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# MAGNETIC STRUCTURE OF SUBSURFACE LAYERS OF SINGLE CRYSTALLINE YTTRIUM-IRON GARNET FILMS IMPLANTED WITH Si+ IONS WITH VARIOUS ENERGIES

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Findings from the investigation of subsurface layers of epitaxial single crystalline yttrium-iron garnet $Y_3Fe_5O_{12}$ films implanted with $Si^+$ ions with the dose of $5\cdot10^{13}cm^{-2}$ and the energies of 100 – 150 keV using conversion electron Mossbauer spectroscopy are presented and the comparison with previously obtained results from simulations and x-ray diffractometry studies is carried out. The analysis of energy dependence of the components Mossbauer spectra, the integral intensity of the deformation profiles and the integral lattice disorder in the subsurface layers confirms the validity of theoretical models used in this work.

**Keywords:** yttrium-iron garnet, ion implantation, conversion electron Mossbauer spectroscopy.

Представлено результати досліджень приповерхневих шарів епітаксійних монокристалічних плівок залізо-ітрієвого гранату $Y_3Fe_5O_{12}$, імплантованих іонами $Si^+$ з дозою $5\cdot10^{13}$ см$^{-2}$ в діапазоні енергій 100 – 150 кеВ методом месбауерської спектроскопії конверсійних електронів та здійснено їх порівняння з попередньо отриманими результатами моделювання та рентгенівської дифрактометрії. Аналіз характерів енергетичних залежностей компонент месбауерівських спектрів, інтегральної інтенсивності профілю деформації та інтегрального розупорядкування структури приповерхневого шару підтвердив правомірність застосованих моделей.

**Ключові слова**: залізо-ітрієвий гранат, іонна імплантація, конверсійна електронна месбауерівська спектроскопія.

Представлены результаты исследований приповерхностных слоев эпитаксиальных монокристаллических пленок железо-итриевого граната $Y_3Fe_5O_{12}$, имплантированных ионами $Si^+$ с дозой $5\cdot10^{13}$ см$^{-2}$ в диапазоне энергий 100 – 150 кэВ методом мессбауэровскои спектроскопии конверсионных электронов и осуществлено их сравнение с ранее полученными результатами моделирования и рентгеновской дифрактометрии. Анализ характеров энергетических зависимостей компонент мессбауэровских спектров, интегральной интенсивности профиля деформации и интегрального разупорядочения структуры приповерхностного слоя подтвердил правомерность примененных моделей.

**Ключевые слова:** железо-иттриевый гранат, ионная имплантация, конверсионная электронная мессбауэровская спектроскопия.

## INTRODUCTION

Yttrium iron garnets (YIG) are very interesting ferromagnetic materials due to their unique combination of high potential for microwave application and ability to tailor properties using wide range of dopants [1]. In particular, epitaxial single crystalline yittrium-iron garnet (YIG) films are promising materials for making devices for detection, control and processing of electronic signals in the centimeter range. Operational characteristics of these devices are determined by a thin subsurface layer with artificially created gradient of physical and chemical properties.

Discovery and development of techniques to enhance properties of the subsurface layers of epitaxial iron garnet films, including YIG, is therefore important for the advancement of modern microelectronic technology. Ion implantation is a promising technique to modify properties of the subsurface layers. Its unique advantages come from the flexibility of the process itself, which allows controlling concentration of implanted ions as well as distribution of displaced matrix ions and mechanical stresses in the damaged layer, opens possibility of modifying crystalline and magnetic microstructure and overcomes limitation of traditional physical and chemical processing techniques. Several new physical effects and features with important theoretical and practical implications were observed in the implanted YIG films.

## EXPERIMENTAL

Single crystalline iron-yttrium garnet films (YIG) $Y_3Fe_5O_{12}$ were grown on 500 nm thick (111) dielectric $Gd_3Ga_5O_{12}$ substrates ($a_s$ = 12.3820 Å) using liquid phase epitaxy in a five-zone Garnet-3 oven (NVP Karat) at 10 °C, temperature at which the solution/melt is supercooled. The films thickness was 4.28 nm. The implantation was done using $Si^+$ ions accelerated to the energies of 100 – 150 keV. The implantation dose was $D = 5 \cdot 10^{13}$ cm$^{-2}$. No channeling effects or self-annealing was observed during the implantation.

Conversion electron Mossbauer spectroscopy (CEMS) was used to study magnetic microstructure of the implanted films. This technique is highly sensitivite to the changes in structural and magnetic properties of the crystalline lattice. The signal is acquired from the surface layer with depths under 1500 Å [2]. CEMS is especially effective for the investigations of implanted layers, since its acquisition depth is very similar to the thickness of the implantation layer. YAGRS-4M spectrometer was operated in the constant accelerations mode. $Fe_2O_3$ iron oxide enriched to 8% with $Fe^{57}$ isotope was used in the exit batch to improve quality of the CEMS spectra. These spectra were acquired at room temperature using $Co^{57}$ gamma rays source in chromium matrix operating in constant accelerations mode. Proportional gas counter with 96% He and 4% $CH_4$ gas mixture was used to detect conversion electrons. CEMS spectra were calibrated relative to metallic α-Fe.

The objective of this work was to investigate magnetic microstructure of subsurface layers of YIG thin films. In particular, the effects of implanting these films with $5 \cdot 10^{13}$ cm$^{-2}$ $Si^+$ ions at 100 – 150 keV were studied. The experimental results were compared with our previous simulations and x-ray diffractometry studies described elsewhere [3 – 4].

## RESULTS AND DISCUSSIONS

The dependence of the conversion electrons emission probability on the distance from the surface was calculated based on the model [5]. This model assumes that the emission probability does not depend on the angle and energy and is a function of the distance from the surface $x$. The electron energy was assumed to be inversely proportional to $x$. For the case of iron matrix, the emission probability is given by:

$$\dot{P}^{\mathrm{Fe}}(x) = 0.74 - 2.7x/R_B^k + 2.5\left[\frac{x}{R_B^k}\right]^2,$$

$$\text{then } x \ \ 0.55\, R_B^k,$$

$$P(x) = 0 \text{ then } x \leq 0.55, \qquad (1)$$

where $R_B^k$ – is the conversion electrons mean path length in iron, $R_B^k = 320$ nm. For other materials the following equation holds:

$$P(x) = P^{\mathrm{Fe}}\left(\frac{\rho}{\rho^{\mathrm{Fe}}} \cdot x\right), \qquad (2)$$

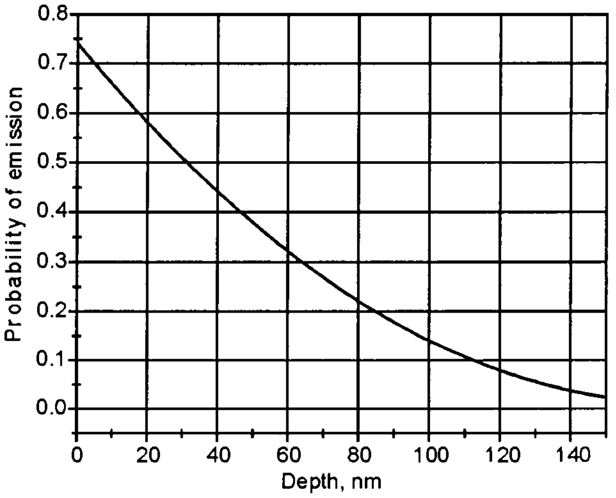

Fig. 1. Conversion electrons emission probability as a function of generation depth for surface layer of iron-yttrium garnet $Y_3Fe_5O_{12}$.

where ρ – is the target density; $\rho^{Fe}$ – is iron density. The calculation has been carried out for conversion electrons emitted during the transition of $Fe^{57}$ core from its excited to baseline energy state in $Y_3Fe_5O_{12}$ (fig. 1).

As a result, conversion electrons generated at depths over 150 nm do not contribute much to the formation of Mossbauer spectrum. In particular, 92% of Mossbauer signal comes from the 100 nm layer, while 75% of the signal comes from only 65 nm layer.

Integral lattice damage was calculated from the simulation of defect formation process (fig. 2) for 65 nm thick subsurface layer of YIG films, which is most relevant for the CEMS spectroscopy.

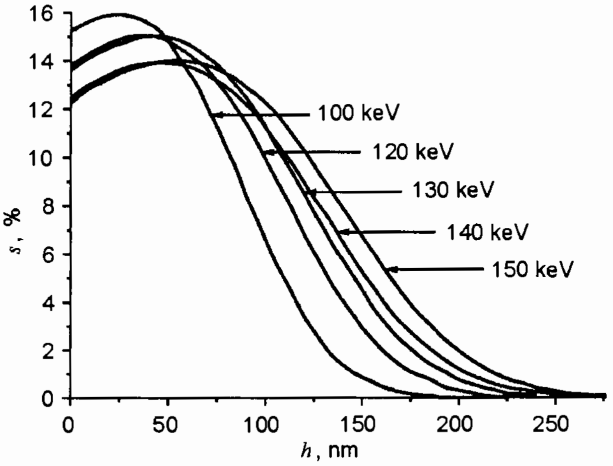

Fig. 2. Structural disorder in YIG films implanted with $Si^+$ ions with various energies.

These observations are supported by the analysis of CEMS spectra from YIG films implanted with $Si^+$ ions with various energies.

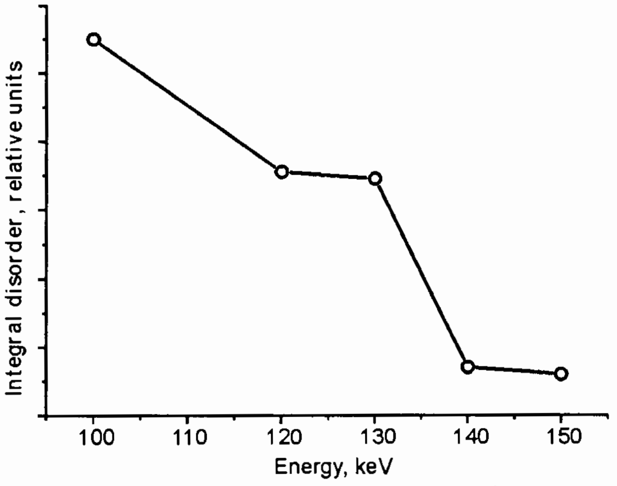

Fig. 3. Integral lattice disorder for a 65 nm thick subsurface layer of a YIG film implanted with $Si^+$ ions ($D = 5 \cdot 10^{13}$ cm$^{-2}$) as a function of implantation energy.

Table 1

Parameters of partial components of CEMS spectra from $Y_3Fe_5O_{12}$ films implanted with $Si^+$ ions with 100, 120, 130, 140 and 150 keV energies. ($D = 5 \cdot 10^{13}$ cm$^{-2}$)

| | | $I_s$, mm/s | $Q_s$, mm/s | $H$, kE | $s^*$ | $G$, mm/s |
|---|---|---|---|---|---|---|
| 100 | $a_1$ | 0.963 | 0.3034 | 491.26 | 8.7 | 0.3943 |
| | $d_2$ | 0.6191 | 0.1987 | 394.95 | 38.78 | 0.7308 |
| | $d_1$ | 0.4094 | –0.5046 | 308.28 | 22.8 | 0.9139 |
| | $a_2$ | 1..3005 | –0.7799 | 475.94 | 11.32 | 0.6553 |
| | $D$ | 0.5909 | 1.7145 | – | 18.4 | 1.0765 |
| 120 | $a_1$ | 0.8947 | 0.3034 | 484.07 | 12.71 | 0.581 |
| | $d_2$ | 0.615 | 0.1883 | 388.05 | 45.54 | 0.8079 |
| | $d_1$ | 0.4262 | –0.3524 | 304.31 | 14.28 | 0.7935 |
| | $a_2$ | 1.1906 | –0.7799 | 457.5 | 18.17 | 1.2035 |
| | $D$ | 0.6052 | 2.3954 | – | 9.31 | 1.439 |
| 130 | $a_1$ | 0.8947 | 0.3034 | 483.77 | 15.11 | 0.5595 |
| | $d_2$ | 0.617 | 0.2248 | 389.41 | 52.88 | 0.7915 |
| | $d_1$ | 0.5285 | –0.5255 | 304.47 | 11.3 | 0.6288 |
| | $a_2$ | 1.1906 | –0.7799 | 460.81 | 15.22 | 0.9461 |
| | $D$ | 0.6443 | 2.2403 | – | 5.49 | 0.8 |
| 140 | $a_1$ | 0.9324 | 0.2602 | 489.79 | 24.79 | 0.4309 |
| | $d_2$ | 0.6777 | 0.1138 | 386.67 | 35.77 | 0.5801 |
| | $d_1$ | 0.5422 | 0.4099 | 401.94 | 18.11 | 0.3848 |
| | $a_2$ | 1.1015 | –0.393 | 459.7 | 19.24 | 1.1691 |
| | $D$ | 0.5824 | 2.2912 | – | 2.1 | 0.3205 |
| 150 | $a_1$ | 0.9138 | 0.1568 | 479.5 | 21.2 | 0.5498 |
| | $d_2$ | 0.6323 | 0.1827 | 370.29 | 37.08 | 0.8818 |
| | $d_1$ | 0.6016 | 0.2971 | 394.83 | 18.73 | 0.4722 |
| | $a_2$ | 0.9475 | –0.6296 | 411.68 | 16.89 | 0.8511 |
| | $D$ | 0.6361 | 2.1097 | – | 6.1 | 0.5022 |

The spectra were assumed to be a superposition of four sextants that correspond to different magneto-equivalent positions of iron ions and the duplet component. The presence of two octa-coordinated positions of Mossbauer cores comes from two possible orientations of the main axes of the electric field tensor relative to the magnetization direction [6]. Two magneto-nonequivalent tetra-coordinated positions were first introduced by the authors [7] as a result of deviation from anion stoichiometry of garnet lattice due to lack of equilibrium and entrance of dopant atoms in the final stages of the film growth. The change in the local atomic configuration and the formation of defects during the implantation leads to broadening of lines of hyperfine magnetic splitting and (as a result of breaking exchange bonds) appearance of paramagnetic iron ions, which are recorded as the doublet components in the spectra

There is a close similarity in the energy dependence of the relative composition of the doublet component of CEMS spectra and the integral intensity of the deformation profile for the 65 nm thick subsurface layers. This observation provides evidence to support the validity of our theoretical

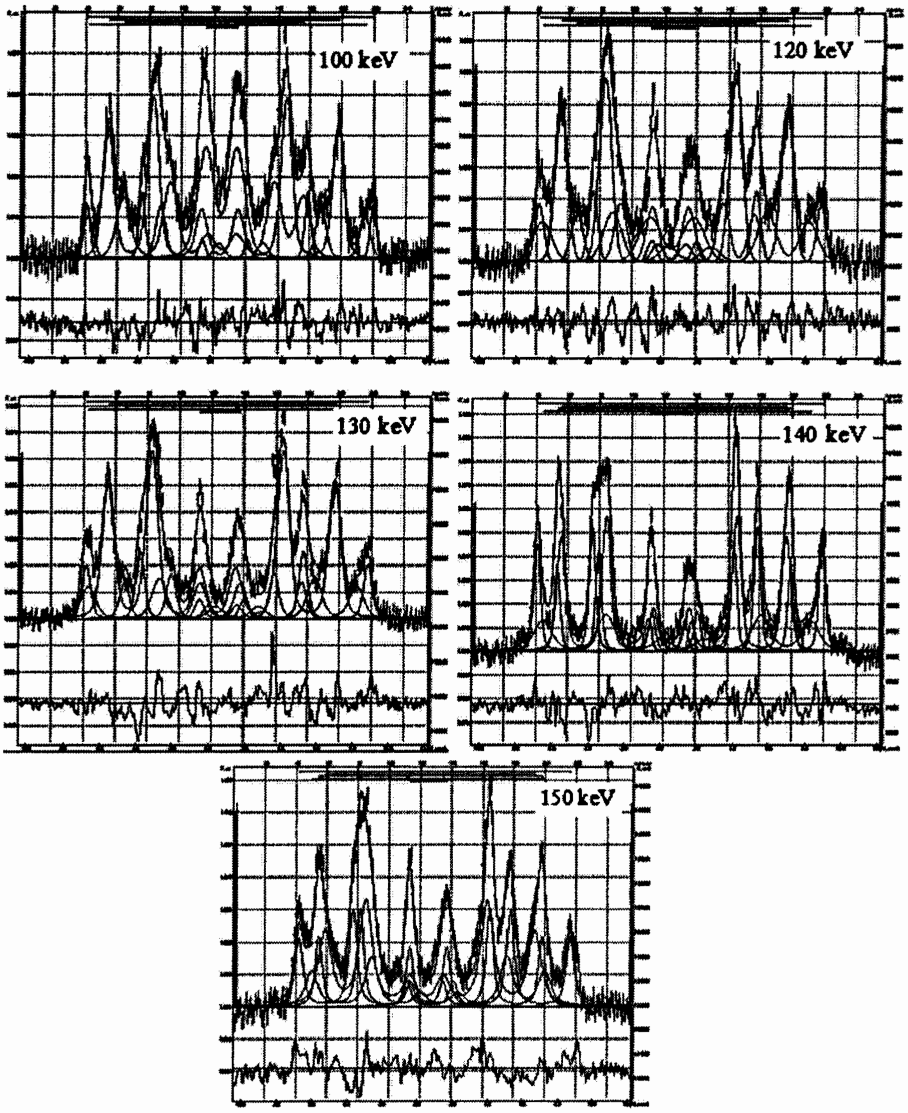

Fig. 4. CEMS spectra of subsurface layers of iron-yttrium garnet films $Y_3Fe_5O_{12}$ implanted with $Si^+$ ions with energies of 100, 120, 130, 140 and 150 keV, ($D = 5\cdot10^{13}$ cm$^{-2}$).

models and correctness of the mathematical interpretation of the x-ray diffractometry results (fig. 5(a) and (b)). Simultaneously, these findings are consistent with the variation of the amount of integral lattice disorder in the subsurface layer of YIG films with the increase in the energy of implant $Si^+$ ions (fig. 3).

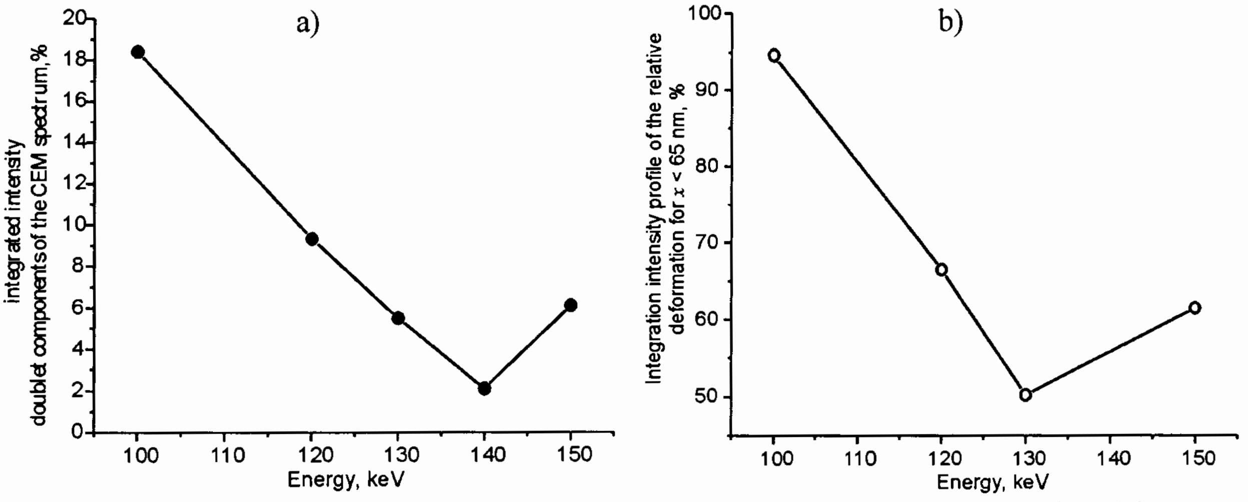

Fig. 5. The dependence of relative composition of (a) doublet component of the CEMS spectra and (b) and the integral intensity of the deformation profile in 65 nm think subsurface layer on the energy of $Si^+$ ions ($D = 5\cdot10^{13}$ cm$^{-2}$).

It is worth noting that the increase in the implant energy with the implantation dose kept constant leads to larger change of the combined integral intensity of the spectral components that correspond to octa-coordinated iron ions compared to tetra-coordinated ones. The relative concentration of a-lattice iron ions in magneto-ordered state is increasing with the increase in energy and approaching stoichiometry level (fig. 6). This is another confirmation of higher radiation stability of tetra-coordinated cation sub lattice resulting from smaller amount of oxygen anions in the first coordination sphere [8].

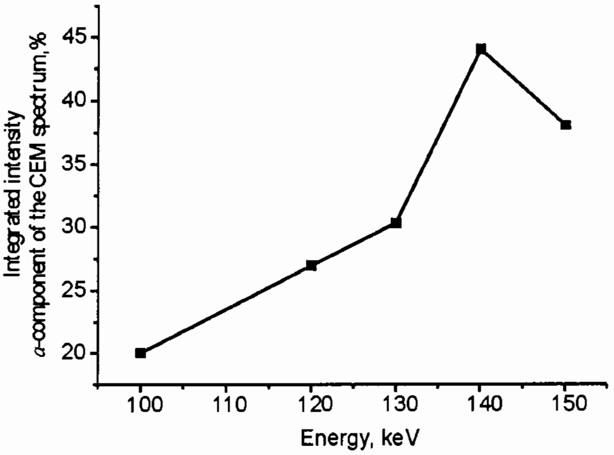

Fig. 6. The energy dependence of the integral intensity of the magnetic components of CEMS spectra generated by resonance scattering of x-rays from the cores of $Fe^{3+}$ ions in octa sub lattices for epitaxial YIG film implanted with $Si^{+}$ ions ($D = 5\cdot10^{13}$ cm$^{-2}$).

## CONCLUSIONS

1. The integral lattice disorder in the 65 nm thick sub surface layer of YIG film decreases with the increase in the implant $Si^{+}$ ions energy, which is consistent with the energy dependence of the mean free path of implant ions in the crystals.
2. The presence of two octa-coordinated positions of Mossbauer cores comes from two possible orientations of the main axes of the electric field tensor relative to the magnetization direction.
3. The presence of two magneto-inequivalent tetra-coordinated positions is caused by the deviations in anion stoichiometry driven by the lack of equilibrium and introduction of dopant atoms in the final stages of epitaxial growth process.
4. The validity of the theoretical model is supported by the similarity in the energy dependences of the relative composition of the doublet component of Mossbauer spectrum acquired from (65 nm think) subsurface layer and the integral intensity of the deformation profile obtained from *x*-ray diffractometry studies correlated to the changes in the amount of integral lattice disorder in subsurface layer.

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