Electric field gradient in nanostructured SnO$_2$ studied by means of PAC spectroscopy using $^{111}$Cd or $^{181}$Ta as probe nuclei

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Abstract Electric quadrupole interactions were studied in pure and Mn-doped powder samples and thin films of SnO$_2$ using perturbed $\gamma\gamma$ angular correlation spectroscopy (PAC). The powder samples were prepared by Sol gel method and the thin film were prepared on the Si (100) substrate by sputtering technique using Sn in the oxygen atmosphere. The samples were characterized by x-ray diffraction, energy dispersive spectroscopy and scanning electron microscopy. The thickness of the film was 100 nm. The average particle size of the SnO$_2$ powder samples was determined to be smaller than 60 nm. The radioactive $^{111}$In and $^{181}$Hf tracers were introduced in the powder samples during the sol gel chemical process. Radioactive $^{111}$In was implanted on the SnO$_2$ thin films using the University of Bonn ion implanter (BONIS). PAC measurements were carried out in a four BaF$_2$ detector spectrometer in the temperature range of 77–973 K for samples annealed at different temperatures. The PAC results for both nuclear probes show the presence of two electric quadrupole interactions. The major fractions in both cases correspond to the substitutional sites in the rutile phase of SnO$_2$. The results are compared with previous PAC measurements.

Keywords SnO$_2$ · PAC spectroscopy · Electric-field gradient
1 Introduction

The tin dioxide SnO$_2$ semiconductor with a wide band gap is quite attractive for the fabrication of diluted magnetic semiconductors (DMS) because of its excellent optical transparency and the fact of having native oxygen vacancies with high carrier density. DMS are a class of materials that have been studied intensively in recent years due to their main application in the area of spintronics. There are other applications such as flat panel display devices, solar cells and lasers [1–7]. Some experiments have been conducted to investigate the magnetism in SnO$_2$ [8–11] doped with transition metals however controversial results have been reported in compounds prepared by different methods. More recently it has been observed that the sintering temperature, to which the SnO$_2$ is submitted, can influence the magnetic ordering of the same sample [12]. Other studies concluded that the defects in the crystal structure are responsible for the ferromagnetism, instead of 3d or 4f ions doping. Recent research in this area includes experimental and theoretical investigation and involves several different metal oxides as well as experimental techniques. Whereas there are reports of several studies involving SnO$_2$, TiO$_2$ and other oxides in the literature using different experimental technique there are very few investigations which used microscopic technique such as PAC to study SnO$_2$.

2 Experimental

The powder samples of pure and 3 and 5 % Mn-doped SnO$_2$ were prepared using the sol gel method [1]. The resulting powders were pressed into pellets and submitted to a thermal treatment at 1173 K for 12 h. Thin films of SnO$_2$ were prepared on silicon substrate (100) with sputtering technique using 99,999 % pure Sn target in Ar gon and oxygen atmosphere. The thickness of the film was $\sim 100$ nm, measured with a white-light-interferometer (Filmetrics Model F40 230–243). The samples were characterized by x-ray diffraction (XRD), scanning electron microscopy (SEM) and energy dispersive spectroscopy (EDS) at the Polytechnic Institute, University of Sao Paulo using samples without radioactive nuclear probes. The radioactive probe $^{111}$In-$^{111}$Cd was introduced in the samples during the sol gel chemical process by adding carrier free $^{111}$InCl$_3$ solution with an activity of $\sim 20 \mu$Ci. In a similar process the probe $^{181}$Hf was introduced by adding the $^{181}$HfF$_4$ solution, obtained by dissolving a small quantity ($\sim 0.1$ mg) of neutron irradiated Hf metal in a few drops of dilute hydrofluoric acid [1]. Radioactive $^{111}$In was implanted into thin film using University of Bonn ion implanter BONIS with beam energy of 160 keV. A rapid thermal annealing ($T_A$) was carried out at 873 K for 10 min before PAC measurements. The PAC measurements were performed using a spectrometer consisting of four conical BaF$_2$ detectors and a slow-fast coincidence system for measuring the delayed gamma-gamma coincidence spectra.

3 Results and discussion

The EDS, SEM and XRD results for 3 % Mn-doped powder sample are shown in Figs. 1, 2 and 3, respectively. The EDS spectra show mainly the presence of Mn in