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II-12 Electric Quadrupole Interaction in the 5/2⁺ First Excited State of ¹¹¹Cd in Indium

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It is well known that metals provide excellent hosts for the measurement of hyperfine interactions using γ - γ perturbed angular correlations.¹⁾ In order to be able to use non-cubic metals as hosts for the determination of the nuclear quadrupole moments of excited states, one must first be able to predict the electric field gradients (EFG). It is partly with this object in mind that we have undertaken measurements of the electric quadrupole interaction of the 247 keV level of ¹¹¹Cd in indium metal using the 172-247 keV y-cascade from the decay of 2.8 day ¹¹¹In. Such measurements were first carried out by Albers-Schönberg et al. in 1953²⁾ and 1954³⁾ by using timeintegral angular correlations at room temperature. The first time-differential measurement was made by Lehmann and Miller in 1956.4) In 1957, Steffen⁵⁾ reported on the temperature dependence of the electric quadrupole interaction measured by timeintegral correlations. In 1962, H.-J. Behrend and D. Budnick reported a polarization-directional correlation at room temperature⁶⁾ which allowed the measurement of the sign of the quadrupole interac-An unpublished time-differential result by tion. Böstrom and Jonsson in 1971 studied the effects of combined electric and magnetic interactions on polycrystalline indium.⁷⁾ Again, in 1971, a timedifferential experiment on polycrystalline indium was carried out by Haas and Shirley.⁸⁾ The results of these room temperature measurements are shown in Table I.

In Table I, we have used the conventional definitions⁹⁾ where $\omega_0 = e^2 Qq/h$ for I = 5/2. Q is the nuclear quadrupole moment and eq is the EFG acting at the nucleus. The table clearly indicates that the timedifferential measurements are in very good agreement with each other, but in poor agreement with all the time-integral measurements. In such cases it is always preferable to trust the time-differential measurement, since time-integral measurements are subject to possible severe systematic errors.

In our data analysis, we have assumed that the static EFG is axially symmetric and randomly oriented (polycrystalline sample). A typical room temperature measurement is shown in Fig. 1. The data were taken with a 3-detector apparatus, using $1.5'' \text{ long } \times 1.5''$ diameter NaI (T1) crystals, so that coincidences vs time could be measured simultaneously at 180° and 90° angles.¹⁰ We subtracted the random coincidence rate from the two sets of data and formed the ratio

Table I.	Electric	quadrupole	interaction	of	¹¹¹ Cd in	Indium metal.	
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Reference	Method [°]	$\omega_0 \tau$	$e^2 Qq/h$ (MHz)
Albers-Schönberg et al. ²⁾	IPAC	1.59(2)	13.9(8)
Albers-Schönberg et al. ³⁾	IPAC	1.606(17)	13.80(15)
Lehmann and Miller ⁴⁾	TDPAC	_	20 17. 8(4)ª
Steffen ⁵⁾	IPAC	not reported	14. 5(8) ^b
Behrend and Budnick ⁶⁾	IPAC	-1.5(5)	+12.7(40)
Böstrom and Jonsson ⁷⁾	TDPAC		17.7(1)
Haas and Shirley ⁸⁾	TDPAC		17.3(2)
this paper	TDPAC	_	17.62(6)

^a calculated in ref. 9, p. 1178.

^b *ibid*, p. 1175.

° IPAC-time integral perturbed angular correlations.

TDPAC-time differential perturbed angular correlations.

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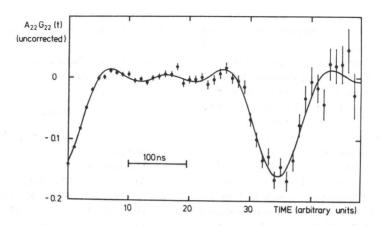


Fig. 1. Time-differential perturbed angular correlation spectrum for ¹¹¹Cd in In metal at T = 295 K. The ordinate, R, is calculated as described in the text.

$$R = 2 rac{W(\pi, t) - W(\pi/2, t)}{W(\pi, t) + 2W(\pi/2, t)}$$

= $A_{22}G_{22}(t)$,

assuming $A_{44} = 0$.

We then performed a least squares fit to a function of the form

$$R = A_2 \cdot \frac{1}{5} \left| 1 + \frac{13}{7} \cos(\omega_0 t + \phi) + \frac{10}{7} \cos 2(\omega_0 t + \phi) + \frac{5}{7} \cos 3(\omega_0 t + \phi) \right| + C .$$

This curve clearly fits the data well and confirms our assumption about the interaction.

The sharp disagreement between our value of the interaction frequency and that of Behrend and Budnick (see Table I) casts serious doubt on their value for the quadrupole moment of this level, which is the accepted value at present.¹¹⁾

Since the nuclear quadrupole resonance (NQR) of ¹¹⁵In and ¹¹³In has already been measured as a function of temperature,¹²⁾ and, since the quadrupole moments of both nuclei are known, we felt that a comparison of these experiments with that for ¹¹¹Cd in In might be illuminating.

In Fig. 2 is plotted our measured interaction frequency as a function of temperature, normalized to the frequency at 77 K,¹³ along with the NQR results of Hewitt and Taylor¹²) for ¹¹⁵In in In metal, similarly normalized. The solid curve is the temperature dependence calculated with the theory of De-Wette¹⁴) which assumes point ions and a uniform electron distribution. The lattice parameters were taken from X-ray work on pure In.¹⁵) The most striking aspect of these results is that both ¹¹¹Cd and

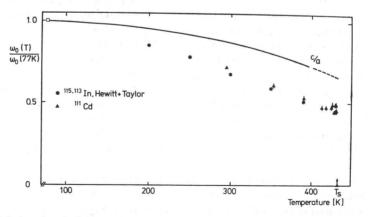


Fig. 2. Relative electric field gradients in indium metal as a function of temperature.

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¹¹⁵In in indium have almost the same temperature dependence, which is much stronger than that of the pure lattice ionic charge distribution. Since Behrend and Budnick calculated the EFG assuming point ions and homogeneously distributed electrons, this casts even further doubt on their value of the quadrupole moment. In addition, Hewitt and Taylor¹²⁾ point out that for ¹¹⁵In in In the absolute value of the EFG predicted by this model is a factor of 4 too small. Because of this, we would prefer to trust the systematics of the measured values of Q for 5/2 levels in neighboring odd neutron nuclei and the calculations of Kisslinger and Sorenson¹⁶⁾ which lead to the conclusion that Q = 0.6(1) barns. With this value we find that the EFG for ¹¹¹Cd and ¹¹⁵In in indium metal are equal within the large error caused by uncertainty in Q. If we turn this argument around and say that the EFG's are exactly equal (forgetting for the moment that the Sternheimer factors, γ_{∞} , are not exactly equal), we calculate

$$Q(^{111}\text{Cd}, 5/2) = \frac{\nu_Q(^{111}\text{CdIn})}{\nu_Q(^{115}\text{In})} \cdot Q(^{115}\text{In})$$
$$= \frac{17.62 \text{ MHz}}{28.9 \text{ MHz}} \cdot 0.861 \text{ barns}$$
$$= 0.52 \text{ barns},$$

 $v_Q(^{115}In)$ is taken from Hewitt and Taylor¹²) and $Q(^{115}In)$ is the recalculated value as discussed by Raghavan and Raghavan.¹⁷)

A test of this assumption can be made by comparing the results for the electric quadrupole interaction of ${}^{117}In_{.01}Cd_{.99}{}^{17}$ with that of ${}^{111}CdCd{}^{18}$ which yields

$$Q(^{111}\text{Cd}, 5/2) = \frac{125 \text{ MHz}}{145.1 \text{ MHz}} \cdot 0.64 \text{ barns}$$

= 0.55 barns.

The striking agreement between these values leads one to the conclusion that, at least for the system In-Cd, the EFG is characteristic of the host alone. This hypothesis invites further experimental and theoretical investigation of binary alloy systems in which (i) both components have completely filled d-shells so that the band-structure is well-behaved and (ii) the impurity is "chemically similar" to the host so that the host electronic structure will not be strongly perturbed. Examples of such systems might be In-Sn, Cd-Tl, and Cd-Sn.

For ¹¹¹Cd the pairing-plus-quadrupole model of Kisslinger and Sorenson¹⁶⁾ is applicable. They predict a value for this level of $Q(^{111}$ Cd, 5/2) = 0.62 barns. This is in quite good agreement with the values

obtained above.

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