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PARAMAGNETIC GARNET FILM MONOCHROMATOR FOR SYNCHROTRON RADIATION

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Abstract. – Epitaxial films of paramagnetic garnets are shown to present, in principle, noticeable advantages over ferrimagnetic ones when used as Mössbauer monochromators of synchrotron radiation. The growth of such paramagnetic garnet films of composition \( \{Y_{2.9}La_{0.1}\} \{Sc_{1.8}Fe_{0.8}\} \{Fe_9\} O_{12} \) is reported.

A Mössbauer beam from synchrotron radiation has been recently obtained for the first time at Doris [1, 2], using a pure nuclear reflection from an epitaxially grown film of ferrimagnetic Yttrium Iron Garnet (YIG) \( Y_3Fe_5O_{12} \), highly enriched in \( ^{57}Fe \). A significant improvement towards the realization of a practical Mössbauer source from synchrotron radiation would be the utilization of a paramagnetic garnet film as monochromator, which should give a nuclear diffraction energy spectrum with fewer and broader peaks, due to the simpler hyperfine splitting and due to stronger oscillator strength.

Indeed, in the garnet structure, pure nuclear reflections can be obtained also from paramagnetic crystals. The general formula unit of garnets is \( \{D_3\} \{A_2\} \{C_3\} O_{12} \), where the cations C, A, and D occupy distorted dodecahedral, octahedral and tetrahedral sites, respectively. The quadrupole interaction varies from site to site. It also varies periodically within the unit cell for the cations of a single site because of changes in the orientation of the symmetry axis of the electric field gradient. This causes pure nuclear reflections from subgroups of the paramagnetic cations for each site (C, A, or D).

We will focus on pure nuclear reflections from \( ^{57}Fe \) ions. In the garnet structure iron ions enter essentially octahedral and tetrahedral sites. Because of intensity considerations [1] we discuss briefly only the paramagnetic pure nuclear diffraction from the tetrahedral site. The unit cell contains 24 ions with tetrahedral surrounding, divided into three equally populated subgroups each one having its local symmetry axis (a fourfold inversion axis) along one of the three (100) crystallographic directions. All ions of the tetrahedral site have the same cross section for electronic scattering of X-ray radiation. On the other hand the cross section for resonant nuclear scattering for linear polarized X-rays depends on the angle between the axis of the local electric field gradient (i.e. the local symmetry axis) and the polarization direction [3]. Therefore using a X-ray beam of suitable energy (for instance from synchrotron radiation) it is possible to make the three subgroups of the tetrahedral site inequivalent, but only as far as the resonant nuclear scattering is concerned. This leads to the occurrence of a number of pure nuclear reflections. For instance this is the case for the (002) electronic forbidden reflection when the incident X-ray beam is polarized along a [100] direction.

From the point of view of the material design, the main requirements are:

i) to bring the Neel temperature of YIG below room temperature, by substituting with diamagnetic ions only octahedral irons, in order to keep the maximum intensity for nuclear reflections from tetrahedral sites;

ii) to use a growth method requiring a minimal amount of iron oxide, due to the very high cost of enriched \( ^{57}Fe \).

Requirement i) can be fulfilled [4] by substitution of Sc\(^{3+}\) (which has a very strong preference for octahedral garnet sites), in amounts larger than 1 atom per formula unit (AFU). Scandium has also the important advantage of giving rather low absorption for the intended 14.4 keV X-rays. The resulting lattice parameter would be \( a_0 = 1.2457 \) nm, too large to be compatible with the lattice parameter of conventional Gadolinium Gallium Garnet (GGG) substrates \( a_{GGG} = 1.2381 \) nm. We therefore decided to adopt as substrate the "SGGG" , of nominal composition \( \{CaGd\}_3 \{MgZrGa\}_2 \{Ga\}_5 O_{12} \), originally developed by Mateika and Rusche [7], that has a lattice constant \( a_{SGGG} = 1.2497 \) nm. This value just corresponds to a scandium substitution of 1.5 AFU in YIG.

Requirement ii) suggested us to use, for liquid phase film growth, the Lithium Rare-earth Molybdate melt, developed by Bonner [6]. It has a solubility for \( Fe_2O_3 \) much lower than the conventional PbO – B\(_2\)O\(_3\) melt, so that only about 2.5 g of enriched iron are required. We have performed a series of preliminary experiments.
(using, of course, non-enriched iron) concerning high scandium substitutions. The amount of scandium substituted in YIG was found to be an increasing function of the growth temperature, and to be also favoured by tiny simultaneous substitution of $\text{La}^{3+}$ for $\text{Y}^{3+}$ in dodecahedral garnet sites. Films of composition $\text{Y}_{2.5}\text{La}_{0.1}\text{Sc}_{1.5}\text{Fe}_{0.5}\text{(Fe)}\text{O}_{12}$ were grown on (111) and on (100) SGGG substrates. CEMS spectra (conversion electron Mössbauer spectroscopy) of the epitaxial films are shown in figure 1, which clearly confirm their paramagnetic nature.

Figure 2 shows a computer simulation (based on the dynamical theory) of the time response of a film of the same composition, enriched in $^{57}\text{Fe}$, following a (002) nuclear Bragg reflection (the procedure is outlined in Ref. [7]). It is seen that most of the reflected intensity is located in the first dominant peak. Although the integral intensity is less than that of the pure YIG response, the peak reflectivity is increased and could be even more than two times higher if the octahedral sites are fully substituted by scandium.

In conclusion, among the crystals which show pure nuclear reflections, the paramagnetic garnet crystals appear to be well suited for a nuclear monochromator due to their simple time response and high reflectivity. The growth of films adequate to this purpose (highly enriched in $^{57}\text{Fe}$) has been demonstrated to be attainable, and is presently being pursued.

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