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ELECTRIC QUADRUPOLE INTERACTIONS OF ^{177}Hf IN LUTETIUM METAL AND LuIG

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Abstract: Electric quadrupole interaction of ^{177}Hf-probe in lutetium metal and lutetium iron garnet (LuIG) was determined by the PAC method. The quadrupole coupling constant $\frac{e Q V_{zz}^{eff}}{h}$ is 255 ± 65 MHz and 688 ± 53 MHz respectively. EQI in the latter case was found to be temperature independent in the range 78 — 1200 K. The origin and the magnitude of the electric field gradient in LuIG are discussed.

1. Introduction

In magnetically ordered compounds of rare earth ions the effect of nuclear electric quadrupole interaction is usually considerably weaker than that of nuclear magnetic interaction. There are, however, cases when both interactions are of comparable strengths and then, as a rule, quite complex patterns are observed in the NMR, Mössbauer and PAC-spectra. For this reason it seems desirable to know independently some basic parameters of EQI in typical rare earth magnetic compounds. In this paper we report on PAC-investigations of the strength and the temperature dependence of EQI on the diamagnetic probe-ion positioned at the »magnetic« c-site in the rare earth iron garnet.

The investigated system was lutetium iron garnet (LuIG) while the probe-ion was hafnium, generated by radioactive decay of ^{177}Lu and presumably isoelectronic with the parent ion ($Lu^{3+} \rightarrow Hf^{4+}$). The contribution of the 4f-shell to EFG,

typical for other rare earths in magnetic media, vanishes in the present case at all temperatures, thus making the probe particularly sensitive to the lattice contribution to the EFG.

As there is no favourable TDPAC- or Mössbauer case satisfying the above requirements, the best choice is the well known 208-113 keV cascade ($\tau_{1/2} = 0.5$ ns) of ^{177}Hf, which is accessible only by integral PAC-technique. On the other hand, it is practically impossible to determine by this method such important parameters of EQI as the asymmetry parameter η and the width of EFG distribution δ. However, the integral attenuation factor is inherently only slightly sensitive to these parameters. In a quite large range of η and δ, covering most of really existing cases, IPAC can supply sufficiently accurate values for the basic parameter — the EQI strength. The subsequent analysis is therefore correct only to the influence of η and δ.

2. *Measurements and results*

The integral $\gamma - \gamma$ angular correlation 9/2 + (208 keV) 9/2 — (113 keV) 7/2 — in ^{177}Hf was recorded by an automatic, three-detector setup. The sources were prepared by short time thermal neutron irradiation of lutetium chloride, lutetium metal and LuIG. Particular care was taken to minimize all external effects leading to attenuation of angular correlation. For similar reasons, each source used in these experiments had a mass of less than one miligram.

Table 1

Run	$LuCl_3$ water solution	Run	Lu metal	Run	Lu iron garnet
1	—0.187(2)	7	—0.179(3)	12	—0.162(2)
2	—0.184(2)	8	—0.180(1)	13	—0.165(3)
3	—0.184(2)	9	—0.176(1)	14	—0.164(2)
4	—0.182(3)	10	—0.174(2)	15	—0.160(3)
5	—0.186(2)	11	—0.185(3)	16	—0.164(2)
6	—0.185(2)				
	—0.185(1)		—0.179(1)		—0.163(1)

In Table 1 series of experimental values are presented for anisotropy of angular correlation for three different sources:

- water solution of $LuCl_3$ (first column);
- lutetium metal (second column), and
- LuIG (third column).

The bottom row presents corresponding mean values. All the measurements were performed under identical conditions — at room temperature, without application of external magnetic field and under the same geometry.

In the case of LuIG we attempted to detect an eventual angular shift of the correlation pattern in a perpendicular magnetizing field. Within the limit of experimental error we could not detect any effect produced by changing the direction of the perpendicular state of magnetization of the garnet.

Table 2

T[K]	Anisotropy
78	—0.163(2)
300	—0.163(1)
633	—0.166(2)
878	—0.165(2)
1060	—0.165(2)
1200	—0.166(2)

Finally, in Table 2 anisotropies determined for the LuIG — source at different temperatures from 78 to 1200 K are presented. These values are corrected for scattering in the dewar of furnace, respectively.

3. *Discussion*

Liquid source. The mean value of the first column, Table 1, when corrected for finite angular resolution of the system, gives $G_{22} A_{22} = -0.160 \pm 0.001$. This compares well to the value of $A_{22} = -0.159 \pm 0.001$ reported for a liquid source by Holmberg and Stefansson[1]. Subsequent determinations of attenuation in other media are based on the assumption that $G_{22} = 1$ for the liquid source.

Lutetium metal. This system exibits small but definite attenuation, $G_{22} = 0.97 \pm 0.01$ It is safe to assume that it originates in the static EQI alone. For an axially symmetric, randomly oriented interaction the attenuation coefficient is given by

$$G_{kk}(\infty) = \sum_n s_{kn} \frac{1}{1 + (n \omega_0 \tau_N)^2}. \quad (1)$$

The quadrupole coupling constant for Lu-metal $\frac{e Q V_{zz}^{eff}}{h} = (255 \pm 65)$ MHz was obtained from the experimental attenuation coefficient. Relatively large error is due to small $\omega_0 \tau_N$ — value.

The quadrupole moment Q, of the 113 keV state of ^{177}Hf is not known. However, remembering that this state is the first excited state of the rotational family ($K = 7/2$, $I = 9/2$), its quadrupole moment can be calculated from the strong coupling model formula

$$Q(K, I) = Q_0 \frac{3K^2 - I(I+I)}{(I+1)(2I+3)}, \tag{2}$$

where Q_0 is the intrinsic quadrupole moment. Taking into account the spectroscopic quadrupole moment value of the ground state, $Q^{7/2} = (4.5 \pm 0.5)$ b[2], we obtained for the 9/2 — state $Q^{9/2} = (1.75 \pm 0.2)$ b. This value is used for the determination of the electric field gradient from quadrupolle coupling constant The result is $V_{zz}^{eff} = (6.0 \pm 1.7)\, 10^{17}$ V/cm^2. It agrees well with the value of $V_{zz}^{eff} = (5.0 \pm 0.3)\ 10^{17}$ V/cm reported by Butz[3], who applied the TDPAC method on ^{181}Ta probe in lutetium metal.

Lutetium iron garnet. As this compound is ferrimagnetic below the Neel temperature ($T_N = 549$ K), nuclear magnetic interaction can not be a priori excluded, even in the case of diamagnetic ion. In the present case, however, we could not detect any shift of the angular correlation pattern in a perpendicularly magnetized sample, nor any temperature dependence of the attenuation (Table 2). These facts suggest that eventual magnetic interaction is probably quite weak. We therefore assume that here again the major part of attenuation originates from a static EQI.

Similar to the case of metal, we deduce for the garnet system $\frac{e Q V_{zz}^{eff}}{h} =$ $= (688 \pm 53)$ MHz and the gradient of the electric field $V_{zz}^{eff} = (16.3 \pm 2.4) \cdot 10^{17}$ V/cm^2. If the gradient V_{zz}^{eff} in the present system is interpreted as

$$V_{zz}^{eff} = (1 - \gamma_\infty)\, V_{zz}^{latt}, \tag{3}$$

we can deduce the lattice contribution by applying $\gamma_\infty = -68.12$, as the lattice antishielding factor of Hf[4]. The result is $V_{zz}^{latt} = (23.5 \pm 3) \cdot 10^{15}$ V/cm^2.

In Table 2 the anisotropies of the angular correlation are presented for different temperatures between 78 and 1200 K. It can be observed that attenuation is virtually temperature-independent in this temperature range. Such behaviour is compatible with the lattice origin of EFG.

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ELEKTRIČNA KVADRUPOLNA INTERAKCIJA JEZGRA ^{177}Hf U LUTECIJUM METALU I LuIG

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Sadržaj

Metodom perturbiranih ugaonih korelacija određena je električna kvadrupolna interakcija jezgra ^{177}Hf u lutecijum metalu i feritu-granatu LuIG. Kvadrupulna konstanta interakcije $\frac{eQV_{zz}^{eff}}{h}$ iznosi 255 ± 65 MHz i 688 ± 53 MHz respektivno. Temperaturska zavisnost za slučaj granata je konstantna u intervalu 78 — 1200 K. Razmatrano je poreklo i veličina gradijenta električnog polja za slučaj granata.