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DIRECT IRRADIATION OF $LaMn_2Si_2$ AND $LaMn_2Ge_2$ COMPOUNDS WITH THERMAL NEUTRONS TO PRODUCE $^{140}La(^{140}Ce)$ PROBE NUCLEI FOR PAC SPECTROSCOPY: ANALYSIS OF POSSIBLE INTERFERENCES FROM ^{56}Mn AND $^{75,77}Ge$ GAMMA RAYS

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ABSTRACT

In this paper we describe a method to introduce radioactive ^{140}La nuclei with a half-life of ~40 h, into samples of intermetallic compounds $LaMn_2X_2$ (X = Si, Ge) to carry out Perturbed gamma-gamma angular correlation (PAC) spectroscopy measurements using $^{140}La(^{140}Ce)$ as probe nuclei. While there are several methods to introduce this probe nucleus in a variety of samples, an alternate method is described which produces $^{140}La(^{140}Ce)$ probe in $LaMn_2Ge_2$ and $LaMn_2Si_2$ compounds by directly irradiating for a short time with thermal neutrons in the IEA-R1 nuclear reactor at IPEN/CNEN-SP. This method could be used because La is a component of the samples. Natural La contains (99.9%) as ^{139}La isotope which, when irradiated with neutrons produces ^{140}La, the parent radioisotope of ^{140}Ce used for PAC measurements. However, other elements present in the compounds are also activated during neutron irradiation to produce gamma emitting radioisotopes in particular ^{56}Mn and $^{75,77}Ge$. In order to verify if these would interfere in the PAC measurements, we have measured the gamma spectrum of these isotopes in the samples at different time intervals after the end of irradiation using a high resolution HPGe spectrometer. PAC spectra at room temperature were measured for both $LaMn_2Si_2$ and $LaMn_2Ge_2$ compounds using $^{140}La(^{140}Ce)$ as probe nuclei and showed that interference from other gamma-rays is negligible if PAC measurements started 24h after the end of irradiation of the sample.

1. INTRODUCTION

Hyperfine interaction measurements using perturbed gamma-gamma angular correlation (PAC) spectroscopy is based on the emission of two gamma radiations in a cascade arising from the decay of the excited states of a probe nucleus. It provides information about the hyperfine interaction between external nuclear fields and nuclear moments of probe nuclei in a certain atomic site in the crystalline structure of solid materials. As a consequence it allows extracting information of the involved hyperfine parameters, as well as the characterization of structural and magnetic transitions of the crystal. The most commonly used probe nuclei for PAC measurements are $^{111}In(^{111}Cd)$, $^{140}La(^{140}Ce)$ e $^{181}Hf(^{181}Ta)$. In this paper, we present a method to introduce $^{140}La(^{140}Ce)$ probe nuclei into samples of $LaMn_2Si_2$ and $LaMn_2Ge_2$ compounds to carry out PAC measurements. There are several ways to introduce this radioisotope into the samples. One way is through the ion implantation technique of the ^{140}La which β^- decays to produce ^{140}Ce. Another way is by means of radioactive-isotope-beam technique, where, ^{140}Cs is implanted into the sample where the radioactive equilibrium (^{140}Ba

– ^{140}La) is achieved followed by the β^- decay to produce ^{140}Ce [1]. Still another method would be through irradiation of a small quantity of metallic La with neutrons to produce ^{140}La which is then introduced into the sample (replacing approximately 0.1% of the desired atomic sites) through arc melting of the component metals followed by thermal annealing [2]. The present paper describes a new methodology to obtain the $^{140}La(^{140}Ce)$ in the $LaMn_2Si_2$ and $LaMn_2Ge_2$ compounds. These compounds have the natural La that contains ^{139}La isotope, (with 99.9% natural abundance) which when irradiated with neutrons produces the ^{140}La radioisotope. The specific facility, known as fast pneumatic station irradiation was used to irradiate samples with neutrons in the IEA-R1 research reactor of IPEN/CNEN. One of the advantages of this method is the certainty that the probe nuclei is in the position of the La atom. However other component elements of the compounds such as Mn and Ge and Si will also be activated by neutron irradiation and it should be made sure that gamma rays from these radioisotopes do not interfere in the measurement of PAC spectra using $^{140}La(^{140}Ce)$ probe. High resolution gamma ray spectroscopy using HPGe detector can be used to identify the presence of all the gamma ray emitting nuclei in the sample. The only other gamma emitting radionuclides produced after the neutron irradiation of the samples $LaMn_2Si_2$ and $LaMn_2Ge_2$ are ^{56}Mn ($t_{1/2}$ =2.6h), ^{75}Ge ($t_{1/2}$ =83min) and ^{77}Ge ($t_{1/2}$ =11.3h) apart from ^{140}La. The other radionuclides such as ^{71}Ge ($t_{1/2}$ =11.4d) and ^{31}Si ($t_{1/2}$ =2.6h) also produced from neutron activation are pure beta emitters [3].

2. EXPERIMENTAL

Samples of $LaMn_2Ge_2$ and $LaMn_2Si_2$ (La = 99.9 %, Mn, Si, Ge = 99.999 % purity) were prepared by arc-melting the metallic the components in stoichiometric proportion in argon atmosphere. A small excess of Mn was used for the case of $LaMn_2Ge_2$ as a small part of Mn was found to evaporate during arc melting. After repeated melting of the samples in the arc furnace they were sealed in vacuum (2.10^{-4} mbar) and submitted to thermal annealing at 850 ^{0}C for 12 hours. The samples were characterized by X–ray diffraction (XRD) and the results showed the expected tetragonal structure of $ThCr_2Si_2$ type, which belongs to the I_4/mmm space group (see Figure 1). The XRD measurements were performed at LCT-laboratory at Polytechnic Institute of University of São Paulo (USP). The data were analyzed by the Rietveld method with software Rietica [4]. The results are presented in Fig. 1.

The samples of $LaMn_2Ge_2$ and $LaMn_2Si_2$ were irradiated in the pneumatic station for 4 minutes with thermal neutrons in a flux of about 6 x 10^{12}n cm^{-2}s^{-1} in the pneumatic station of the IEA-R1 Nuclear Research Reactor at IPEN-CNEN/SP. The time of irradiation in this station is limited to a maximum of 5 minutes. The station is connected to the nuclear reactor core through a tubular pneumatic system. Samples to be irradiated are sent through this tube and return automatically after pre-determined irradiation time. Samples to be irradiated are enclosed in a polyethylene container called rabbits. After short irradiation, the gamma ray spectra were recorded using HPGe spectrometer.

PAC measurements were made at room temperature using the spectrometer with six BaF_2 detectors positioned on the face of a cube (Figure 2) and associated with a "slow-fast" electronic arrangement for measuring delayed gamma-gamma coincidence spectra. The spectrometer allows simultaneous accumulation 30 coincidence spectra [5]. The experimental data were acquired using Lab-View software then analyzed by the software PAC-Fit.

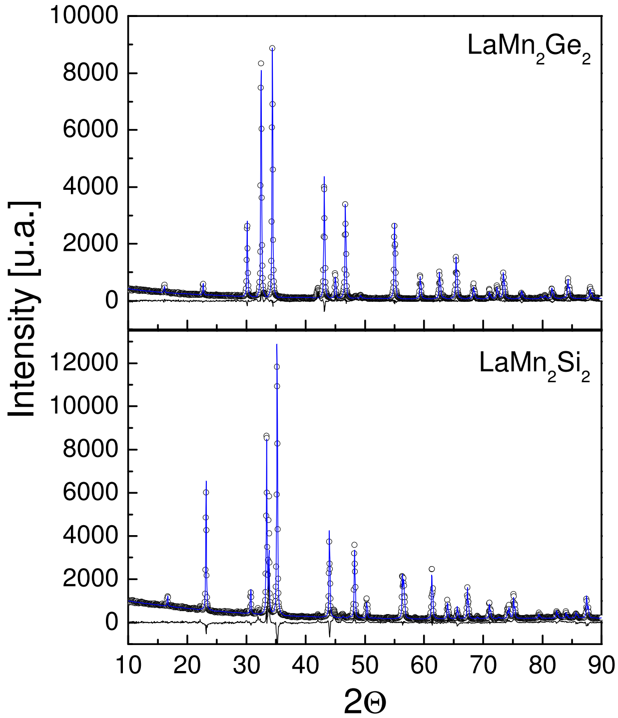

Figure 1. X-ray diffraction spectra of $LaMn_2Si_2$ and $LaMn_2Ge_2$ compounds. Both compounds present only one phase corresponding to tetragonal $ThCr_2Si_2$ type structure.

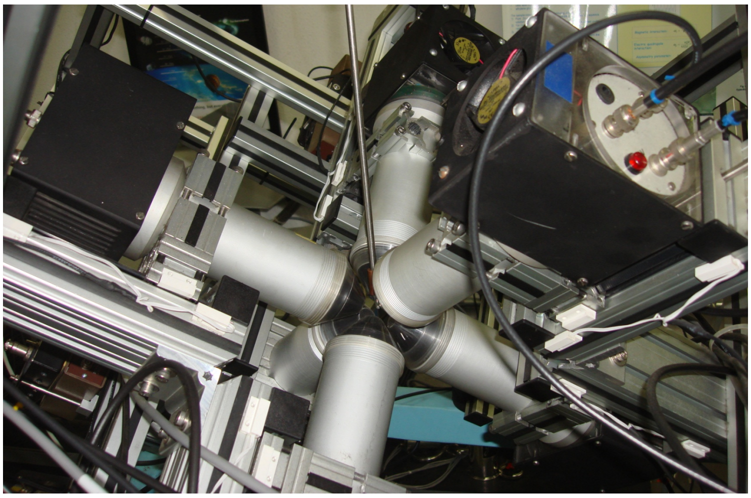

Figure 2. Cubic arrangement of the six BaF_2 detectors.

3. RESULTS AND DISCUSSION

3.1. Neutron Irradiation of the Sample and Gamma Spectroscopy

After neutron irradiation the gamma ray measurements were performed using the HP(Ge) detector (Model GC 3020) coupled to digital spectrum analyzer (DSA-1000) multichannel analyzer, both from Canberra. Gamma ray spectra were collected and processed using a Canberra Genie 2000 version 3.1 of gamma spectroscopy software. Samples were measured at 1, 24 and 72 hours after the irradiation with counting time of 600 seconds (see the spectra in Fig. 3).

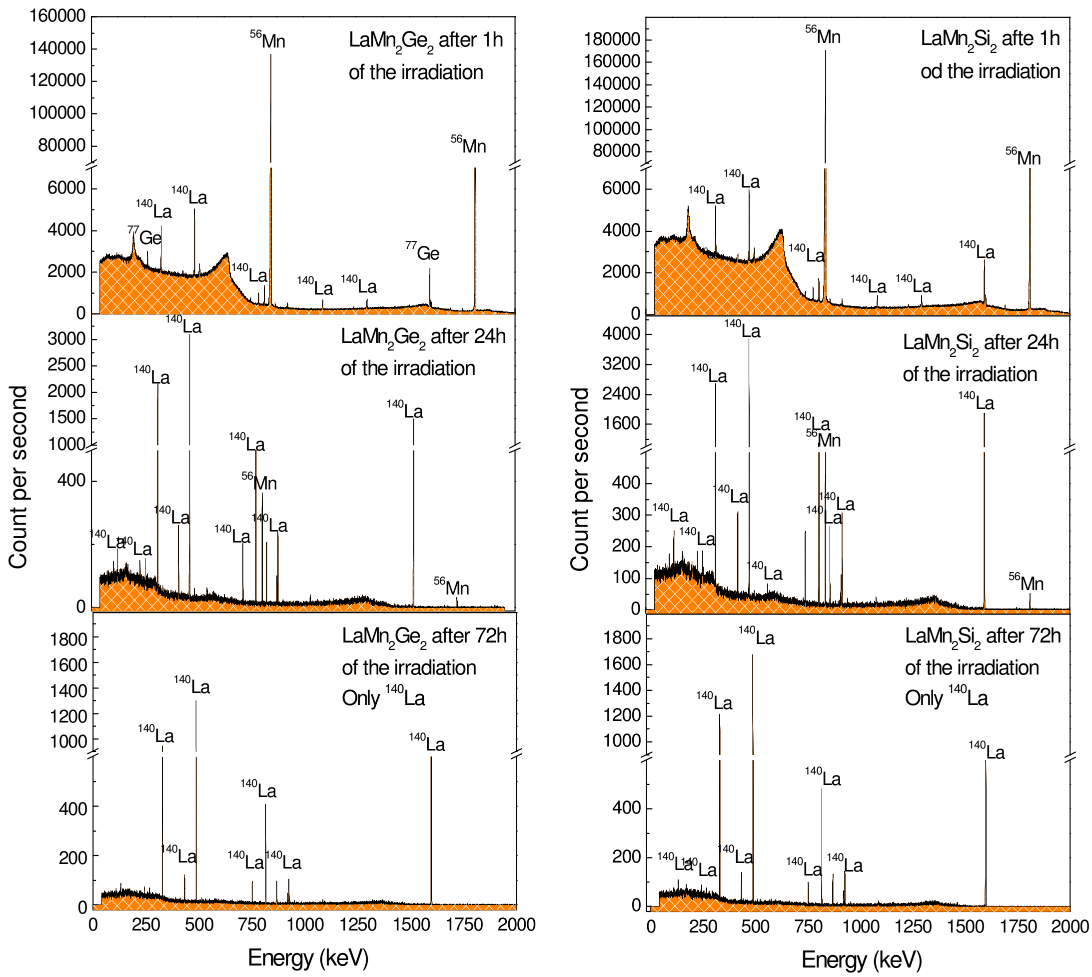

Figure 3. Gamma ray spectra measured with a HP(Ge) detector. $LaMn_2Si_2$ and $LaMn_2Ge_2$ compounds irradiated in IEA-R1 and measurements performed after 1, 24 and 72 hours of irradiation.

In the first measurement, 1h after the end of irradiation, the gamma ray energies of 328.8 keV and 487.0 keV, in the decay o^{140}La, used in the PAC spectroscopy are clearly observed. However, gamma-rays of ^{56}Mn (847.3 keV and 1812.9 keV) and ^{77}Ge (264.7 keV and 1603.8 keV) are observed with much larger intensities. The spectra obtained 24 hours after the irradiation show ^{56}Mn gamma rays with reduced intensity in addition to ^{140}La gamma rays. Hardly any gamma rays of ^{77}Ge are visible. The spectra obtained 72 hours after the irradiation show only ^{140}La gamma rays as can be seen in Fig. 3. Thus it was concluded that PAC measurements of $LaMn_2Si_2$ and $LaMn_2Ge_2$ could be started after 72 with no interference from other radioactivities in the sample.

3.2. PAC Measurements

PAC measurements were performed after 24 hours of sample irradiation. During this period the sample was annealed at 850^0C. As shown in the gamma ray spectra after 24 hours the ^{56}Mn gamma rays practically do not interfere in the PAC measurements. PAC measurements were carried out only at room temperature (Figure 4).

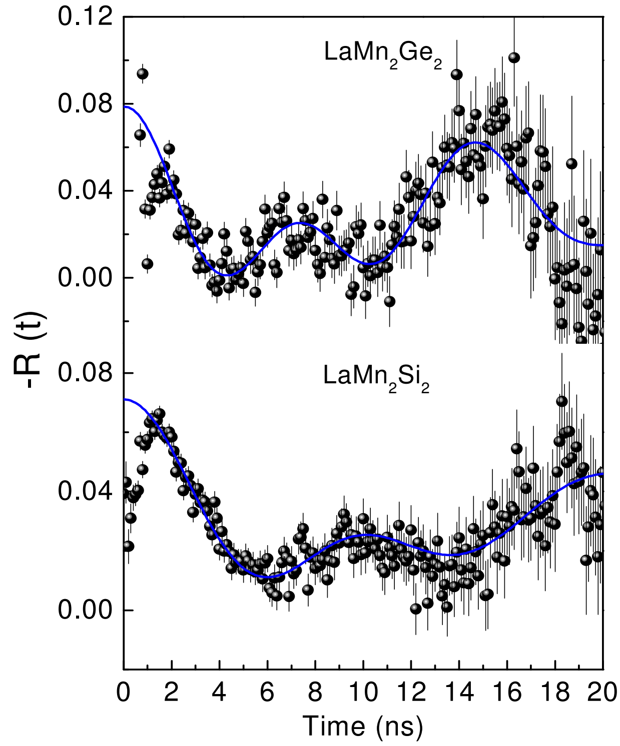

Figure 4. PAC spectra for $^{140}La(^{140}Ce)$ probe nuclei in $LaMn_2Ge_2$ and $LaMn_2Si_2$ compounds at room temperature. The blue solid lines are the least square fit to the theoretical function.

The results of the PAC measurements show only magnetic interaction because the ^{140}Ce probe nucleus has a small electric quadrupole moment. This fact makes this probe practically insensitive to electric quadrupole interactions, being sensitive only to magnetic dipole interactions. The analyses showed only single site with magnetic interaction for both samples and the hyperfine magnetic frequency were 48.1MHz and 67.3 MHz for $LaMn_2Si_2$ and $LaMn_2Ge_2$, respectively.

3. CONCLUSIONS

The results of gamma ray measurements showed that the technique used in this work for the production $^{140}La(^{140}Ce)$ probe nuclei in $LaMn_2Si_2$ and $LaMn_2Ge$ compounds is more effective than other techniques used currently and cited in this paper. The tests showed that after 24 hours the energies of ^{77}Ge gamma rays are not present in the spectrum and the 847keV gamma ray in the decay of ^{56}Mn was the only one present other than the gamma rays in the decay of ^{140}La. The ^{56}Mn gamma ray at 847keV does not interfere in the PAC measure as the energies of ^{140}La gamma rays used for the PAC measurement are 328 keV and 487 keV and these can be selected without interference from other gamma rays. The results showed that other component elements of the compounds $LaMn_2Si_2$ and $LaMn_2Ge_2$ which get activated by neutrons in addition to ^{140}La do not interfere in the PAC measurements as long as the measured are performed at least 24h after the end of irradiation. Additional advantage of the method is the possibility of producing the probe nuclei again in the same sample after its activity has reduced just by repeating the irradiation with neutrons in the pneumatic station facility thus eliminating the need to produce new samples each time. Moreover this technique can be use for others samples that have ^{139}La since the others radioisotopes in the sample not interfere in the measure.

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