

Quadrupole Interaction of ^{172}Yb and ^{168}Er Nuclei in the First Excited 2^+ State

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We have measured the hyperfine interactions in YbVO_4 and YbPO_4 using ^{172}Yb perturbed angular correlation spectroscopy for 90 - 1094 keV and 1094 - 79 keV γ - γ cascades. The quadrupole interaction frequency varied with temperature from 0 to 2 GHz. Precise information was obtained from both cascades independently even using the cascade with the intermediate state of the half life as short as 1.6 ns.

Key words: Quadrupole Interaction; Perturbed Angular Correlations; ^{172}Yb ; ^{168}Er ; YbVO_4 .

1. Introduction

The perturbed angular correlation (PAC) spectroscopy is less frequently applied to study nuclear quadrupole interaction in solids than traditional techniques, such as nuclear and quadrupole resonances and Mössbauer spectroscopy. PAC technique, however, has a remarkable advantage: its sensitivity is absolutely temperature independent. On the other hand it is rather limited to a relatively small number of radioactive isotopes which have an excited state with appropriate characteristics.

Among PAC isotopes used for nuclear quadrupole interaction studies ^{172}Yb is exceptional since its two different excited states are suitable for measurements in the time-differential way [1, 2]. These states are populated by γ transitions following the EC-decay of ^{172}Lu with the convenient half-life of 6.7 days. Three from over one hundred γ transitions form a triple cascade with the energies 90 - 1094 - 79 keV. The half-life and spin of the successive intermediate states are: 8.3 ns and $I = 3$, 1.6 ns and $I = 2$, respectively.

When the 90 - 1094 keV cascade is chosen for hyperfine interaction measurements, the perturbation factor can be observed for a relatively long time of more than 40 ns, i. e. for a period about 5 times longer than the half-life of the isomeric state. For the 1094 - 79 keV cascade the half-life of the intermediate state

is much shorter, and therefore the observation time is less than 10 ns. Usually, such a short observation time makes it difficult to determine the quadrupole interaction frequency precisely. As a consequence, PAC experiments are very seldom performed using nuclear probes with an isomeric state of about 1 ns half-life. In the following we will compare the results which were obtained using two different γ - γ cascades of ^{172}Yb and will show that quite precise information can be obtained even using the short living isomeric state.

2. PAC Measurements with ^{172}Yb Probe

The measurements were performed with ^{172}Yb in two ytterbium compounds: YbVO_4 and YbPO_4 . The parent isotope ^{172}Lu was produced directly in the samples during 20 MeV proton beam irradiations. After the irradiations the samples were sealed in a quartz tube and then placed into a small oven or cryostat in a PAC setup. The temperature ranged from 20 to 1000 K.

The fortuitous energy and placement of γ transitions in the decay scheme allows the time spectrum of the two cascades to be recorded simultaneously. We measured four time spectra with two start and two stop BaF_2 counters. In each spectrum the events due to 90 - 1094 keV and 1094 - 79 keV coincidences were

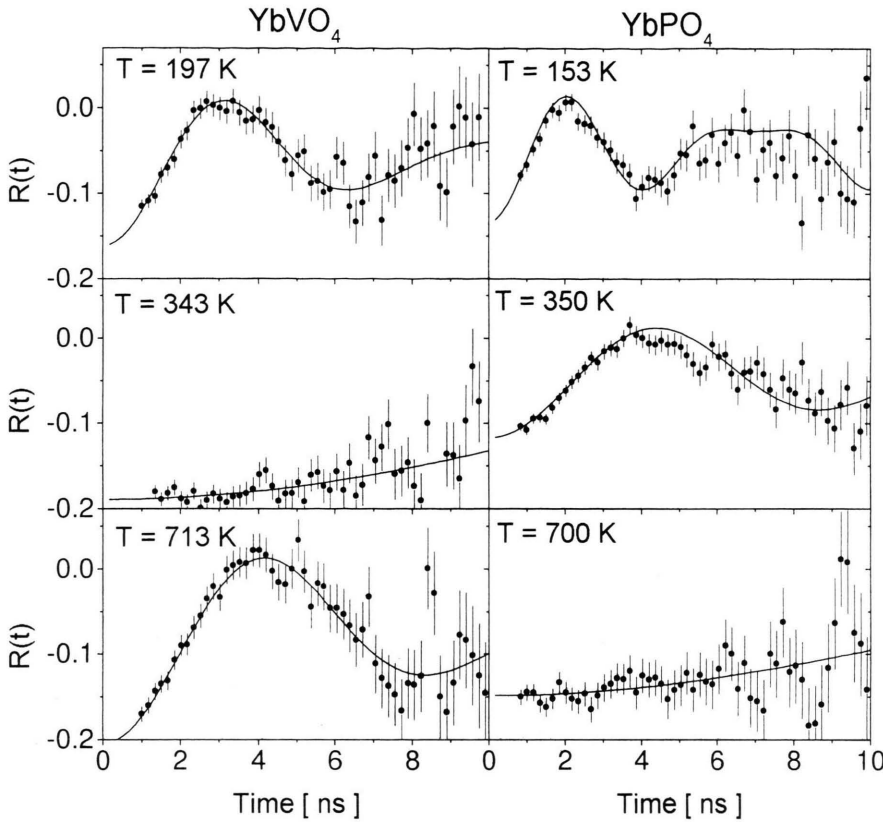


Fig. 1. Examples of PAC spectra for the 1094 - 79 keV cascade of ^{172}Yb in YbVO_4 and YbPO_4 . The prompt coincidence contribution affects strongly the experimental points up to 1.5 ns.

stored, respectively, to the right and to the left from the time zero point. Examples of the usual ratio $R(t)$, derived from these spectra, are shown in Figure 1.

3. Results

The YbVO_4 and YbPO_4 compounds crystallize in the tetragonal zircon structure with all Yb ions being at equivalent sites of D_{2d} symmetry. Therefore, a unique component in the PAC spectra is expected with the shape corresponding to an axial electric field gradient. Indeed, all the spectra could be very well fitted assuming one component. There were two fitted parameters: the effective anisotropy A^{eff} and the quadrupole interaction frequency $\nu_Q = eQV_{zz}/h$.

The fitted values of A^{eff} depend on the experimental conditions and range from +0.06 to +0.09 and from -0.14 to -0.19 for the 90 - 1094 keV and 1094 - 79 keV, respectively. These values are in fair agreement with the reported values of A_2 for both cascades

[3] if one takes into account the geometrical factors and the fact that the coincidence spectra contain substantial admixture of coincidences with the Yb x-rays and the Compton background. The fitted values of ν_Q are plotted in Figs. 2 and 3 for both observed cascades and for both investigated compounds. The difference in ν_Q^1 for the 90 - 1094 keV cascade and ν_Q^2 for the 1094 - 79 keV cascade comes from the different quadrupole moments of Yb nuclei in two involved excited states. The ratio of ν_Q^1 and ν_Q^2 gives the ratio of the quadrupole moment of ^{172}Yb at 1173 and 79 keV excited states being 1.35(3).

The YbVO_4 and YbPO_4 appeared to be appropriate to compare the results derived from two cascades since in these compounds the strength of the quadrupole interaction varies in a wide range. As it was expected, the quadrupole interaction frequency ν_Q^1 was generally determined more precisely than ν_Q^2 . Nevertheless, the results obtained using the 1094 - 79 keV cascade are accurate enough to reproduce

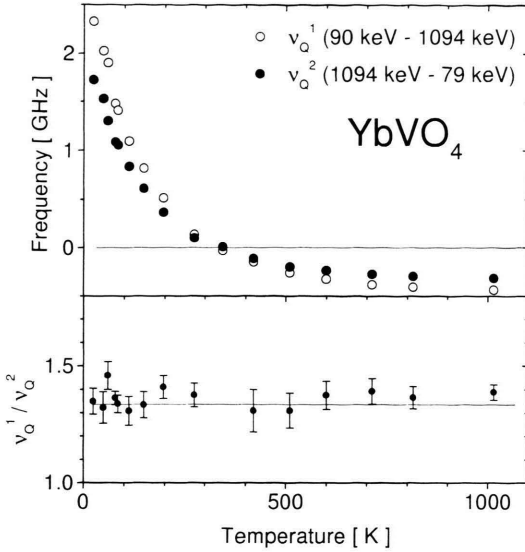


Fig. 2. Temperature dependence of the quadrupole interaction constant ν_Q in YbVO_4 derived from the PAC spectra for the 90 - 1094 keV (open points) and 1094 - 79 keV (solid points) cascades. At the bottom the ratio $\nu_Q(1094 - 79)/\nu_Q(90 - 1094)$ is plotted.

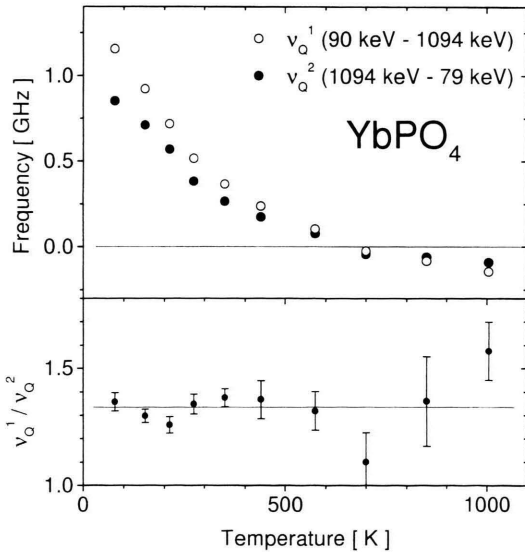


Fig. 3. Temperature dependence of the quadrupole interaction constant ν_Q in YbPO_4 derived from the PAC spectra for the 90 - 1094 keV (open points) and 1094 - 79 keV (solid points) cascades.

the same temperature dependence. It is particularly valid when the quadrupole interaction frequency is large. For a weak quadrupole interaction however,

the results are less certain and the ν_Q^1/ν_Q^2 ratio deviates from the average value. This is well seen at about 350 K for YbVO_4 and at about 650 K for YbPO_4 . This anomaly may be attributed to the known effect: when the quadrupole interaction pattern is observed only for a very short time it becomes difficult to distinguish between a small but unique EFG and a distribution of the EFG's magnitude around a small average value. One may conclude that the application of the 1094 - 79 keV cascade is seriously limited when the quadrupole interaction frequency drops below 60 MHz.

4. Electric Field Gradient in YbVO_4 and YbPO_4

The hyperfine quadrupole interaction was investigated in the YbVO_4 and YbPO_4 compounds with Mössbauer spectroscopy below 70 K [4]. The present measurements extend these investigations to high temperatures, giving a clear indication of two significant sources of the electric field gradient at Yb nuclei. The first source is the charge of the neighbouring ions distributed in the crystal lattice. The lattice contribution practically doesn't change with temperature. The second contribution is induced by the 4f electron shell of the probe ion itself. For Yb^{3+} ion, the ground state of the partially filled 4f electron shell has $J = 7/2$. It splits in a crystal electric field (CEF) into 4 doublets. Each CEF level contributes to the EFG with a different amount. The resultant EFG changes with temperature together with the gradually varying population of the four CEF levels according to the Boltzmann distribution.

Figures 2 and 3 show that the maximum value of the ν_Q is at low temperature when the 4f electron contribution to the EFG is fully determined by the wave function of the ground state. The decrease of ν_Q with increasing temperature demonstrates thermal excitations of the Yb ion to the first and then to the next excited states. At about 350 K in YbVO_4 and about 650 K in YbPO_4 , the quadrupole interaction frequency equals zero. The 4f electron and the lattice contributions must then be equal. The total EFG vanishes, since these contributions are of the opposite signs [5]. We attributed the negative sign to ν_Q above these temperatures to emphasize the change of sign of the total EFG. When the temperature increases further, the 4f electron contribution approaches zero since all 4f levels become equally populated. The total EFG saturates at the value equal to the lattice contribution. The quantitative description of the

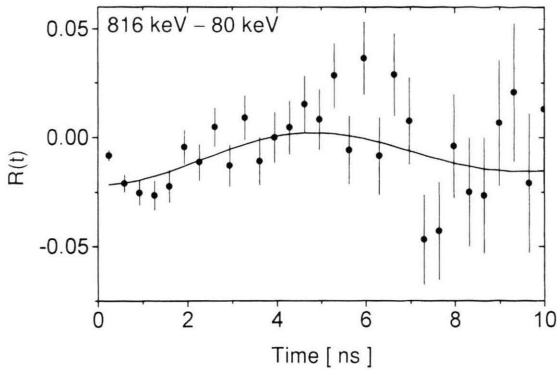


Fig. 4. PAC spectrum for ^{168}Er in erbium metal measured at 295 K.

EFG temperature dependence involves a crystal field hamiltonian [6] and it will be discussed elsewhere.

5. PAC Measurements with ^{168}Er Probe

The quadrupole interaction investigations were extended to another PAC probe with a short half-life

of the isomeric state, which, to our knowledge, has not been applied yet. We have used the 816 - 80 keV γ - γ cascade passing through the 1.8 ns, $I = 2^+$, intermediate level in ^{168}Er , which is populated in the EC decay of 90 days ^{168}Tm . The ^{168}Er probe was in erbium metal. Figure 4 shows the PAC spectrum which was fitted with the same perturbation factor as the spectra of the ^{172}Yb 1094 - 79 keV cascade. The following results were obtained: $A^{\text{eff}} = -0.03(1)$ and $\nu_Q = 240(20)$ MHz. The effective anisotropy is somewhat smaller than expected based on the reported value of $A_2 = -0.165(6)$ [7]. The obtained quadrupole interaction frequency together with the low temperature Mössbauer effect data [8] indicate that the EFG is strongly temperature dependent. The 4f shell must contribute significantly to the EFG at Er nuclei in erbium metal.

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