A Comparative Time Differential Perturbed Angular Correlation Study of the Nuclear Quadrupole Interaction in $HfF_4 \cdot HF \cdot 2H_2O$ Using ^{180m}Hf and ^{181}Hf(β^-)^{181}Ta as Nuclear Probes: Is Ta an Innocent Spy?

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We report on a comparative study of the nuclear quadrupole interaction of the nuclear probes ^{180m}Hf and ^{181}Hf(β^-)^{181}Ta in $HfF_4 \cdot HF \cdot 2H_2O$ using time differential perturbed angular correlations (TDPAC) at 300 K. For the first probe, assuming a Lorentzian frequency distribution, we obtained $\omega_Q = 103(4)$ Mrad/s, an asymmetry parameter $\eta = 0.68(3)$, a linewidth $\delta = 7.3(3.9)\%$, and full anisotropy within experimental accuracy. For the second probe, assuming a Lorentzian frequency distribution, we obtained three fractions: (1) with 56.5(7)%, $\omega_Q = 126.64(4)$ Mrad/s and $\eta = 0.9241(4)$ with a rather small distribution $\delta = 0.40(8)\%$ which is attributed to $HfF_4 \cdot HF \cdot 2H_2O$; (2) with 4.6(4)%, $\omega_Q = 161.7(3)$ Mrad/s and $\eta = 0.761(4)$ assuming no line broadening which is tentatively attributed to a small admixture of $Hf_2OF_6 \cdot H_2O$; (3) the remainder of 39.0(7)% accounts for a rapid loss of anisotropy and is modelled by a perturbation function with a sharp frequency multiplied by an exponential factor $\exp(-\lambda t)$ with $\lambda = 0.55(2)$ ns^{-1}. Whereas the small admixture of $Hf_2OF_6 \cdot H_2O$ escapes detection by the ^{180m}Hf probe, there is no rapid loss of roughly half the anisotropy as is the case with ^{181}Hf(β^-)^{181}Ta. This loss could in principle be due to fluctuating electric field gradients originating from movements of nearest neighbour HF adducts and/or H_2O molecules after nuclear transmutation to the foreign atom Ta which are absent for the isomeric probe. Alternatively, paramagnetic Ta ions could lead to fluctuating magnetic dipole fields which, when combined with fluctuating electric field gradients, could also lead to a rapid loss of anisotropy. In any case, Ta is not an "innocent spy" in this compound.

Although ^{180m}Hf is not a convenient probe for conventional spectrometers, the use of fast digitizers and software coincidences would allow to use all γ-quanta in the stretched cascade which would greatly improve the efficiency of the spectrometer. ^{180m}Hf could also serve as a Pu analogue in toxicity studies.

Key words: TDPAC; Nuclear Quadrupole Interaction; ^{180m}Hf vs. ^{181}Hf(β^-)^{181}Ta.
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