

Andrzej TUROS*, Andrzej PODGÓRSKI**, Jacek JAGIELSKI**,
Marek WIELUŃSKI*

BACKSCATTERING ANALYSIS OF MAGNETIC LIQUID PHASE EPITAXIAL LAYERS ON GADOLINIUM GALLIUM GARNETS

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Abstract. Backscattering analysis of magnetic, epitaxial layers on gadolinium gallium garnets is described. The obtained results have been compared with the values estimated using segregation coefficients. The good agreement of these results testifies to the usefulness of the backscattering method in nondestructive analysis of the layers composition. The incorporation of lead (originated from fluxed melts) into crystal structure has also been found.

INTRODUCTION

In recent years the considerable development of the backscattering analysis, both in the basic and the applied research, has been observed (Chu *et al.* 1977; Turos 1974, 1978; Mayer, Ziegler 1974; Meyer *et al.* 1976). This method has been particularly useful for the structural and analytical studies.

In order to investigate the properties of the magnetic layers of GGG materials it has been necessary to determine precisely an elemental composition of these layers.

The determination of the composition by classical methods has not been accurate enough (O'Dell 1974).

The purpose of the present paper is the analysis of usability of the backscattering method in the accurate determination of the composition of GGG layers.

METHOD DESCRIPTION

The backscattering method is based on utilization of the phenomena occurring during interaction of charged species, like protons, deuterons or helium ions with the surface of solids. The charged particles accelerated in the energy range of 0.5 –

* Institute of Nuclear Research, ul. Hoża 69, 00-681 Warszawa.

** Institute of Electronic Materials Technology, ul. Konstruktorska 6, 02-673 Warszawa.

2 MeV by means of an electrostatic accelerator, are then elastically scattered by the target atoms. After scattering the particles are analysed by the semiconductor detector. The detector allows to record the number and the energy of scattered particles. The energy of scattered particles depends on the mass of scattering atoms and the depth at which scattering occurred. The analysis of the backscattering energy spectrum obtained in that manner enables to determine the target composition. The investigations of the crystal materials in which one of the crystallographic axis has been aligned with beam directions (channeling), allows to obtain additional informations on the extent of crystal damage.

In the experiments carried out, directions of the crystallographic axis in the target were not aligned with the direction of the incident beam. It allowed us to consider investigated crystal as an amorphous material. The geometry of the experiment and different particle trajectories depending on the scattering depth are shown in Fig. 1.

The energy of the incident particles is E_0 and the angle between the target normal and the direction of the incident beam is θ_1 . Let us consider the particles scatter-

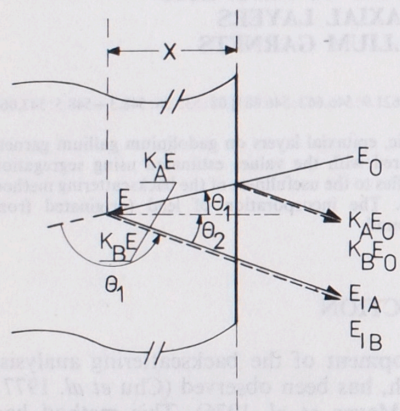


Fig. 1. Backscattering process in a sample composed of homogenous mixture of two monoisotopic elements A and B

ed by surface atoms and by the atoms located at the depth x of the material. In the first case the energy of scattered particle depends only on the mass of the scattering atom A and is equal $k_A E_0$. k_A is the scattering parameter depending in this case only on the mass of the scattering atom. In the second case the energy of the scattered particles is additionally diminished because of the energy loss in the target material and after emerging from the target is equal $E_{1,A}$. The difference $k_A E_0 - E_{1,A}$ is proportional to the depth at which scattering occurred. For the target containing n elements (compound sample), energies of the particles scattered by surface atoms $A_1, A_2 \dots A_n$ are equal to $K_{A_1} E_0, K_{A_2} E_0 \dots K_{A_n} E_0$, respectively. The energies of particles scattered by the atoms located at the depth x are equal to $E_{1,A_1} \dots E_{1,A_n}$.

The energy spectrum of particles scattered by a compound target is therefore composed by the superposition of component spectra (each of them being due to the scattering by different atoms). The formation of such a spectrum in the case of two component target ($A_x B_y$) is schematically shown in Fig. 2.

In the present work we have investigated the LPE layers of the garnets the composition of which can generally be denoted by formula: $(Y_x B_{3-x})(Fe_y C_{5-y})O_{12}$, where B is one of the elements, like Sm, Eu, Ca, C-Ge or Ga. Such compounds

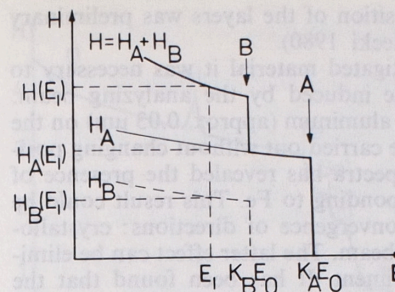


Fig. 2. The energy spectrum of the particles scattered by the target composed of two elements A and B

can be therefore denoted as $X_8 O_{12}$. In the work (Chu *et al.* 1977) the interpretation method of the spectrum for that kind of compounds was presented. The concentration ratio of two components can be determined as follows:

$$N_{A_1}/N_{A_2} = \frac{H_{A_1}}{H_{A_2}} \frac{\sigma_{A_2}}{\sigma_{A_1}} \quad [1]$$

where:

N_{A_i} - concentration of a given component in the target,
 H_{A_i} - number of counts corresponding to the component i ,
 σ_{A_i} - scattering cross-section for i component of the target.
 For $Z \geq 20$ eq. 1 can be approximated by the expression:

$$N_{A_1}/N_{A_2} = \frac{H_{A_1}}{H_{A_2}} \frac{Z_{A_2}^2}{Z_{A_1}^2} \quad [2]$$

where:

Z_{A_i} - atomic number of the component i .

Assuming additionally that:

$$(N_{A_1}/N) + (N_{A_2}/N) + \dots + (N_{A_n}/N) = 8 \quad [3]$$

where:

N - concentration of "molecules" $X_8 O_{12}$ in target, one obtains:

$$\frac{N_{A_1}}{N} \left(1 + \frac{N_{A_2}}{N_{A_1}} + \frac{N_{A_3}}{N_{A_1}} + \dots + \frac{N_{A_n}}{N_{A_1}} \right) = 8 \quad [4]$$

The content of A_i component in "molecule" $X_8 O_{12}$ has been determined by the ratio N_{A_i}/N . Using equations (2) and (4) the content of any component in epitaxial layer of the garnet can be determined.

EXPERIMENTAL

The experiments were carried out using beam of 2 MeV He^+ ions obtained from the Van de Graaff accelerator of the Institute of Nuclear Research. The proper experiments were preceded by calibration of the measuring system using standard sample of a known composition. On the basis of this procedure the energy calibration of the spectra has been made. This, in turn, made it possible to establish the energy of ions scattered by given elements and therefore to determine the masses of particular component of the target, i.e. to perform the quantitative

analysis of the layer composition. The composition of the layers was preliminary estimated using segregation coefficients (Sarnecki 1980).

Because of dielectrical properties of investigated material it was necessary to remove from the sample the electrical charge induced by the analyzing beam. This was achieved by evaporating thin layer of aluminum (approx. 0.03 μm) on the top of the samples. The early experiments were carried out without changing position of the samples. The analysis of energy spectra has revealed the presence of a pronounced peak in the energy region corresponding to Fe. This result could be explained by iron segregation or accidental convergence of directions: crystallographic axis of the iron sublattice and incident beam. The latter effect can be eliminated by rotation of the sample during experiment. It has been found that the mentioned Fe peak is no longer present in the spectra obtained for rotated samples. Analysis of the results was done using exclusively the spectra of the samples coated with Al layer and rotated during measurements.

RESULTS AND DISCUSSION

The energy spectra of the He^+ ions, obtained for the samples of different elemental composition, are shown in figures 3–5. The spectra analysis revealed the presence of Al surface layer of the thickness of about several hundred \AA . The presence of lead of uniform concentration has also been found. Gadolinium-gallium garnets are commonly grown by liquid phase epitaxy from $\text{PbO}-\text{B}_2\text{O}_3$ fluxed melts. Consequently some incorporation of Pb may be observed. Because of its

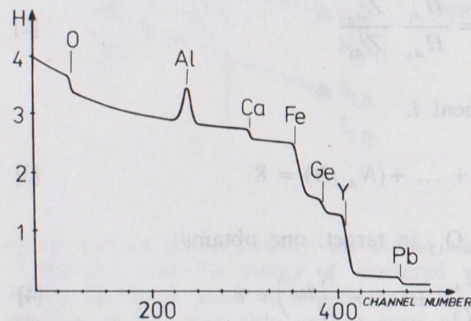


Fig. 3. The energy spectrum for the sample no 1. H is number of counts per channel/1000

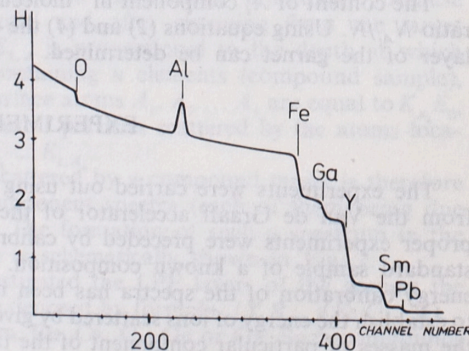


Fig. 4. The energy spectrum for the sample no 2. H is number of counts per channel/1000

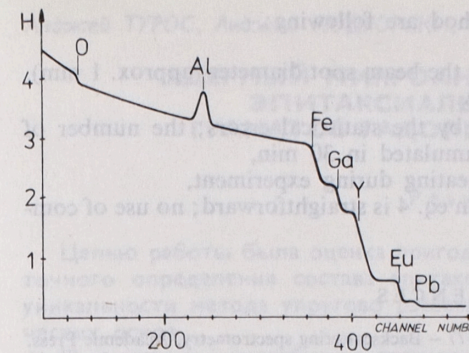


Fig. 5. The energy spectrum for the sample no 3. H is number of counts per channel/1000

large ionic radius, Pb^{2+} is most likely to be accommodated in GGG in the largest cation site, the dodecahedral site, which normally is occupied by Y^{3+} (Glass and Elliott 1974). Thus, instead of a single crystal generally denoted by $(\text{Y}_x\text{B}_{3-x})(\text{Fe}_y\text{C}_{5-y})\text{O}_{12}$ where B – rare-earth element or Ca, C – Ga or Ge, the single crystal denoted by $(\text{Y}_{x-a}\text{Pb}_a\text{B}_{3-x})(\text{Fe}_y\text{C}_{5-y})\text{O}_{12}$ is formed.

The compositions of layers were calculated using the previously described method, the results being given in Table 1. The composition of layers estimated using segregation coefficients (Sarnecki 1980) are also presented in the Table. Because of the strong temperature effect on segregation coefficient the estimation of the layer composition using that coefficient is a very inaccurate method. Also the accurate control of epitaxy process parameters is impossible. It is incapable of detecting fluxed melts components (such, as Pb) in the layer and determining their content. Backscattering method enables to determine composition of the chosen layers independently of the preparation manner.

The method is a nondestructive one and allows to determine distribution of the impurities. No special standards are required except the universal one, for the energy calibration. The basic limitation of the method seems to be the difficulty in the content determination of elements of atomic mass considerably smaller than the mean atomic mass of the layer. However, it is possible in some cases to determine the content of light elements using nuclear reaction method, which is a modification of the backscattering method.

Table 1
The composition of layers

Sample	Layer composition estimated using segregation coefficients	Layer composition calculated using backscattering method
1	$(\text{Y}_{2,05}\text{Ca}_{0,95})(\text{Fe}_{4,1}\text{Ge}_{0,9})\text{O}_{12}$	$(\text{Y}_{1,92}\text{Pb}_{0,03}\text{Ca}_{1,04})$ $(\text{Fe}_{4,22}\text{Ge}_{0,81})\text{O}_{12}$
2	$(\text{Y}_{2,55}\text{Sm}_{0,45})(\text{Fe}_{3,8}\text{Ga}_{1,2})\text{O}_{12}$	$(\text{Y}_{2,62}\text{Pb}_{0,02}\text{Sm}_{0,36})$ $(\text{Fe}_{3,72}\text{Ga}_{1,29})\text{O}_{12}$
3	$(\text{Y}_{2,4}\text{Eu}_{0,6})(\text{Fe}_{3,9}\text{Ga}_{1,1})\text{O}_{12}$	$(\text{Y}_{2,57}\text{Pb}_{0,04}\text{Eu}_{0,34})$ $(\text{Fe}_{3,86}\text{Ga}_{1,19})\text{O}_{12}$

The main characteristics of the method are following:

- method is undestructive,
- sample sizes are limited only by the beam spot diameter (approx. 1 mm),
- typical depth resolution is 200 Å.
- the measurement time is limited by the statistical errors; the number of counts giving 1% accuracy can be accumulated in 30 min,
- there is no measurable sample heating during experiment,
- the analysis of the spectra based on eq. 4 is straightforward; no use of computers is necessary.

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*Andrzej TUROS, Andrzej PODGÓRSKI, Jacek JAGIELSKI,
Marek WIELUŃSKI*

JĄDROWA MIKROANALIZA MAGNETYCZNYCH WARSTW EPITAKSJALNYCH GRANATÓW GADOLINOWO GALOWYCH

Streszczenie

Praca miała na celu ocenę przydatności metody rozpraszania wstecznego do precyzyjnego wyznaczania składu epitaksjalnych warstw granatów.

Ze względu na unikalność metody opisano pokrótce jej fizyczne podstawy. Przedstawiono sposób interpretacji widma dla związków typu granatów. Warunki eksperymentalne dostosowano do własności badanego materiału. Wyznaczone składy warstw porównano z szacunkowymi wartościami uzyskanymi na podstawie współczynników segregacji. Stwierdzono zgodność wyników. W warstwie zaobserwowano obecność ołowiu o równomiernym rozkładzie koncentracji. Występowanie ołowiu wynika z użycia $PbO-B_2O_3$ jako topnika w procesie epitaksji.

Uzyskane wyniki świadczą o przewadze metody rozpraszania wstecznego nad klasycznymi metodami wyznaczania składu warstw epitaksjalnych.

OBJAŚNIENIA FIGUR

- Fig. 1. Schemat procesu rozpraszania cząstki przez tarczę zawierającą homogeniczną mieszaninę dwóch monoizotopowych pierwiastków A i B
- Fig. 2. Widmo energetyczne cząstek rozproszonych przez tarczę zawierającą dwa pierwiastki A i B
- Fig. 3. Widmo energetyczne dla próbki 1
- Fig. 4. Widmo energetyczne dla próbki 2
- Fig. 5. Widmo energetyczne dla próbki 3

Андрей ТУРОС, Андрей ПОДГОРСКИ, Яцек ЯГЕЛЬСКИ, Марек ВЕЛУНЬСКИ

ЯДЕРНЫЙ МИКРОАНАЛИЗ МАГНИТНЫХ ЭПИТАКСИАЛЬНЫХ ПЛЁНОК ГРАНАТОВ ГАДОЛИНОВО-ГАЛЬОВЫХ

Резюме

Целью работы была оценка пригодности метода упруго-рассеяния для точного определения состава эпитаксиальных гранатовых плёнок. В виду уникальности метода упруго-рассеяния дано краткое описание его физических основ.

Представлено способ интерпретации спектра для соединения типа граната. Параметры эксперимента были обусловлены свойствами испытуемого материала. Определённые составы плёнок сопоставлено с приблизительными данными полученными из коэффициентов сегрегации. Результаты показали сходными. В плёнках обнаружено свинец с однородным распределением концентрации. Присутствие свинца является следствием применения $PbO-B_2O_3$ в качестве флюса в процессе эпитаксиального наращивания из жидкой фазы.

ОБЪЯСНЕНИЯ К ФИГУРАМ

- Fig. 1. Схема процесса рассеяния частиц на мишени содержащей два моноизотопные элементы A и B
- Fig. 2. Зависимость числа зарегистрированных частиц от энергии для мишени содержащей два моноизотопные элементы A и B
- Fig. 3. Энергетический спектр рассеяния для плёнки no 1.
- Fig. 4. Энергетический спектр рассеяния для плёнки no 2.
- Fig. 5. Энергетический спектр рассеяния для плёнки no 3.