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NUCLEAR QUADRUPOLE ALIGNMENT OF $^{180m}$Hf AND $^{175}$Hf IN HAFNIUM METAL

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ABSTRACT

$^{180m}$Hf and $^{175}$Hf nuclei have been aligned in the electric field gradient of single-crystal hafnium metal at temperatures down to 3 mK. The temperature dependence of gamma ray anisotropies yielded the electric quadrupole interaction energies for $^{180m}$Hf, $e^2qQ(8^-)=+(6.61\pm0.40) \times 10^{-18}$ erg, and for $^{175}$Hf, $e^2qQ(5/2^-)=+(1.05\pm0.50) \times 10^{-18}$ erg, as well as the E2/M1-mixing ratio of the 343-keV gamma transition of $^{175}$Lu, $\delta_{343}=-0.27\pm0.03$. 

Thermal equilibrium nuclear orientation of radioactive nuclei in metallic ferromagnetic host lattices has been applied extensively in the past to the study of magnetic hyperfine interactions. In these experiments the degree of nuclear orientation is usually determined from the anisotropy of nuclear radiations. On the other hand, nuclear quadrupole alignment has been observed in this way until now only in paramagnetic salts. In the present paper we report on the first radiative detection of pure nuclear quadrupole alignment in the electric field gradient (EFG) of a hexagonal metal.

$^{175}$Hf and $^{180m}$Hf nuclei were aligned in single-crystal hafnium metal at temperatures down to 3 mK. From the temperature dependence of the anisotropies of gamma rays emitted from oriented nuclei values for the electric quadrupole interaction (EQI) energies of both nuclear states in hafnium metal, as well as the E2/M1 mixing ratio of the 343-keV gamma transition of $^{175}$Lu were obtained. This method yields also the sign of the electric quadrupole hyperfine interaction if the angular distribution parameters of the relevant nuclear radiations are known to within at least their signs.

The single crystal hafnium metal samples were obtained from a 99.99% pure rod by spark cutting perpendicular to the [0001] direction followed by mechanical polishing and electro-
polishing to the desired thickness. The discs were then irradiated with fast neutrons which were generated by the impact of a 45 MeV deuteron beam on a beryllium target. Besides $^{175}\text{Hf} \left( t_{1/2} = 70 \text{ d} \right)$ and $^{180m}\text{Hf} \left( t_{1/2} = 5.5 \text{ h} \right)$ practically no other disturbing activities were produced in this way. The irradiation was so arranged that the two activities had a ratio close to one at the time of the experiment.

The hafnium metal sample and a hexagonal $^{60}\text{Co( Co)}$ single crystal, which served as thermometer, were soft soldered, with their respective C-axes parallel, to one end of a copper fin assembly. Since a good thermal contact between copper fin and sample was of crucial importance, two different soldering techniques were employed, one using ultrasonic soldering of hafnium with indium metal, and the other using regular soldering of a nickel-plated hafnium disk with Bi/Cd eutectic alloy. For more details of the experimental low temperature technique we refer to Ref. 4. A coaxial Ge(Li)-diode positioned at $0^\circ$ with respect to the common $[0001]$ -direction of both the Hf and Co single crystals was employed to record gamma ray spectra continuously over time spans of 15 minutes during the slow warming up of the samples over a typical period of 8 hours following adiabatic demagnetization.

The temperature dependence of the reduced intensities $W(\Theta=0) -1$ of the 443-keV gamma rays of $^{180}\text{Hf}$ and of the 343-keV gamma rays of $^{175}\text{Lu}$ are presented in Fig. 1. The
data points are the results of several independent experiments with separate Hf single crystals with different thicknesses (250μ and 560μ), different strong activities, and two different soldering techniques. The fact that the results of these separate experiments agree well within statistics supports the assumption that the temperature measured with the 60Co(Co) single crystal was identical to the lattice temperature of the hafnium metal single crystal.

The solid curves in Fig. 1 are the results of least-squares fits of the data with the theoretical anisotropy function

\[ W(\Theta,T) = 1 + \sum_{k=2,4} B_k \cdot U_k \cdot F_k \cdot Q_k \cdot P_k (\cos \Theta), \]

using suitable solid-angle correction factors \( Q_k \). In both cases axial symmetry was assumed for the EFG in hafnium metal. Only in the 180mHf case, however, is the decay scheme known well enough to permit an unambiguous calculation of the re-orientation parameters \( U_k \) and of the angular distribution parameters \( F_k \). The orientation parameters \( B_k \) are statistical tensors which describe the populations of the nuclear sub-states and contain the entire temperature dependence of \( W(\Theta,T) \).

From a one parameter least-squares fit of the anisotropy versus inverse temperature data for the 443-keV gamma rays of 180Hf, a value of
\[ e^2 q Q(8^-) = + (6.61 \pm 0.40) \times 10^{-18} \text{ erg} \]

was obtained for the EQI of the 1142-keV 8^- isomeric state of \(^{180}\text{Hf}\) in hafnium metal. The positive sign follows directly from the observed positive anisotropy at \(\theta=0^\circ\). It corresponds to a positive electric quadrupole moment of the 8^- state of \(^{180}\text{Hf}\), since eq is known to be positive in hafnium metal 8.

In the \(^{175}\text{Hf}\) case the analysis is not quite as straight-forward due to less complete knowledge of the decay scheme. First, there is some uncertainty in the reorientation parameters \(U_k\). The 343-keV level of \(^{175}\text{Lu}\) is populated by first-forbidden EC decay both directly from \(^{175}\text{Hf}\) and via an 89-keV mixed transition from the 433-keV level of \(^{175}\text{Lu}\) 9. Since the relative sizes of the various first-forbidden matrix elements in the EC decay are unknown, an uncertainty in the \(U_2\) and \(U_4\) is introduced, even though an appreciable \(L=2\) component can be excluded from the smallness of the logft value 10. Second, only the magnitude of the E2/M1-mixing ratio of the 343-keV gamma transition of \(^{175}\text{Lu}\) is known, \(|\delta_{343}| = 0.26 \pm 0.03\), 12 causing an ambiguity in both the signs and the magnitudes of the angular distribution parameters \(F_k\).

Accordingly, the anisotropy versus inverse temperature data for the 343-keV gamma rays of \(^{175}\text{Lu}\) were least-
squares fitted with both $\delta_{343}$ and the EQI energy as free adjustable parameters; thereby, $U_2$ and $U_4$ were varied between the extremes corresponding to pure $L=0$ and $L=1$ EC-decay transitions of $^{175}\text{Hf}$. This procedure leads to increased errors in the final values for the two fit parameters. Assuming a positive sign for the EQI energy, corresponding to a positive electric quadrupole moment of $^{175}\text{Hf}$, a value of

$$\delta_{343} = -0.27 \pm 0.03$$

was obtained for the mixing ratio of the 343-keV transition, in excellent agreement with the result of Erman et al.\textsuperscript{12} A negative sign for the EQI energy would lead to values for $\delta_{343}$ which are in complete disagreement with the result of Ref.\textsuperscript{12}. For the EQI energy of $^{175}\text{Hf}$ in hafnium metal we obtain

$$e^2 qQ(5/2) = +(4.05 \pm 0.50) \cdot 10^{-18} \text{ erg}.$$  

A comparison of the present result with the EQI energy of the $2^+$ state of $^{178}\text{Hf}$ in hafnium metal, $e^2 qQ(2^+) = -(2.93 \pm 0.10) \cdot 10^{-18} \text{ erg}$, using $Q^_Q(178\text{Hf}) = +6.81 \pm 0.06$ barn\textsuperscript{13}, yields values for the spectroscopic and intrinsic nuclear quadrupole moments, $Q$ and $Q_0$, respectively, of both nuclear states. Table 1 summarizes the results.

Since the intrinsic quadrupole moment of the $K=0$ groundstate band of $^{180}\text{Hf}$ is known\textsuperscript{13}, we may form the ratio

$$Q_0(180\text{Hf}, K=8)/Q_0(180\text{Hf}, K=0) = 0.92 \pm 0.11.$$  

Within the limits
of error no drastic difference in deformation is observed between these two intrinsic nuclear states of $^{180}\text{Hf}$. It should also be noted that our result for the intrinsic quadrupole moment of the $K=5/2$ band of $^{175}\text{Hf}$ fits well into the systematics of intrinsic quadrupole moments known for various hafnium isotopes.  

The present method, which is especially useful due to its sensitivity to the sign of the EQI, can readily be applied to the study of EQIs of several other relatively long-lived nuclear states. It may be hoped that a drastic increase in accuracy is possible due to observation of electric quadrupole resonance on oriented nuclei in hexagonal metals.

ACKNOWLEDGEMENT

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Table 1. Summary of EQI energies of $^{180m}_{\text{Hf}}$ and $^{175}_{\text{Hf}}$ in hafnium metal, including derived values for $Q$ and $Q_o$, respectively.

<table>
<thead>
<tr>
<th>nucleus</th>
<th>$I^\pi$</th>
<th>$K$</th>
<th>$e^2qQ$ (10^{-18} \text{ erg})$</th>
<th>$Q$ (barn)</th>
<th>$Q_o$ (barn)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{180m}_{\text{Hf}}$</td>
<td>8⁻ 8</td>
<td>+6.61±0.40</td>
<td>+4.4±0.5</td>
<td>+6.3±0.7</td>
<td></td>
</tr>
<tr>
<td>$^{175}_{\text{Hf}}$</td>
<td>5/2⁻ 5/2</td>
<td>+4.05±0.50</td>
<td>+2.7±0.4</td>
<td>+7.5±1.2</td>
<td></td>
</tr>
</tbody>
</table>
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§ In the definition of Ref. 11.


Figure Caption:

Fig. 1. Temperature dependence of the reduced intensities of the 443-keV gamma rays of $^{180}_{\text{Hf}}$ (a) and of the 343-keV gamma rays of $^{175}_{\text{Lu}}$ (b), emitted parallel to the [0001] - axis ($\theta = 0^\circ$) of hafnium metal after the decay of aligned $^{180m}_{\text{Hf}}$ and $^{175}_{\text{Hf}}$, respectively. Note the different scales of figures (a) and (b). The solid lines are the results of the fits described in the text.
(a) 180m\textsuperscript{Hf} 443 keV

(b) 175\textsuperscript{Hf} 343 keV
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