

PAC Spectroscopy and Diffusion Effects

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Perturbed angular correlation (PAC) spectroscopy is a technique to study the local structure around probe atoms in materials. It belongs to the nuclear spectroscopy methods such as NMR and Mößbauer spectroscopy. In PAC spectroscopy the hyperfine interaction between electric and magnetic fields produced by the sample material and the nuclear moments of the probe atoms results in a perturbation of the angular correlation of the emitted γ -rays. Fig. 1 illustrates the fundamentals. An appropriate probe atom decays over an intermediate state, which has a short life time. γ -rays emitted in transitions

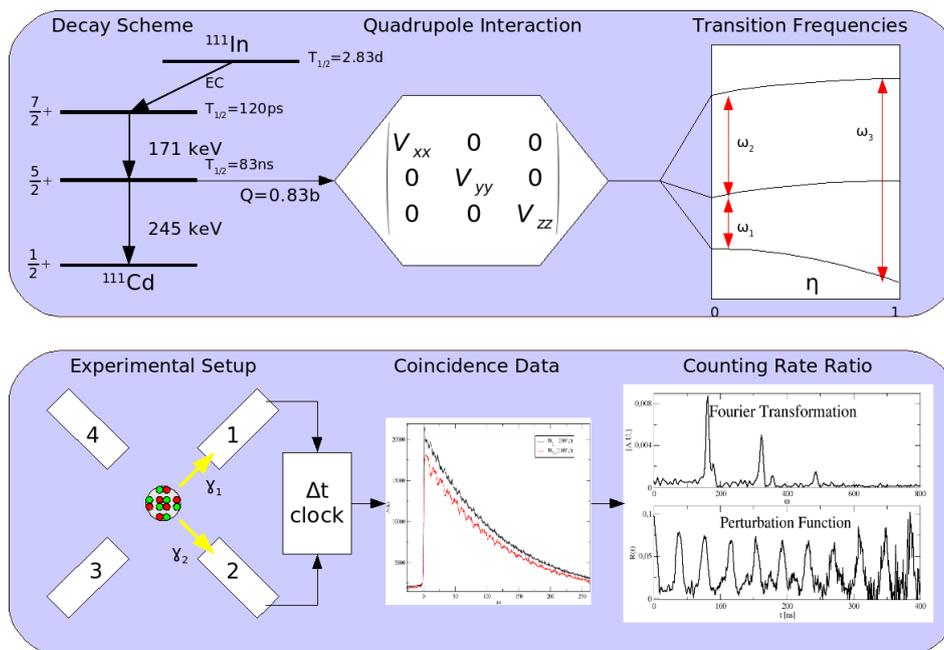


Fig. 1 PAC schema.

to and from that sensitive level are used as start and stop signals to detect the decay curve of the intermediate state. Because of the anisotropy of the two angularly correlated γ -rays and the perturbation by electric and magnetic fields, the intensity of emitted γ -rays varies slightly over time which can be ob-

served as ripples on the decay curve of the sensitive level. For quadrupole interactions the interesting parameters which can be obtained by PAC spectroscopy are: fraction of sites (f), quadrupole interaction frequency (ν_Q) and its damping (δ) and the asymmetry parameter (η). For magnetic dipole interactions: fraction of sites (f), Larmor frequency (ω_L) and its damping (δ). Measurements are usually recorded at different temperatures. Changes in the phase or the magnetism can be studied as well as defects or changes of the site's fraction. Ref. [1] provides an overview.

Atomic dynamics in the material have also an effect on the PAC data. When the first γ -quantum is emitted and the probe's neighborhood changes before the second γ -quantum is emitted, the correlated information gets basically lost. For example, if the change is caused by hopping of atoms close to the probe atom, the probability that a hop will occur within the time window of this probe atom increases with time. In the PAC time diagram this results in a decrease of the perturbation's amplitude with increasing time. From the data, the hopping frequency can be obtained [2,3]. Fig. 2 illustrates such a PAC spectrum.

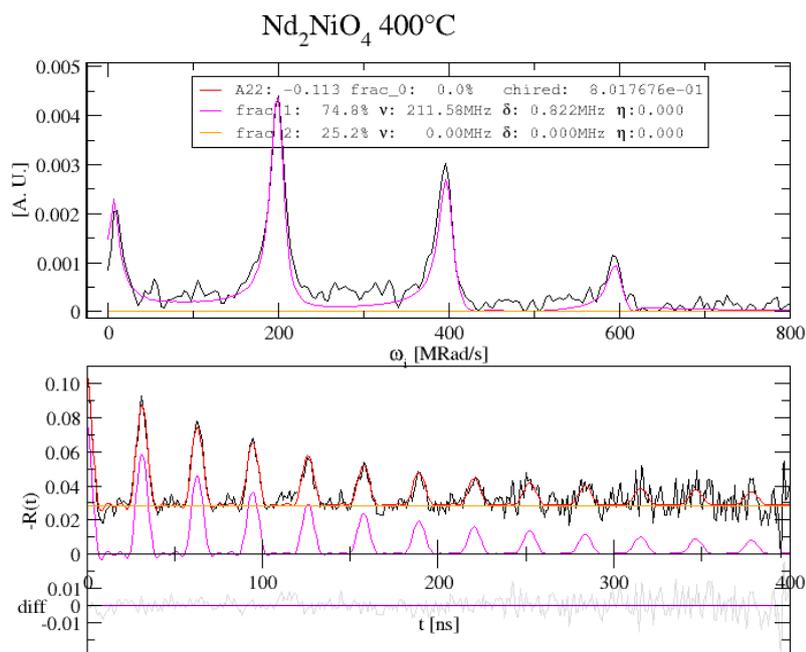


Fig. 2 $^{111}\text{In}/^{111}\text{Cd}$ PAC on Neodymium nickel oxide at 400 °C.

Fig. 3 shows another effect of dynamic processes in a PAC spectrum. With increasing temperature a sharpening in the spectrum is observed. Such a case has been described and simulated in Ref. [4]. Here, a nucleus is considered which undergoes a transition from a situation at where it “sees” many different EFGs to a situation which can be characterized by one EFG only. A PAC spectrum with many different EFGs would correspond to the spectrum at lower temperature in Fig. 3. The spectrum at the highest temperature in Fig. 3 would correspond to the single EFG case (here in the spectrum actually two EFGs exist).

For the $^{111}\text{In}/^{111}\text{Cd}$ probe the first level has a decay time of 120 ps as seen in Fig. 1. If the dynamic process is in the time scale of the first decay level, a certain part of the probe's population is in the final single EFG when the decay starts from the first level. This population experiences an undisturbed decay. The other population has a wide distribution due to the many EFGs. In total, this results in a PAC spectrum with sharp frequencies at lower amplitude but with no damping over time. With increasing temperature the jump rate increases and increasingly populates the group of probe atoms of the final single EFG, which results in an increase of the amplitude. In Fig. 3 two final EFG are observed.

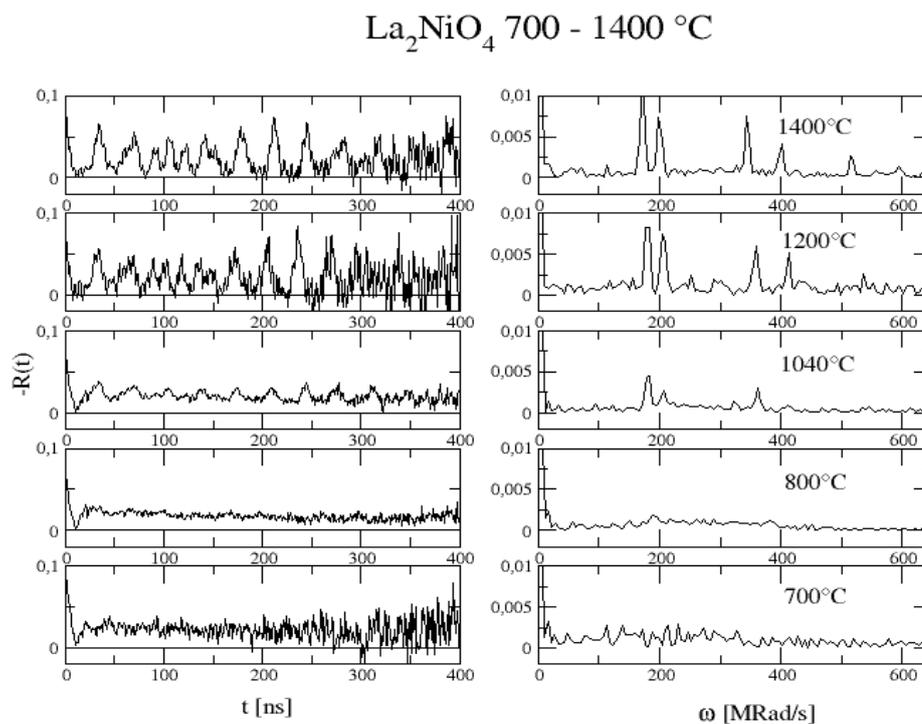


Fig. 3 $^{111}\text{In}/^{111}\text{Cd}$ PAC on Lanthanum nickel oxide at different temperatures [5].

PAC spectroscopy is a challenging method to study local structure in materials. It also allows studying dynamic effects in materials by selecting appropriate probe atoms.

References

- [1] G. Schatz, A. Weidinger: Nukleare Festkörperphysik, Teubner Studienbücher (1992).
- [2] A. Abragam, R. Pound, Phys. Rev. 92 (1953) 943.
- [3] U. Bäverstam, R. Othaz, N. de Sousa, B. Ringstöm, Nuc. Phys. A 186 (1972) 500.
- [4] D. Lupascu, Dissertation Universität Göttingen (1995).
- [5] J. Röder, M. Uhrmacher, K. D. Becker, Hyp. Int. 178 (2007) 31.