Magnetic and magnetoresistive characteristics of neutron-irradiated Si$_{0.97}$Ge$_{0.03}$ whiskers

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Abstract. The effect of $8.6 \times 10^{17}$ n/cm$^2$ with fast neutron irradiation on the magnetic susceptibility of Si$_{0.97}$Ge$_{0.03}$ thread-like crystals (whiskers) with impurity concentration near metal-insulator junction has been studied. Significant differences have been observed in the change of magnetic susceptibility of irradiated whiskers and bulk Cz-Si. The low-temperature (4.2…40 K) changes of magnetoresistance in magnetic fields up to 14 T, caused by irradiation, have been studied. It has been established that at temperatures near 4.2 K, a significant contribution to the conductivity is made by light charge carriers of low concentration but with high mobility. The level supplying these charge carriers has the energy of $\varepsilon = 2.1$ meV, and with application of magnetic field it increases up to $\varepsilon = 2.5$ meV in approx. 10 T field. It demonstrates the fact that the reason of magnetoresistance, beside the magneto-field decrease of mobility, is the magneto-field decrease in the free carrier concentration. At temperatures approx. 40 K, conductivity is due to holes, the activation energy whereof is $\varepsilon = 11.5$ meV, which is practically independent of the magnetic field.

Keywords: whiskers, silicon-germanium, neutron irradiation, magnetic susceptibility, magnetoresistance.

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1. Introduction

An important scientific task that has been not completely solved is producing physical value sensors, which are operable under unfavorable conditions (strong magnetic fields, low temperatures, irradiation) [1, 2]. On the other hand, as the structure of thread-like crystals (whiskers) is perfect, it allows to model defects, arising in crystals during irradiation [3].

The papers [4-6] present investigation results of the effect of strong magnetic fields as well as electron and $\gamma$-quantum irradiation on electric and magnetoresistive characteristics of Si$_{0.97}$Ge$_{0.03}$ whiskers. The research shows that the main defects in crystals are the secondary radiation defects of interstitial boron atoms. It has been found that the revealed changes of magnetoresistance are related to the defects arising during the irradiation process, which cause the dislocation of charge carriers in the crystal impurity zone; the optimal conditions for crystal doping have been defined to create sensitive deformation sensors, resistive to $\gamma$-quantum irradiation operable in strong magnetic fields.
Investigated in [7-9] is the effect of 6.8-MeV proton irradiation with up to $8.6 \times 10^{17} \text{p/cm}^2$ dose and annealing at 100...300 °C on the electric conductivity of $\text{Si}_{1-x} \text{Ge}_x$ ($x = 0.03$) whiskers with impurity concentration near the metal dielectric transition within the temperature range 4.2...300 K in 14-T magnetic fields. The conductivity mechanism at low temperatures in whiskers irradiated by the dose $5 \times 10^{17} \text{p/cm}^2$ due to the annealing series at 100 °C and 280 °C has been explained. The results have been explained within the suggested theory that irradiation leads to an increase in the concentration of impurity states of double filled carriers, which is accompanied by increase in the crystal conductivity. The annealing of samples, in its turn, destroys these states, resulting in the reduced conductivity of the upper Hubbard band, which leads to an increase in magnetoresistance and to a decrease in the value of negative magnetoresistance at 13 to 30 K. It has been shown that the essential magnetoresistance of SiGe whiskers irradiated by high doses (over $1 \times 10^{17}$) of 6.8 MeV protons is related to magneto-field decrease in the mobility of free charge carriers (holes).

At present, the neutron irradiation effect on the electro-physical properties of SiGe whiskers practically has not been studied.

The objective of the research is to study the magnetic and magnetoresistive properties of $\text{Si}_{0.97} \text{Ge}_{0.03}$ whiskers after the fast neutron irradiation dose $8.6 \times 10^{17} \text{n/cm}^2$.

2. Experimental results and discussion

Irradiation with the fast neutron effect dose $8.6 \times 10^{17} \text{n/cm}^2$ on magnetic and magnetoresistive properties of $\text{Si}_{1-x} \text{Ge}_x$ ($x = 0.03$) has been studied. The irradiation was carried out using the Pressurized water reactor (PWR) of the Institute for Nuclear Researches of the National Academy of Sciences of Ukraine. The whisker with the diameter of 30 to 40 µm, the length of 2 to 3 mm and of $p$-type conductivity with the resistivity close to $\rho = 0.018 \text{Ohm-cm}$ were selected for the experiment.

After irradiation, the magnetic susceptibility in the magnetic fields 0.3 to 4.0 kOe at the ambient temperature was measured.

Fig. 1 shows the magnetic susceptibility dependences on the value of magnetic field for neutron irradiated $\text{Si}_{1-x} \text{Ge}_x$ whiskers ($x = 0.03$) (curve 1) and initial samples. To compare the neutron irradiation effect on the magnetic susceptibility, Fig. 2 presents the same dependence for bulk samples of monocrystalline Cz-Si, grown in the direction (100) with the dissolved oxygen concentration $(7-8) \times 10^{17} \text{cm}^{-3}$ and resistivity $\sim 10 \text{Ohm-cm}$.

Comparing Fig. 1 and Fig. 2, one can see that the neutron irradiation effect is more essential in the monocrystalline silicon samples. In particular, the change in the magnetic susceptibility in Cz-Si is $\Delta \chi = 0.35 \times 10^{-8} \text{cm}^3 \text{g}^{-1}$, whereas in $\text{Si}_{1-x} \text{Ge}_x$ ($x = 0.03$) whiskers it is $\Delta \chi = 0.23 \times 10^{-8} \text{cm}^3 \text{g}^{-1}$. One can assume that they are more resistive to neutron irradiation.

The rise of non-linearity, depending on the magnetic susceptibility, with the magnetic field value for $\text{Si}_{1-x} \text{Ge}_x$ ($x = 0.03$) whiskers after neutron irradiation demonstrates formation of some clusters in addition to dispersed paramagnetic centers in crystals, which behave like Langevin paramagnetism of atoms possessing a magnetic moment $10^2...10^3$ times higher than the magnetic moment of individual atoms. As it is known, neutron irradiation causes formation not only of a variety of point defects in the material but also formation of the so-called regions of disorder, which, apparently, can serve as centers responsible for creation of magnetic nanoclusters.

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To analyze the experimental dependences of magnetic susceptibility on the magnetic field value \( \chi(H) \) (Figs 1 and 2, curves 2), we use the theoretical model, which is described in detail in \([10, 11]\):

\[
\chi(H) = \chi^{\text{ord}}(H) + \chi^{\text{par}} + \chi_L = \\
= N_{cl} m_{cl} L' \left( \frac{m_{cl} H}{kT} \right) + \chi^{\text{par}} + \chi_L, \quad (1)
\]

where \( \chi^{\text{ord}} \) is the magnetic susceptibility component depending on the field value, \( \chi^{\text{par}} \) – paramagnetic component independent of the magnetic field intensity, \( \chi_L \) – susceptibility of the lattice, \( N_{cl} \) – concentration of magnetically ordered clusters, \( m_{cl} \) – magnetic moment of one such cluster (we assume the magnetic moments of the clusters are the same in the first approximation), \( L'(x) \) – derivative from Langevin function, \( k \) – Boltzmann constant, \( T \) – temperature. \( m_{cl} = N_0 \mu_B \sqrt{s(s+1)} \), where \( N_0 \) is the amount of paramagnetic centers in one magnetic cluster, \( \mu_B \) – Bohr magneton, \( g \) – g-factor (for evaluation, we assume \( g = 2 \)), \( s \) – spin of paramagnetic center that makes up the cluster (we assume \( s = 1/2 \)).

Taking into account the above comments, the expression (1) takes the following form:

\[
\chi(H) = N_{cl} N_0 \mu_B g \sqrt{s(s+1)} \times \\
\left[ \frac{N_0 \mu_B g \sqrt{s(s+1)} \cdot H}{\sinh \left( \frac{N_0 \mu_B g \sqrt{s(s+1)} \cdot H}{kT} \right)^2} \right]^2 kT \\
+ \frac{kT}{N_0 \mu_B g \sqrt{s(s+1)} \cdot H^2} + \chi^{\text{par}} + \chi_L. \quad (2)
\]

As a result of approximation of experimental dependences with the theoretical expression (2) the respective values have been assessed. The results are given in Table.

Table. Calculated parameters of samples based on the results of approximation of the experimental dependence \( \chi(H) \) by using the theoretical expression (2).

<table>
<thead>
<tr>
<th>Sample</th>
<th>( \chi^{\text{par}}, \text{cm}^3 \cdot \text{g}^{-1} )</th>
<th>( N_0, \text{1/cluster} )</th>
<th>( N_{cl}, \text{cm}^3 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Si_{0.97}Ge_{0.03}</td>
<td>0.23\cdot10^{-8}</td>
<td>1.8\cdot10^{3}</td>
<td>8.6\cdot10^{8}</td>
</tr>
<tr>
<td>Cz-Si</td>
<td>0.35\cdot10^{-8}</td>
<td>1.9\cdot10^{3}</td>
<td>1.2\cdot10^{8}</td>
</tr>
</tbody>
</table>

As shown in Table, the effect of neutron irradiation is more significant in samples Cz-Si. After irradiation, they show a significantly higher paramagnetic \( (\chi^{\text{par}}) \) component of the magnetic susceptibility and concentration of magnetically ordered clusters. Judging by the fact that the amount of paramagnetic centers in a single magnetic cluster \( (N_0) \) in both types of samples is approximately the same, the magnetic moments of the formed nanoclusters are comparable. Further, the samples of Si_{1-x}Ge_x whiskers (\( x = 0.03 \)) were subjected to thermal treatment within the temperature range 250…350 °C with increments 50 °C for 30 min and magnetic susceptibility being measured after each increment. The results are presented in Fig. 3.

It has been ascertained that, after annealing of SiGe whiskers irradiated with neutrons at 350 °C, the value of their magnetic susceptibility coincides with that of the initial sample. This indicates the destruction of secondary radiation defects.

Measuring the magnetoresistance in the magnetic fields up to 14 T in the temperature interval from 4.2 up to 300 K was performed with the same samples in the International Laboratory of High Magnetic Fields and Low Temperatures (Wroclaw, Poland).

After irradiating the whisker samples, the resistance grew approximately by \( 10^5 \) times up to 5 MOhm at ambient temperature. It did not allow us to perform low-temperature measurements of the magnetoresistance. Therefore, the annealing of the samples within the temperature range 50…350 °C with the 25 °C increment were carried out. The whiskers annealed at each temperature for 20 min were cooled down to ambient temperature, and thereafter the resistance was measured. It has been found out that the resistance of the irradiated whiskers decreases drastically approaching that of the initial sample at temperatures higher than 280 °C (Fig. 4).
Thereafter low-temperature measurements of the resistance field dependence were carried out (Fig. 5). Magnetoresistance changes insignificantly in the weak magnetic fields and raises with the increase of the magnetic field as it is shown in the figure. A change in the magnetoresistance curvature has been also observed.

The change in magnetoresistance curvature from concave to convex at 4.22 K shows that light charge carriers (electrons or light holes) that have a high mobility and very low concentration contribute to this magnetoresistance, that is, they are free minority charge carriers [12].

Let us calculate the parameters of these minority carriers [12]:

$$\mu = \frac{\sqrt{3} 4\rho(B_n) - 1}{B_n 3\rho_0},$$

where $\mu$ and $n$ are the mobility and concentration of minority carriers, respectively, $B_n$ – a point of magnetic field of the magnetoresistance bend, $\rho(B_n)$ – resistivity in the point of magnetoresistance bend, $\rho_0$ – resistivity without field.

According to these formulas for $T = 4.22$ K, we obtain: $\mu = 1400 \text{ cm}^2/(\text{V} \cdot \text{s})$, $n = 3.9 \times 10^{10}$, that is, at this temperature a very small amount of light charge carriers with a high mobility contributes to the conductivity. Let us calculate conductivity ratio of minority carriers to majority carriers by using the formula [12]

$$\frac{\sigma_m}{\sigma_0} = 4 \left( \frac{\rho(B_n)}{\rho_0} - 1 \right),$$

whence we obtain $\sigma_m/\sigma_0 = 0.32$, that is, minority carriers make a considerable contribution to conductivity at this temperature. The energy of impurity activation that causes these charge carriers is calculated at temperature of resistance and is $\varepsilon = 2.1 \text{ meV}$ (Fig. 6). When the temperature increases, this level is quickly exhausted, and at 40 K it is the level with energy 11.5 meV that contributes to conductivity.

In order to define the influence of magnetic field on the parameters of free charge carriers, dependences $\ln R(1/T)$ were plotted at different magnetic field induction values (Fig. 6), whence the energy of impurities activation at these field values were calculated. The figure shows that $\ln R(1/T)$ curve slope
obviously increases at low temperatures, and it affirms the rise in the activation energy together with the magnetic field increase. At 10 T, this activation energy is 2.5 meV, and that means it is considerably higher than the activation energy without field (2.1 meV). It indicates the fact that resistance increases together with the magnetic field not only as a result of the reduction of free charge carriers’ mobility, which is inevitable in these fields, but also in the consequence of their magneto-field concentration decrease. As it is shown in Fig. 6, at higher temperatures (≈40 K) for all magnetic field values, the curves practically merge, which shows that the activation energy \( \varepsilon = 11.5 \text{ meV} \) is independent of the field, and it demonstrates the independence of the free charge carrier concentration on the magnetic field value, and thus magnetoresistance is defined by the mobility decrease of free charge carriers (at these temperature holes).

3. Conclusions

It has been ascertained that the effect of neutron irradiation on magnetic susceptibility of Si\(_{0.95}\)Ge\(_{0.05}\) whiskers is less appreciable than in Cz-Si case. After irradiation, Cz-Si contains essentially more paramagnetic components, and the concentration of the magnetically ordered clusters is higher. At the same time, the magnetic moments of created nanoclusters in both samples are comparable. It has been determined that the magnetic susceptibility value of neutron irradiated Si\(_{0.95}\)Ge\(_{0.05}\) whiskers matches that of the initial sample after the annealing at 350 °C. This suggests that radiation-induced defects are annealed out.

An analysis of the magnetoresistance curves for neutron irradiated SiGe whiskers shows that the light charge carriers (electrons or light holes) essentially contribute to the conductivity at low temperatures (approx. 4.2 K), but they are in a small amount and have high mobility. The level that supplies these charge carriers has the energy \( \varepsilon = 2.1 \text{ meV} \) and increases up to \( \varepsilon = 2.5 \text{ meV} \) when applying the magnetic field 10 T. Thus, the reason of magnetoresistance is not only the magnetic field reduction of mobility, but also the magnetic field reduction in the free charge carrier concentration. At higher temperatures (approx. 40 K), the conductivity is carried out by the holes that have the activation energy \( \varepsilon = 11.5 \text{ meV} \), and this energy is practically independent of the magnetic field.

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