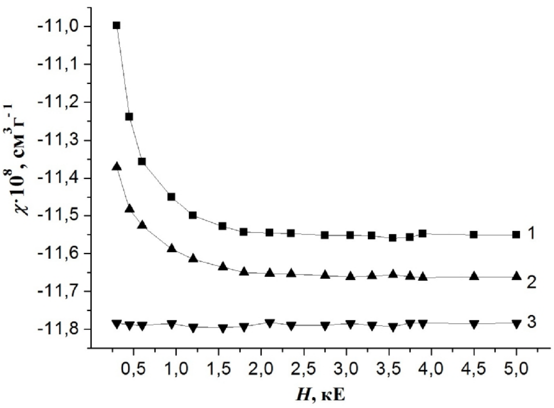

Fig. 4. Dependence of magnetic susceptibility of $Si_{0,97}Ge_{0,03}$ FC: 1 – crystals irradiated with 6.8 MeV protons at a dose of $1{\cdot}10^{17}$ p^+/cm^2; 2 – re-irradiated 6.8 MeV protons with a dose of $1{\cdot}10^{17}$ p^+/cm^2, pre-annealed at 500°C; 3 – original samples.

Table 2

The parameters of the annealed samples $Si_{0,97}Ge_{0,03}$ calculated by the results of the approximation of the experimental dependences χ (H) (Figure 2) by theoretical expression (1)

№	Sample $Si_{0,97}Ge_{0,03}$	c_{par}, $cm^3 \cdot g^{-1}$	N_0, 1/cluster	N_C, cm^{-3}	D, nm
1	F = $1 \cdot 10^{17}$ p^+/cm^2	$0.24 \cdot 10^{-8}$	$1.62 \cdot 10^4$	$1.85 \cdot 10^9$	6
2	annealed at 200°C	$0.18 \cdot 10^{-8}$	$1.58 \cdot 10^4$	$1.41 \cdot 10^9$	5.8
3	annealed at 300°C	$0.11 \cdot 10^{-8}$	$1.6 \cdot 10^4$	$8.2 \cdot 10^8$	5.7
4	annealed at 500°C	–	–	–	–

Table 3

The calculated parameters of the irradiated samples $Si_{0,97}Ge_{0,03}$ by the results of the approximation of the experimental dependences χ (H) (Fig. 3) by theoretical expression (1)

№	Sample $Si_{0,97}Ge_{0,03}$	c_{par}, $cm^3 \cdot g^{-1}$	N_0, 1/cluster	N_C, cm^{-3}	D, nm
1	Φ = $1 \cdot 10^{17}$ p^+/cm^2	$0.24 \cdot 10^{-8}$	$1.62 \cdot 10^4$	$1.85 \cdot 10^9$	6
2	F = $1 \cdot 10^{17}$ $p^+/см^2$ pre-annealed at 500°C	$0.12 \cdot 10^{-8}$	$8.4 \cdot 10^3$	$9.1 \cdot 10^8$	4.7
3	Output Sample	–	–	–	–

serve as wastewater for radiation defects. However, these suggested assumptions need further research.

Conclusions

1. We evaluated the concentration and magnitudes of magnetically sensitive defects in irradiated specimens $Si_{0,97}Ge_{0,03}$ on the basis of the Langevin paramagnetism model of atoms.

2. Based on the conducted thermal treatments in the range of 200-500°C, we determined the vacancy nature of radiation defects in irradiated protons in $Si_{0,97}Ge_{0,03}$ FC and estimated the dynamics of their variation at different annealing temperatures of the irradiated specimen.

3. We found that repeated proton irradiation of samples $Si_{0,97}Ge_{0,03}$, which passed all the stages of annealing in the range of 300-500°C, had a significantly lesser influence on the change in their magnetic properties, which indicates to an increase in radiation resistance and opens up new prospects for the use of micron $Si_{1-x}Ge_x$ FC in electronic technology and micro-sensors.

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Магнітна сприйнятливість ниткоподібних кристалів $Si_{0,97}Ge_{0,03}$ опромінених протонами

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Методом хімічних транспортних реакцій в закритій бромідній системі, з використанням золота в якості ініціатора росту, вирощено ниткоподібні кристали $Si_{0,97}Ge_{0,03}$ поперечними розмірами 40 ± 2 мкм. Досліджено вплив протонного опромінення дозами до $1\cdot10^{17}$ $p^+/см^{-2}$ та наступних термічних обробок за температур 200 – 500 °C на магнітну сприйнятливість цих кристалів. Залежності магнітної сприйнятливості від напруженості магнітного поля ниткоподібних кристалів $Si_{0,97}Ge_{0,03}$, опромінених протонами, описано в рамках моделі ланжевенівського парамагнетизму атомів та пояснено утворенням дефектів вакансійного типу. Виявлено підвищення радіаційної стійкості кристалів $Si_{0,97}Ge_{0,03}$ після комбінованої дії опромінення та наступних термічних обробок.

Ключові слова: кремній-германій, ниткоподібні кристали, протонне опромінення, термічний відпал, магнітна сприйнятливість.