DETERMINATION OF THE NUCLEAR PARAMETERS OF $^{155}$Gd EXCITED STATES USING THE MÖSSBAUER EFFECT

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From Mössbauer spectra of GdAlO$_3$ and GdVO$_4$ above and below the Néel temperature and fitted using a transmission integral, we have determined the following parameters of the 86.5 keV and 105 keV levels: $g(86)/g(0) = +1.217 \pm 0.005$, $Q(86)/Q(0) = +0.10 \pm 0.02$, $g(105)/g(0) = -0.55 \pm 0.02$, $Q(105)/Q(0) = +0.74 \pm 0.02$, $\Delta r^2_{105}/\Delta r^2_{86} = +1.30 \pm 0.05$. The linewidth observed for the 105 keV transition is less than the calculated natural linewidth.

1. Introduction

The Mössbauer effect study of the nuclear properties of the $^{155}$Gd isotope has been the subject of many reports over the last ten years. Of the three possible gamma transitions able to be studied in this nucleus, the majority of workers have investigated the 86.5 keV transition because of its high intensity and narrow linewidth ($2\Gamma_0 = 0.499$ mm $\cdot$ s$^{-1}$). The 60.0 keV transition has a short lifetime of 0.13 ns, ($2\Gamma_0 = 23.5$ mm $\cdot$ s$^{-1}$) and resolution of the hyperfine interactions is impossible to obtain. The 105.3 keV transition has a theoretical linewidth of 2.22 mm $\cdot$ s$^{-1}$ making it difficult to obtain spectra which are well resolved and coupled with its small recoilless fraction, this makes the unique determination of the nuclear parameters of this level a hard task.

We have recently reported some of the magnetic properties of two antiferromagnets, GdVO$_4$ and GdAlO$_3$, using the Mössbauer effect of the 86.5 keV transition [1,2]. As a result, we have been able to redetermine the magnetic and quadrupole moments of the 86.5 keV excited state. We have also investigated the 105.3 keV level using GdAlO$_3$ which, because of its high Debye temperature, produces spectra having a higher absorption than is normally observed.

GdVO$_4$ ($T_N = 2.49$ K) and GdAlO$_3$ ($T_N = 3.87$ K) both order antiferromagnetically below 4 K. Above this temperature, the gadolinium nucleus is subject to an

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electric field gradient (EFG) only and the resultant Mössbauer spectra exhibit quadrupole splitting. Below their ordering temperature, magnetic and quadrupole interactions split the nuclear energy levels, allowing the moments of each Mössbauer level to be determined.

The preparation of single crystal and powdered absorbers of the two compounds is reported in refs. [1,2]. Spectra were recorded for each powdered absorber and both along directions parallel and perpendicular to the magnetic ordering axis using single crystals. A total of 33 spectra were recorded using the 86.5 keV transition. Two single line sources were used in the study. A $^{154}\text{SmPd}_3$ source was used to record all spectra of GdAlO$_3$ and some of GdVO$_4$. The remaining work on GdVO$_4$ was undertaken using the $^{155}\text{EuPd}$ source reported by Prowse et al. [3].

2. Spectrum fitting

The spectra were fitted using a least-square analysis incorporating the Gauss-Newton method of numerical differentiation. Each line was assumed to have the same width and to be of the modified Lorentzian shape described below.

$$I(E) = I_0 \left[ \frac{\frac{1}{4}\Gamma^2}{(E - E_0)^2 + \frac{1}{4}\Gamma^2} \right] \left[ 1 + \frac{2\xi(E - E_0)}{\frac{1}{2}\Gamma} \right].$$  \hspace{1cm} (1)

The amplitude of the dispersion or interference term is significant for radiation of multipolarity EI and is given by

$$\xi = \left( \frac{\alpha\sigma_k}{6\pi\lambda} \right)^{1/2}.$$

Initial fitting used the thin-absorber approximation but is was found that this method was inadequate for the partially resolved spectra obtained with combined magnetic and quadrupole splittings. The line positions in the theoretical fits appeared to be accurate, however their intensities were not able to be determined well enough to give a satisfactory fit to the experimental spectra. It was apparent that the effect of absorber thickness was important and so each spectrum was refitted using the transmission integral described by Shenoy et al. [4]. The improvement to the fits of spectra obtained using the single crystal absorbers was significant and indicates the importance of this fitting procedure when partially resolved spectra are being analysed.

The high temperature quadrupole split spectra however could be fitted fairly well using the thin-absorber approximation, because of their two well resolved lines. This was also true for the powdered spectra recorded at 1.38 K even though they were only partially resolved. Fitting the powdered spectra using the transmission integral method gave an effective absorber thickness, $T_a$ less than that found for the single crystals.

Comparison of the fits obtained using the two methods can be seen in fig. 1.
showing the spectrum of a GdAlO$_3$ single crystal recorded along the magnetic ordering direction, (a - axis), at 1.38 K. The effect of a thick absorber is to even up the line intensities in unresolved spectra. The solid line (a) was obtained using the thin-absorber approximation with the overall line intensities adjusted to give the best fit to the outer part of the spectrum. The fitting is clearly unacceptable in the central part of the spectrum whereas curve (b), obtained using the transmission integral method with $T_a = 2.2$, represents a good fit to the entire spectrum. Curve (a) could also be obtained using this second method by constraining $T_a < 0.3$ as expected for the thin-absorber approximation.

3. Results

Spectra recorded at 4.2 K, using the 86.5 keV gamma transition, indicate that each sample is quadrupole split only, with the separation of the two lines being due predominantly to the splitting of the $^{155}$Gd ground state. Each peak is slightly broadened because of the small splitting of the 86.5 keV excited state. The spectra obtained from the powdered absorbers are symmetric and only the magnitude of the
EFG can be obtained. However, because the intensities of the $\Delta m = 0$ lines vary as a function of the angle between the direction of propagation of the gamma rays and the principal axis of the EFG, the spectra obtained from the single crystal absorbers are asymmetrical. The sign and magnitude of the EFG can therefore be determined.

Spectra recorded below the magnetic ordering temperature for each compound indicate the presence of a temperature-dependent hyperfine field at the Gd$^{3+}$ nucleus. A series of Mössbauer experiments recorded along the magnetic ordering direction at different temperatures down to 0.4 K has enabled us to determine the sublattice magnetization curves for GdVO$_4$ and GdAlO$_3$ [1,2]. Computer fitting of the spectra has given consistent values for the nuclear moments of the excited state, with the following ratios being determined

$$\frac{Q(86.5)}{Q(0)} = +0.10 \pm 0.02, \quad \frac{g(86.5)}{g(0)} = +1.217 \pm 0.005.$$  

The 105.3 keV gamma transition was used to record four spectra with the GdAlO$_3$ absorbers. This compound was used because of its high Debye temperature, $\theta_D = 214$ K, which increases the recoiless fraction and hence the observed effect. The ob-

Fig. 2. Computer fitted spectra of GdAlO$_3$ recorded above and below the Néel temperature using the 105 keV transition.
served absorption of 1% is three times larger than is generally obtained. GdAlO$_3$ has a large EFG and this has allowed us to obtain good resolution of spectra recorded above $T_N$. The four fitted spectra are shown in fig. 2. The two recorded above $T_N$ exhibit quadrupole splitting only, with the excited state quadrupole moment being similar in magnitude to that of the ground state. The other two spectra were recorded well below $T_N$ where magnetic and quadrupole interactions are present and the following ratios were obtained

$$\frac{Q(105.3)}{Q(0)} = +0.74 \pm 0.02, \quad \frac{g(105.3)}{g(0)} = -0.55 \pm 0.02.$$  

The isomer shifts of GdAlO$_3$ with respect to the SmPd$_3$ source are $0.498 \pm 0.002$ mm $\cdot$ s$^{-1}$ and $0.535 \pm 0.015$ mm $\cdot$ s$^{-1}$ for the 86.5 keV and 105.3 keV transitions respectively. This gives the ratio of the $\Delta \langle r^2 \rangle$ values as

$$\frac{\Delta \langle r^2 \rangle_{105}}{\Delta \langle r^2 \rangle_{86}} = 1.31 \pm 0.04.$$  

Similar spectra taken using an absorber of GdFeO$_3$ gave the ratio of the $\Delta \langle r^2 \rangle$ values as $1.28 \pm 0.06$. Both of these are within the range $1.38 \pm 0.20$ determined by Armon et al. [5].

The values of the interference parameter $2\xi$ were found to be $0.03 \pm 0.01$ for both transitions when corrected to zero absorber thickness. This agrees with the theoretical values of 0.035 and 0.026 for the 86.5 keV and 105 keV transitions respectively.

4. Discussion

There have been many determinations of the moments of the 86.5 keV and 105 keV levels and these are summarized in table 1. Considerable variation has been obtained due to three problems: (a) the ambiguity involved in fitting symmetrical spectra, (b) splitting $\leq$ observed linewidth and (c) poor statistics in many cases.

In addition, two anomalies have been observed in the 105 keV spectra. Blumberg et al. [6] obtained values of the ground state quadrupole splitting from spectra taken with the two transitions which differed by 20%, although Armon et al. [5] did not observe the same problem. Blumberg et al. also observed linewidths for the 105 keV transition in the range 0.64 to 1.55 mm $\cdot$ s$^{-1}$ compared with a natural linewidth $2\Gamma_0$ of $2.22 \pm 0.03$ mm $\cdot$ s$^{-1}$. Armon et al. discounted this result suggesting that it was due to pile-up effects causing “antipeaks” in the 105 keV count rate at velocities where there was an 86.5 keV resonance. However, their observed linewidth of $2.25 \pm 0.15$ mm $\cdot$ s$^{-1}$ is still surprisingly narrow when taken in conjunction with their 86.5 keV linewidths of 0.77 mm $\cdot$ s$^{-1}$ compared to a natural $2\Gamma_0$ of $0.499 \pm 0.005$ mm $\cdot$ s$^{-1}$.

In choosing compounds with combined magnetic and quadrupole splittings, we
Table 1
Parameters reported for the moments of 86.5 keV and 105 keV levels of $^{155}$Gd (The number in parentheses indicates the error in the last figure.)

<table>
<thead>
<tr>
<th>$Q(86.5)$</th>
<th>$Q(105)$</th>
<th>$g(86.5)$</th>
<th>$g(105)$</th>
<th>Method</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Q(0)$</td>
<td>$Q(0)$</td>
<td>$g(0)$</td>
<td>$g(0)$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Q(0) = 1.59 (16) b</td>
<td></td>
<td></td>
<td></td>
<td>Atomic Beam</td>
<td>[i]</td>
</tr>
<tr>
<td>0.20 (10)</td>
<td>0.9 (1)</td>
<td>1.23 (12)</td>
<td>-0.5 (1) or</td>
<td>ME</td>
<td>[ii]</td>
</tr>
<tr>
<td>-0.07 (21)</td>
<td>1.36 (4)</td>
<td>1.22 (1)</td>
<td>1.5 (2)</td>
<td>ME</td>
<td>[iii]</td>
</tr>
<tr>
<td>0.346 (12)</td>
<td></td>
<td>2.01 (5) or</td>
<td></td>
<td>ME</td>
<td>[iv]</td>
</tr>
<tr>
<td>0.12 (1)</td>
<td>0.087 (6)</td>
<td>1.235 (8)</td>
<td>-0.55 (5)</td>
<td>DPAC</td>
<td>[v]</td>
</tr>
<tr>
<td>&lt;0.48</td>
<td>1.34 (6)</td>
<td>0.74 (2)</td>
<td></td>
<td>ME</td>
<td>[vi]</td>
</tr>
<tr>
<td>-0.090 (29)</td>
<td></td>
<td>1.20 (1)</td>
<td></td>
<td>ME</td>
<td>[vii]</td>
</tr>
<tr>
<td>0.35 (6)</td>
<td>0.085 (5)</td>
<td>1.217 (5)</td>
<td>-0.55 (2)</td>
<td>ME</td>
<td>[viii]</td>
</tr>
<tr>
<td>0.10 (2)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>[ix]</td>
</tr>
</tbody>
</table>

ME = Mössbauer Effect
DPAC = Differential Perturbed Angular Correlation

References to table 1

[iii] Ref. [6].
[viii] Ref. [5].
[xi] Ref. [7].
[xiii] This work.
have ensured that our spectra are not all symmetrical and in the case of the 105 keV spectra we used a Ge(Li) detector operating at low count rates (<$10^4$ total counts per second into the detector, 1600 counts/sec into the multi-channel analyzer) to try and eliminate any possibilities of pile-up or gamma resolution problems.

Use of the transmission integral made a marked difference to the fitting and this approach was not reported by any of the previous workers. We believe that this is the first time that the transmission integral has been necessary in order to obtain an acceptable fit to the data (see fig. 1) rather than the more usual occurrence where the thin absorber approximation gives an acceptable fit, but with incorrect parameters. These observations suggest that previous measurements on this and many other nuclides have error limits which are seriously underestimated.

The parameters obtained for the 86.5 keV level are consistent over 30 spectra and representative spectra can be seen in refs. [1,2]. The values are in close agreement with the more recent observations except for the anomalous value for the quadrupole ratio obtained by Bowden et al. [7] which is believed to be due to the use of a thick absorber.

The ratio of g(105)/g(0) is in agreement with the second of the ambiguous set of Armon et al. [5], however our value of $Q(105)/Q(0)$ is at variance with both previous workers. Our fitting was obtained using the same values of the ground state splittings as in the 86.5 keV spectra and acceptable fits could not be obtained with any other values. The value is in agreement with theoretical predictions based on the Nilsson model [5,6].

The linewidths obtained in the 105 keV spectra varied from 1.94 to 2.10 mm $\cdot$ s$^{-1}$ before correcting for absorber thickness and are thus less than the natural $2\Gamma_0$. The correction for absorber thickness is 0.3 mm $\cdot$ s$^{-1}$ which is expected to be greater than the possible error in the linewidth due to deviations from pure Lorentzian shape or due to uncertainty in the baseline. The lifetime of this level has been determined by several groups [8] with good agreement and seems unlikely to be in serious error. We do not believe that the "pile-up" explanation put forward by Armon et al. is applicable to our results and in addition the "anti-peaks" are rarely in the correct position. Consequently, we consider this still to be an unsolved problem with the after-effects of nuclear decay being a credible explanation as suggested by Blumberg et al. [6] but, if so, one would expect this effect to be observed in many more transitions.

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