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Measurements of Perturbed Angular Correlations in Compounds of ^{57}Co

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Measurements of the perturbed angular correlation have been made for the 3/2+ excited state of ^{57}Fe in several compounds of *Co*. The unperturbed *A*, value of the angular correlation for the *122* keV – *14.4* keV cascade in ^{57}Fe has been determined in an aqueous solution of $CoCl_2$ to be $A_{22} = 0.017 \pm 0.009$. Quadrupole interaction frequencies of *1.6* MHz, *13* MHz and *40* MHz were determined in polycrystalline sources of $Co(OH)_3$, $CoCl_2 \cdot 6H_2O$ and $CoSO_4 \cdot 7H_2O$, respectively. Limitations and possibilities of differential measurements for cases of low frequencies are discussed.

Medidas da correlação angular perturbada do estado excitado 3/2+ do ^{57}Fe em vários compostos do *Co* foram efetuadas. O coeficiente da correlação angular não perturbada foi determinado como sendo $A_{22} = 0.017 \pm 0.009$ para a cascata 122 keV – *14.4* keV no ^{57}Fe em uma solução aquosa de $CoCl_2$. Freqüências de interação quadripolar de *1.6* MHz, 13 MHz e *40* MHz foram obtidas em fontes policristalinas de $Co(OH)_3$, $CoCl_2 \cdot 6H_2O$ e $CoSO_4 \cdot 7H_2O$, respectivamente. As limitações e as possibilidades do método diferencial para os casos onde freqüências baixas estão envolvidas são discutidas.

1. Introduction

The time differential perturbed angular correlation method has been used successfully in various cases for the study of nuclear quadrupole interactions in solids[1-4]. In general, however, its use has been limited to a few especially suitable nuclei for various technical reasons. One of the most important limitations up to the present has been that of obtaining good time resolution for cascades involving low energy gamma-rays.

In the following, we report on a set of preliminary experiments designed to test the feasibility of studying hyperfine interactions for the 14.4 keV 3/2+

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excited state of ^{57}Fe with a half life of 97.7 x 10^{-9} seconds through the measurement of differential perturbed angular correlation of the 122 keV-14.4 keV cascade in ^{57}Fe following the decay of ^{57}Co. The study of this state by perturbed angular correlation techniques is particularly interesting as a complement to studies made on the same state using the Mössbauer effect.

In the series of experiments reported here, we have studied sources of $CoCl_2$ in aqueous solution and polycrystalline sources of $CoCl_2 \cdot 6H_2O$, $CoSO_4 \cdot 7H_2O$ and $Co(OH)_3$. The results shown below indicate that the method can be applied successfuly in the investigation of quadrupole interactions in cobalt compounds.

2. Theoretical Background

The angular correlation function describing the coincidence counting rate of two successive radiations emitted in relative directions specified by an angle 8, and separated by a time interval t during which a perturbation acts on the intermediate state, can be written for the case that the source is a liquid or a polycrystalline powder as[1]

$$W(\theta, t) = \sum_k A_k(1)\, A_k(2)\, G_{kk}(t)\, P_k(\cos\theta), \tag{1}$$

where $A_k(1)$ and $A_k(2)$ are the usual angular correlation coefficients and $G_{kk}(t)$ is the perturbation factor. In the present case only the values $k = 0$ and $k = 2$ contribute, and Eq. (1) can be written as

$$W(\theta, t) = 1 + A_2\, G_2(t)\, P_2(\cos\theta), \tag{1a}$$

where for static quadrupole interactions and our spin sequence,

$$G_2(t) = 0.2 + 0.8 \cos \omega_0 t. \tag{2}$$

The fundamental frequency ω_0 is given by $\omega_0 = 6\omega_Q$ where $\hbar\omega_Q = \frac{-eQV_{zz}}{4I(2I-1)}$ corresponding to the usual definitions in ihe literature'.

An expression for the perturbation factor including simultaneous static and time-dependent quadrupole interactions is given in Ref. 5. Effects on the perturbation factor due to a Gaussian distribution of frequencies arising from inhomogeneity of the electric field gradient are described in Ref. 6.

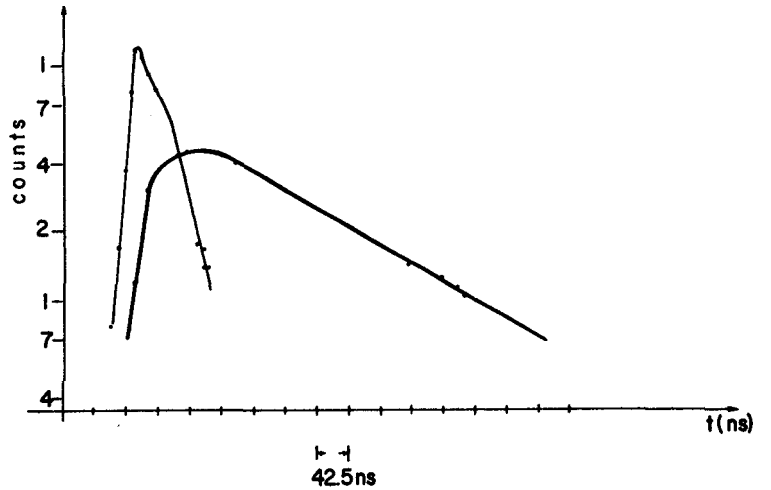

Fig. 1 – Time resolution of 30 nsec and life-time for 133-14 keV gamma rays in ^{57}Fe.

3. Apparatus

The differential measurements were performed with a coincidence system[7] consisting basically of a time-to-pulse-height converter and a 256 channel analyzer. The 122 keV gamma-ray was detected in a 1" × 3/44 $NaI(Tl)$

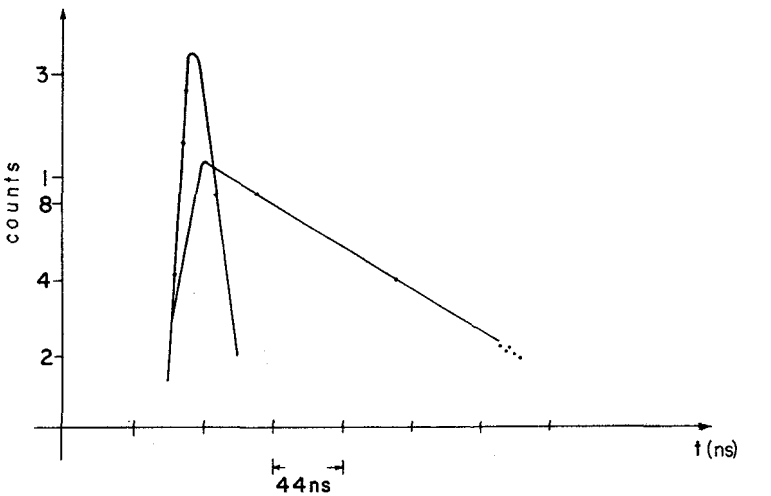

Fig. 2 – Time resolution of 17.6 nsec and life-time for 133-14 keV gamma rays in ^{57}Fe.

scintillator mounted on a RCA 6810*A* photsmultiplier while the 14 keV gamma was detected in a *2.5* mm x *3/44* $NaI(Tl)$ crystal mounted on an *EMI 6097* photomultiplier. Two experimental arrangements were used. In the first, a time pick-off unit was used to generate the fast signal for the low energy radiation, whereas normal leading edge timing was used for the high energy gamma. A time resolution of *30 n*sec at the half-maximum of rhe prompt curve was measured using a ^{22}Na source with the energy settings for the ^{57}Fe cascade. This result is shown in Fig. 1. In the second arrangement a fast amplifier and a tunnel diode discriminator were used instead of the time pick-off unit. With this system, a time resolution of *17.6 n*sec was obtained. The result is shown in Fig. 2.

Time spectra were accumulated in the multichannel analyzer for relative angles between detectors of 90°, 180° and *270".*

4. Results and Conclusions

The results of the differential measurements made on $CoCl_2$ (liquid solution) and on the polycrystalline sources $CoCl_2 \cdot 6H_2O$ and $CoSO_4 \cdot 7H_2O$ are shown in Figs. *3,* 4 and *5* and were obtained with a resolution of 22 r 30 *n*sec. The results of the differential measurements on polycrys-

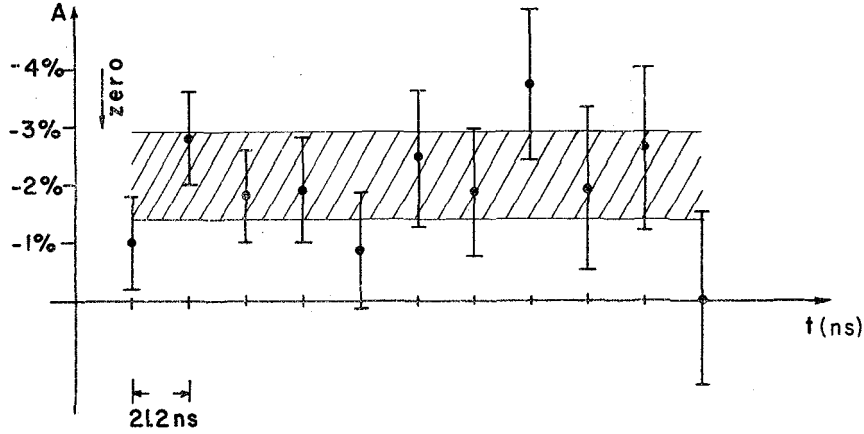

Fig. 3 – Anisotropy in function of time for the 133-14 keV cascade of ^{57}Fe in $CoCl_2$ (liquid).

tailine sources of $Co(OH)_3$ are shown in **Fig.** 6 and were obtained with the experimental system with **22** $\cong$ 17.6 nsec. **All** curves show the **behaviour** of the anisotropy

$$A(t) = W(180, t)/W(90, t) - 1.$$

In all cases we used carrier free sources.

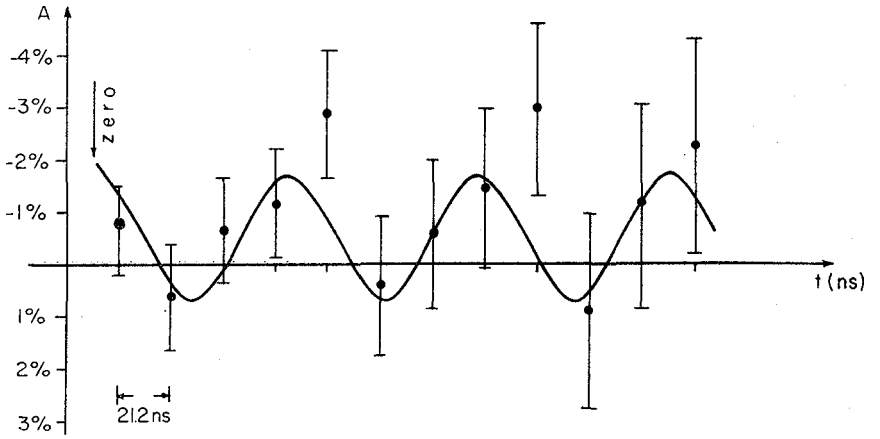

Fig. **4** – Anisotropy **in** function of time for the **133-14 keV cascade** of ^{57}Fe **in** $CoCl_2 \cdot 6H_2O$.

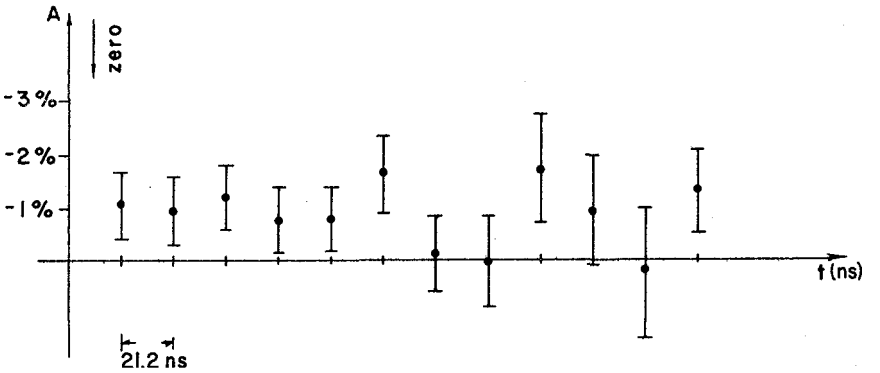

Fig. 5 – Anisotropy in function of time for the **133-14 keV cascade** of ^{57}Fe in $CoSO_4 \cdot 7H_2O$.

In most cases, it is expected that the electric field gradients in liquid solutions average out due to the Brownian motion, leaving the angular correlation unperturbed. The behaviour of the anisotropy for aqueous solution of $CoCl_2$ indicates that this is the case. Since $A_2 = 0$, we can deduce a a value for $A_4 = 0.017 \pm 0.009$ which is in agreement with the value of $A_4 = 0.024 \pm 0.003$ reported by T. Lindqvist and E. Heer[8] deduced from integral measurements. However, G. Chandra[g] has reported $A_4 = 0.042 \pm 0.016$ from a measurement of the integral angular correlation in a single crystal of Co-metal. This discrepancy is probably a result of effects of absorption and scattering of the radiations in the source.

The polycrystalline source of $CoCl_2 \cdot 6H_2O$ (Fig. 4) shows a clear oscillatory behaviour of the anisotropy indicating the presence of a static quadrupole interaction. Comparison with a theoretical curve calculated for a fundamental frequency $\omega_0 \simeq 13$ MHz shows good agreement. Our result can be compared with that obtained by Ingalls and Pasqualli[10] deduced from Mossbauer effect data as $\omega_0 \simeq 17$ MHz.

The polycrystalline source $Co(SO_4) \cdot 7H_2O$ (Fig. 5) shows a flat but non-zero anisotropy indicating that our resolution time was not sufficiently small to determine the quadrupole interaction. In fact, from Mossbauer effect data[11] it is known that $Fe(SO_4) \cdot 7H_2O$ has a quadrupole interaction $\omega_0 = 40$ MHz.

The polycrystalline $Co(OH)_3$ source (Fig. 6) shows an oscillatory behaviour for the anisotropy indicating however a very weak quadrupole interaction

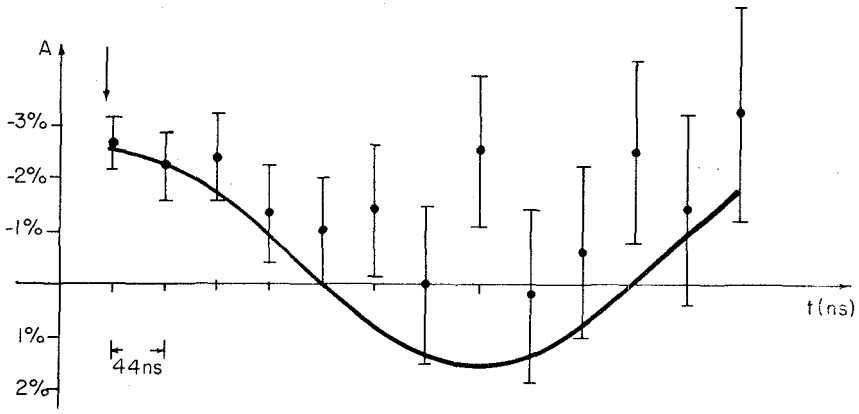

Fig. 6 – Anisotropy in function of time for the 133-14 keV cascade of ^{57}Fe in $Co(OH)_3$.

with $\omega_Q \simeq 1.6$ MHz. Such coupling corresponds to a Mossbauer splitting of 0.12 mm/s, or about 70% of the natural line width. In this case, the perturbed angular correlation measurement can detect the weak quadrupole interaction considerably more accurately than the Mossbauer effect technique.

The results obtained are summarized in Table 1. We note that, in interpreting the results, we have assumed that none of these sources shows magnetic hyperfine structure. In the cases of $CoCl_2$ and $CoSO_4$ the compounds are known to be diamagnetic at room temperature thus assuring absence of magnetic fields. The interpretation is also consistent with what should be expected for $Co(OH)_3$.

Source	Iron Electronic Configuration	Quadrupole Constant	Relaxation
$CoCl_2 \cdot 6H_2O$	d^6	$\cong$ 13 MHz	no
$Co(SO_4) \cdot 7H_2O$	d^6	30 MHz	no
$Co(OH)_3$	d^5	1.6 MHz	no

Table 1.

We conclude from these results that differential measurements of perturbed angular correlations in the decay of ^{57}Co to ^{57}Fe can be a powerful and very useful adjunct to the study of crystalline and molecular properties of compounds containing cobalt. In comparison to studies using cobalt compounds as sources in the Mossbauer effect, they can be especially interesting in cases where a very low quadrupole frequency makes the determination of the quadrupole separation from the Mossbauer effect difficult or impossible, or in cases where exceptionally low concentration of cobalt as an impurity is important.

Analysis of the present results indicate that in order to continue measurements of this type, it will be important both to decrease the statistical uncertainties as well as to improve the resolving time. A system of multiple detectors and significantly better time resolution is presently being tested in our laboratory to be used in further studies.

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