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TDPAC study of a solid-state reaction doping process of 181Hf(→181Ta) impurities in the Ho2O3 semiconductor

Nuclear methods and, in particular, the time-differential perturbed-angular correlation (TDPAC) spectroscopy have been extensively applied to study materials to elucidate at a subnanoscopic scale the environment of impurities or constituent atoms of solids (see, e.g., Ref. 1). In the case of TDPAC, a suitable probe isotope, generally an impurity in the system under study, is used and the information provided, at this probe site, is given as a product of a nuclear and an extra nuclear quantity. In the case of electric-quadrupole interactions, the nuclear quantity is the nuclear quadrupole moment (Q), characteristic of a given nuclear state, which interacts with the electric-field gradient (EFG) acting on the site of the probe atom. Since the EFG mostly originates in the non-spherical electronic charge density close to the impurity nucleus, the TDPAC technique can be used as a powerful tool in order to study the electronic structure (and related structural, electronic or magnetic properties) in the close neighborhood of the probe. The probe can be introduced in the host material by different methods: thermal diffusion, chemical methods, neutron activation, or ionic implantation. In this work we study an alternative doping method: ball-milling-assisted solid-state reaction between neutron-activated m-HfO2 and the system under study (in the present case, the bixbyite Ho2O3). In order to follow the doping process of 181Hf donor impurities in the semiconductor Ho2O3 and to elucidate the effect of each variable involved in the process (milling time, temperature, ball to powder ratio, etc.), TDPAC experiments were carried out after each step of the doping process. The obtained hyperfine parameters were compared to those of m-HfO2, to those expected for Ho2O3 using a well established EFG systematics for 181Ta in bixbyte sesquioxides, and to TDPAC results obtained in Ho2O3 samples doped by ion implantation of $181Hf(\rightarrow 181Ta)$. As we will show, we can determine the effect played by the milling and the thermal treatments, showing the capability of the TDPAC technique to follow the doping process and to give information about the inter-diffusion processes. We also demonstrate the excellent efficiency of the ball-milling-assisted solid-state reaction process to locate Hf donor impurities at the defect-free cationic sites of the Ho2O3 semiconductor, quantifying directly the amount of impurities doped after each step of the process.

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yes

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Poster contribution

Summary

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