Measurements of Quadrupole Interactions by Nuclear Orientation and Quadrupole-Interaction-Resolved NMR on Oriented Nuclei*

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The techniques of quadrupole-interaction nuclear-orientation and quadrupole-interaction-resolved NMR on oriented nuclei were applied to radioactive ^{111}In ($T_{1/2} = 2.8$ d), ^{198}Au ($T_{1/2} = 2.7$ d) and ^{199}Au ($T_{1/2} = 3.1$ d), mass-separator-implanted into single crystals of hcp Co and hcp Gd. For ^{111}In**Gd** the quadrupole interaction was observed via the broadening of the resonance and the dependence of the effective quadrupole interaction on the angle θ between the c-axis of the single crystal and the direction of magnetization. For ^{198}Au**Gd** the large electric field gradient known from the literature could not be confirmed. For ^{198}Au**Co** and ^{199}Au**Co** the quadrupole substructure has been resolved, and the electric field gradient of Au in hcp Co was determined to be $-0.84(4) \times 10^{17}$ V/cm^2. The magnetic hyperfine fields of Au in hcp and fcc Co differ by about 20%. These experiments have shown that hcp Co may be a good host matrix for the determination of the quadrupole interaction of heavy radioactive nuclei with resonance precision.

1. Introduction

The study of the electric quadrupole interaction of radioactive nuclei in noncubic environments with the methods of quadrupole-interaction nuclear-orientation (QI–NO) and quadrupole-interaction-resolved NMR–ON on oriented nuclei (QI–NMR–ON), with which magnitude and sign of the interaction can be determined, may have two different aims: (i) Measurement of quadrupole moments of (short-lived) radioactive nuclei utilizing a matrix for which the electric field gradient (EFG) is known. Nuclear quadrupole moments are interesting as they provide a direct measure of the nonsphericity of the nuclear charge distribution. The sign yields information on the shape and/or the ground state configuration. (ii) Measurement of EFG's using radioactive isotopes with known quadrupole moments. EFG's in non-cubic metals are interesting as they are not well understood theoretically up to now. Even the sign cannot be predicted reliably. A further advantage of QI–NO and QI–NMR–ON is the fact that the measurements can be performed on impurity systems with concentrations far below the ppm level.

The angular distribution of γ-rays emitted in the decay of oriented radioactive nuclei is given by

$$W(\theta) = \sum_k B_k(\nu_M, \nu_Q, T) A_k P_k(\cos\theta) . \qquad (1)$$

The parameters B_k describe the degree of orientation; they depend on the magnetic and electric hyperfine splitting frequencies

$$\nu_M = |g \mu_N B^{eff}/h| ,$$
$$\nu_Q = e^2 q Q/h , \qquad (2)$$

where g and Q are the nuclear g-factor and electric quadrupole moment, B^{eff} and eq are the effective magnetic field and the EFG at the impurity site, and T is the temperature of the system. The A_k in (1) are parameters fixed by the nuclear decay cascade, which can be calculated for favorable cases or be measured with other techniques. In QI–NO measurements $\nu_{M,Q}$ are determined from the measurement of the temperature dependence of $W(\theta)$.

In the QI–NMR–ON method the resonant change of $W(\theta)$ is used as the detector for NMR. In

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the case of a large magnetic interaction onto which a small quadrupole interaction is superimposed, a set of $2I$ subresonances are observed which are grouped around ν_M and which are separated by $\Delta\nu_Q = 3\nu_Q/[2I(2I-1)]$, where I is the nuclear spin. As NQR–ON has not been observed up to now, probably because of the missing enhancement factor for the rf field, QI–NMR–ON is the most promising method for the precise determination of quadrupole moments of radioactive nuclei with resonance precision up to now. The measurement of the quadrupole subresonance structure in ferromagnetic Fe and Ni is limited to a few favorable cases as the EFG's in Fe and Ni are relatively small. Thus matrices are desirable in which a larger EFG is present in addition to a large magnetic hyperfine field, the latter providing a large degree of orientation at the "convenient" temperature of ~ 10 mK and the enhancement factor for the rf-field. Here we report on first QI–NMR–ON measurements with hcp Co and hcp Gd as host matrices.

2. Experimental

All samples were prepared by mass-separator-implantation of the radioactive isotopes ^{111}In, ^{198}Au and ^{199}Au into single crystals of hcp Co and hcp Gd. The samples were cooled to ~ 10 mK using an adiabatic demagnetization cryostat, and the angular distribution was measured either as a function of the temperature, which was determined with a ^{60}Co**Co**(hcp) thermometer, or as a function of the frequency of the applied rf-field. Details of the cryostat and the data acquisition system are described elsewhere [1].

3. Results and Discussion

^{111}In**Gd**

NMR–ON on a radioactive impurity in Gd was detected for the first time. The quadrupole interaction was found to be small, i.e., the quadrupole substructure could not be resolved. It could be determined, however, from the dependence of the NMR line width and the resonance shift on the external magnetic field. We will not describe the details here, as these have been published elsewhere in the mean time [2].

^{198}Au**Gd**

From Mößbaüereffekt measurements on ^{197}Au**Gd** Perscheid and Forker [3] reported a large quadrupole splitting, $\nu_Q = +207(10)$ MHz. With the known ratio of quadrupole moments, $Q(^{198}\text{Au})/Q(^{197}\text{Au}) = +1.39(5)$, the quadrupole interaction for ^{198}Au**Gd** would have been expected to be $+288(17)$ MHz, i.e., comparable or even larger than the expected magnetic interaction. Such a large quadrupole interaction can be detected easily with two NO measurements performed with the magnetization parallel and perpendicular to the c-axis of the single crystal, respectively. In this way a determination of ν_Q is possible, which is nearly independent on the other unknown parameters ν_M and the fraction f of nuclei on substitutional lattice sites. The parameters ν_M and f are strongly correlated and cannot be determined independently. Figure 1 shows a typical result. From the overlap of the two shaded areas the quadrupole splitting is determined to be $\nu_Q = -15(12)$ MHz. Taking into account possible systematic errors resulting from the different magnetization behaviour for $B \parallel c$ and $B \perp c$, an upper limit for the quadrupole interaction is

$$|\nu_Q| \lesssim 35 \text{ MHz}.$$

This is in striking disagreement with the result of Perscheid and Forker [3], and we cannot offer a unique explanation for this discrepancy at present. These authors used melted samples, for which it

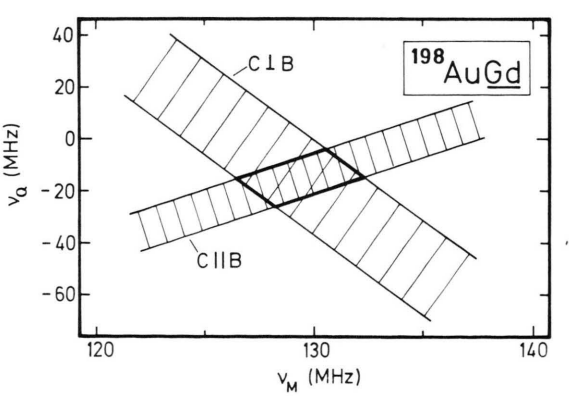

Fig. 1. Results from least-squares fits to the γ-anisotropies of the 412 keV transition of ^{198}Au in Gd, measured fully magnetized parallel and perpendicular to the c-axis of the Gd single crystal.

cannot be anticipated a priori that the Au nuclei are substituted onto regular lattice sites. Thus it is possible that their large EFG results from Au nuclei at nonsubstitutional lattice sites or from cluster compounds. This explanation is supported by the fact that Perscheid and Forker derived $\theta \sim 60°$ for the angle between the easy magnetization and the c-axis which is in striking contradiction to $\theta \sim 30°$ known from many other measurements with different techniques. No final conclusions can be drawn at present, with the exception that Gd seems now to be not the ideal matrix for on-line measurements of quadrupole moments of heavy nuclei.

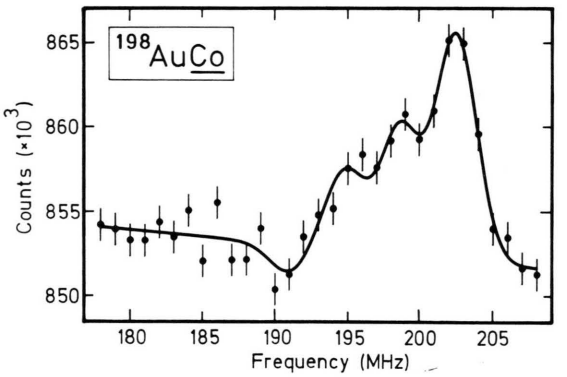

Fig. 2. QI–NMR–ON spectrum of the 412 keV transition of ^{198}Au in a hcp Co single crystal. The line width is 3.3(3) MHz including the frequency modulation bandwidth of 2 MHz.

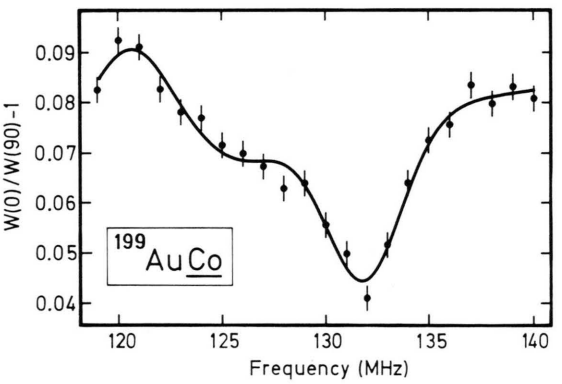

Fig. 3. QI–NMR–ON spectrum of the 208 keV transition of ^{199}Au in a hcp Co single crystal. The line width is 4.4(4) MHz including the frequency modulation bandwidth of 2 MHz.

^{198}Au**Co**(hcp) and ^{199}Au**Co**(hcp)

Figures 2 and 3 show the QI-resolved NMR–ON spectra of ^{198}Au and ^{199}Au in single crystals of hcp Co. The results are listed in Tables 1 and 2. The ratio of the magnetic and electric hyperfine splitting frequencies in hcp Co are in perfect agreement with those in Fe. The EFG of Au in hcp Co is determined to be

$$eq(\text{Au}\mathbf{Co}) = -0.84(4) \times 10^{17}\ \text{V/cm}^2,$$

which is by a factor of 7.4 larger than the EFG in Fe. This means that hcp Co will be a good matrix for the determination of the quadrupole moments of high-spin Au isomers, which are interesting as an odd-even effect for the deformation has been predicted to explain the violation of the additivity of magnetic moments.

The different hyperfine fields for ^{197}Au, ^{198}Au and ^{199}Au in Fe are well understood as being due to hyperfine anomalies. As the ratio for ^{198,199}Au in hcp Co is similar to that in Fe, which means that the ratio of non-contact to contact hyperfine fields is

Table 1. Magnetic hyperfine splitting frequencies and hyperfine fields of Au isotopes in Fe and Co.

System	ν_M/MHz	B_{HF}/kG	$B_{HF}/B_{HF}(^{198}Au)$	Ref.
^{197}Au**Fe**	93.207(10)	1258.5(2)	1.0969(9)	[4]
^{198}Au**Fe**	259.48(3)	1147.3(8)		[5]
^{199}Au**Fe**	166.69(4)	1208.2(3.1)	1.053(3)	[6]
^{197}Au**Co**(hcp)		954.5(1.4) [a]		b
^{198}Au**Co**(hcp)	196.8(2)	870.2(1.1)		b
^{199}Au**Co**(hcp)	126.2(2)	914.7(2.8)	1.051(4)	b
^{199}Au**Co**(fcc)	58.00(5)	783.1(7)		[7]

[a] Calculated with the ratios for Fe. [b] this work.

Table 2. Electric hyperfine splitting frequencies of Au isotopes in Fe and hcp Co.

System	ν_Q/MHz	$\nu_Q/\nu_Q(^{198}Au)$	Ref.
^{197}Au**Fe**	(−)1.50(5)	(+)0.718(28)	[4]
^{198}Au**Fe**	−2.09(4)		[5]
^{199}Au**Fe**	−1.52(2)	+0.727(17)	[6]
^{198}Au**Co**(hcp)	−15.4(5)		a
^{199}Au**Co**(hcp)	−11.3(3)	+0.734(31)	a

[a] This work.

similar, we deduce the hyperfine field of ^{197}Au in hcp Co to be (−)954.5(1.4) kG which is by a factor 1.219(2) larger than the hyperfine field in fcc Co, $B_{HF} = (-)783.1(7)$ kG, determined recently by a spin-echo measurement [7]. For Co in hcp and fcc Co this factor is 1.0112(6), i.e., strongly different from the present case. A systematic investigation with different impurities in hcp and fcc Co could thus introduce new aspects for the quantitative theoretical understanding of EFG's and hyperfine fields.

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