Determination of a Static Hyperfine Interaction Following Recoil into Vacuum

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By means of using a plunger time-differential technique, spin precessions from a static free hyperfine interaction have been observed for low-energy ^{152}Sm nuclei recoiling into vacuum.

I. Introduction

Perturbed angular correlation techniques are being widely used as tools for the determination of static moments of excited nuclear states. If, however, these moments are known, then information on the pertinent fields is available, since generally the deduced quantity is the interaction frequency. In many cases only average or effective values, due to various relaxation mechanisms, can be determined.

Of particular value are those experiments where the interaction is free from any relaxation. In such cases one can relate unattenuated fields with nuclear parameters. Hyperfine magnetic or electric quadrupole coupling is especially desirable because of the magnitude of these couplings.

This article presents the results of an attempt to perform a measurement in relaxation-free conditions: The observation of nuclear spin precession under fields generated by atomic electrons in their ground state, in free ions.

II. The Hyperfine Interaction

The hyperfine interaction couples the atomic spin $\boldsymbol{J}$ with the nuclear spin $\boldsymbol{I}$. In a free ion the magnetic coupling Hamiltonian K is given [1] by

$$K = (\mu \overline{H(0)_J}/I\,J)\,\boldsymbol{I}\cdot\boldsymbol{J}\,,$$

while for an electric quadrupole interaction

$$K = \frac{e\,Q\,\overline{V_{zz}(0)}_J}{8\,I\,J(2\,J-1)\,(2\,I-1)}\,[3\,(\boldsymbol{I}\cdot\boldsymbol{J})^2 + 3/2\,\boldsymbol{I}\cdot\boldsymbol{J} - \boldsymbol{I}^2\,\boldsymbol{J}^2]\,.$$

μ and Q are the magnetic and quadrupole moments of the nuclear state, $H(0)$ and $V_{zz}(0)$ are respectively the magnetic field and the electric field gradient at the nucleus. This interaction affects the nuclear alignment by causing its precession. The changes in the m-state population perturb the original angular distribution, of emitted gamma radiation.

A typical angular distribution of the form

$$W(\Theta) = 1 + \sum_{\text{even}\,k} a_k\,G_k(t)\,P_k(\cos\Theta)$$

will be modified through $G_k(t)$, depending on the hyperfine interaction.

In a free ion, where the atomic spins $\boldsymbol{J}$ are randomly-oriented, one gets in a static case [2]:

$$G_k(t) = \frac{1}{2\,J+1}\sum_{F,F'} \cdot\,(2\,F+1)\,(2\,F'+1)\begin{Bmatrix} F & F' & k \\ I & I & J \end{Bmatrix}^2 e^{-i\omega_{FF'}\,t}$$

where

$$\boldsymbol{F} = \boldsymbol{I} + \boldsymbol{J}$$

and

$$h\,\omega_{FF'} = E_F - E_{F'}\,.$$

Integral-attenuation measurements of angular distribution of gamma rays, following recoil into vacuum of Coulomb excited rare-earth nuclei, may be either time-dependent or static with either magnetic or electric perturbations [3]. These time-dependent and static perturbations can also occur simultaneously. The time-dependent effects originate in fast atomic transitions that change both the atomic spins $\boldsymbol{J}$ and their associated fields. If, however, an excited atomic (or ionic) system can reach its ground state in a time very short compared with the nuclear mean life τ, one may observe a static perturbation.

Time-differential measurements of $G_k(t)$ have a trivial superiority over time-integral techniques, if they can be accomplished at all. It is the aim of the present work to try and determine in a time-differential method the temporal behaviour of $G_k(t)$. The idea is based on the fact that in this way a small static component can be filtered out from a composition having mostly a time-dependent content.

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III. The Time-differential Method

In order to detect attenuation-coefficients $G_k(t)$ or integral attenuation coefficients

$$\overline{G_k(t)} = \frac{1}{\tau} \int_0^\infty G_k(t)\, \mathrm{d}t ,$$

highly anisotropic distributions are necessary. For an even-even nucleus in Coulomb excitation, a known distribution of gamma rays is obtained when working in coincidence with the back scattered particles, namely:

$$W(\Theta, t) = 1 + A_2\, G_2(t)\, P_2(\cos\Theta) + A_4\, G_4(t)\, P_4(\cos\Theta) ,$$
$$A_2 = 5/7 , \qquad A_4 = -12/7 .$$

If a silver plunger is situated at a distance d from a target, then the forward recoiling nuclei of velocity v will hit the plunger at a moment $t' = d/v$. Silver is known to be a hyperfine-free medium which will preserve the nuclear alignment (Ref. [4]). One observes in this case the time dependent angular distribution:

$$I(t', \Theta) = \frac{1}{\tau} \int_0^{t'} \Big[1 + \sum_{k=2,4} A_k\, G_k(t)\, P_k(\cos\Theta) \Big] \cdot e^{-t/\tau}\, \mathrm{d}t + \frac{1}{\tau} \int_{t'}^{\infty} \Big[1 + \sum_{k=2,4} A_k\, G_k(t')\, P_k(\cos\Theta) \Big] \cdot e^{-t/\tau}\, \mathrm{d}t . \qquad (2)$$

We start with a specific simple case. It was assumed that in the case of ^{152}Sm, for which a time-dependent magnetic interaction was found (Ref. [3]) a low-velocity recoil into vacuum would reduce the magnitude of the time-dependent interaction, allowing a possible static component to be observed.

Sm has the following configurations for the neutral, singly and doubly ionized ground states [5]

Sm^{0}	Sm^{+1}	Sm^{+2}
$4f^6\, 6s^2$	$4f^6\, 6s$	$4\, f^6$
$^7F_0$	$^8F_{1/2}$	$^7F_0$

Working with low-velocity recoil one can avoid charge states higher than +2. Under such conditions one hopes to obtain the $J = 0$ of the Sm^0 and Sm^{+2} which do not affect $W(\Theta, t)$, and $J = 1/2$ for Sm^{+1}. We expect then a pure magnetic coupling. The above expressions (1) for $G_k(t)$ give for $J = 1/2$ and $I = 2$:

$$G_2(t) = \frac{19}{25} + \frac{6}{25} \cos \frac{5}{2} \frac{a\, t}{\hbar}$$
$$G_4(t) = \frac{1}{5} + \frac{4}{5} \cos \frac{5}{2} \frac{a\, t}{\hbar} , \qquad (3)$$

where the magnetic hyperfine structure constant is given by:

$$a = \mu\, \overline{H(0)}_J / I\, J .$$

Figure 1 represents two calculated time-dependent intensities of the radiation in a plunger experiment. It is calculated for $\Theta = 45°$ and $\Theta = 90°$ with respect to the beam, in coincidence with back-scattered particles. It was assumed for this calculation that all nuclei are subjected to the static hyperfine field and that the experimental set-up was capable of infinitely narrow time resolution.

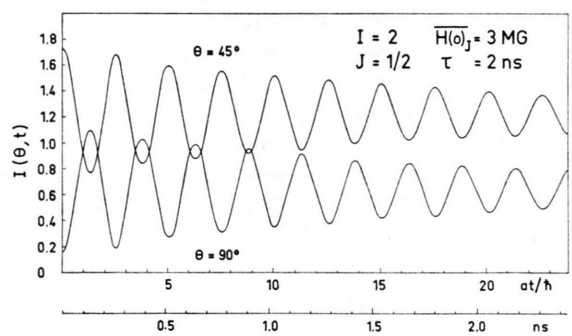

Fig. 1. Theoretical curves for a back-scattered coincidence experiment (see text). Parameters were chosen corresponding to ^{152}Sm. Future measurements with intensity ratios like $I(45°, t)/I(90°, t)$ should facilitate the detection of small amounts of static perturbation.

IV. Measurements and Results

While the method obviously calls for a coincidence arrangement, we are not able at the moment to perform such an experiment. We have therefore limited ourselves to a considerably reduced anisotropy, using the 6 MeV α beam of a Strasbourg Van de Graaff machine. In a Coulomb excitation with 6 MeV α-projectiles, the maximum recoil energy of an excited samarium nucleus is 600 keV, corresponding to $\beta = v/c = 0.29\%$. It is unlikely that a considerable amount of ionization states higher than the 2^+ can be obtained [6].

Without coincidence arrangement the gamma radiation with respect to the beam is given by:

$$W(\Theta,t)=1+\sum_{k=2,4} a_k(\xi)\,A_k\,G_k(t)\,P_k(\cos\Theta)$$

where $a_k(\xi)$ and A_k are Coulomb excitation particle parameter and γ-γ angular correlation coefficients respectively (Ref. [7]).

For our conditions:

$$A_2=0.36,\quad a_2\sim 0.35,\quad A_4=1.14,\quad a_4\sim 0.02.$$

With a target of ^{152}Sm of about 30 μg/cm^2, using Coulomb excitation theory and kinematics as well as the ϱ and ε function of LINDHARD and SCHARFF (Ref. [8]), we estimated the emitted angular distribution of the recoiling Sm ions. A roughly triangular ditribution is obtained, peaked at around 45°.

What was anticipated then was a static component due to the $J=1/2$ ground state of Sm^{+1}. This would lead to $G_2(t)$ and $G_4(t)$, (3), which would oscillate with the single frequency $\omega=5\,a/2\,\hbar$. We also assumed that this was the single static component. Other time-dependent processes, if they did exist, would affect $I(\Theta,t)$ (2), in a smooth monotonic way as predicted by ABRAGAM and POUND [9]. The distribution of the recoiling Sm ions had to be weighted by the relative cross section for producing a single ionized atom. Such phenomena are not well known at present and this made such a calculation impossible. If this *weighted distribution* were to be peaked in any direction, the observation of ω would be possible.

The intensity $I(\Theta,t)$ was recorded for a few Θ as a function of the plunger distance. Figure 2 represents the run at $\Theta=28°$. The initial behavior resembles the time-dependent attenuation as measured in a time-differential technique by a group in FREIBURG [10]. Roughly, an exponential behaviour can be assigned to this part of $I(\Theta,t)$ with a decay constant of $\lambda^{-1}\sim 100$ ps. Such values were usually measured for rare-earth ions [10, 11]. Following the decreasing behaviour we observe an oscillating time-dependence. We relate this to the nuclear spin precession. In such conditions the amplitude of the oscillation has no meaning since the factors that dictate it are not known, as, for example the fraction of the singly ionized Sm in their ground state. Nevertheless it is the *temporal* dependence that contains the interaction strength. If the ^{152}Sm nuclei emitted from the target around 45° are those that play the dominant role, with the available g-factor for the 2^+ state [12], one deduces a field of $\overline{H(0)}\sim 3\,MG$. This value is close to the value found for the Sm^{+3} ion [13].

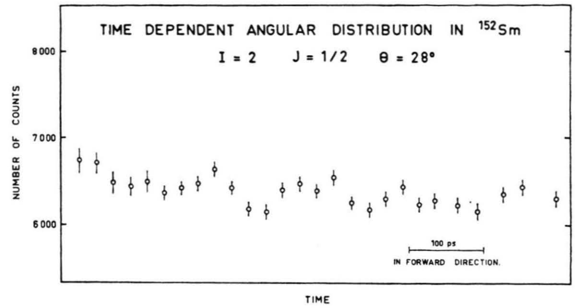

Fig. 2. Observation of the time dependence of the angular distribution at $\Theta=28°$ for ^{152}Sm. The initial behaviour is roughly that of an exponent characterised by $\lambda^{-1}\sim 100$ ps. For emitted Sm ions at 45° the oscillations correspond to a magnetic field at the nucleus of 3 MG.

The system was also run with a thick target where the velocity spread was expected to obscure completely the oscillations. This was verified experimentally.

In conclusion, the phase-space coordinates in the present experiment, of the emitted Sm nuclei, tend to distort the details of the spin rotation following the $\boldsymbol{I}\cdot\boldsymbol{J}$ coupling to a resultant $\boldsymbol{F}$. Moreover, further remote oscillations in time will be more and more obscured. However, this first attempt points at a very promising way for determination of fields at the nucleus, ratios of moments, etc. As mentioned, it seems that highly aligned nuclear systems followed by narrowly defined velocities and directions, are especially suited for such experiments. This is particularly tempting for cases where Coulomb Excitation with back-scattering projectiles in coincidence with the de-exciting gamma radiation are performed. The pattern of $G_k(t)$ becomes more complicated with increasing J, but this introduces no difficulty in the principle of the experiment *.

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* *Note added in proof:* More realistic calculations, taking into account the weighted contribution of Sm nuclei recoiling in all directions, yield a field of 5 MG. These calculations as well as new experimental data will be presented elsewhere.
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Mössbauer-Effekt am Fe^{57} in Additions- und Substitutionsverbindungen des Natrium-Nitrosylprussiates *

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Mössbauer-Effect in Fe^{57} of Addition and Substitution Compounds of Sodium Nitroprusside

Investigating 21 iron complex compounds of the type $[Fe(CN)_5NOL]^{n-}$ or $[Fe(CN)_5L]^{3-}$ it was found that isomeric shift, quadrupol splitting, and Goldanskii effect depend on the purity of the compound. Varying the amount of impurity discrepancies between formerly published data could be explained. The Mössbauer spectra of three compounds showed line widths differing from the theoretical value only by a few per cent. This indicated that a very high purity had been achieved. In all cases information about the molecular structure and the bond strength between the central Fe-atom and its ligands was obtained.

1. Einleitung

Der Mössbauer-Effekt stellt eine experimentelle Methode zur Untersuchung der chemischen Bindung des Leuchtkerns dar: Die Isomerieverschiebung gibt Aufschluß über die Elektronendichte am Kernort, die durch die Bindungsstärke des Leuchtkerns mit seiner Umgebung verändert wird. Die Quadrupolaufspaltung hängt von der Unsymmetrie des elektrischen Feldes ab, das von der Elektronenhülle des Leuchtkerns und von den Bindungspartnern erzeugt wird. Der GOLDANSKII-Effekt [1] polykristalliner Verbindungen zeigt eine vorhandene Kristallanisotropie an, die gestört wird, sobald Verunreinigungen die Ausbildung eines regelmäßigen Kristallgitters verhindern. Die genauesten Mössbauer-Messungen lassen sich mit Fe^{57} als Leuchtkern ausführen, da dieser Kern eine kleine Breite der Resonanzlinie und eine große Resonanzabsorption aufweist. Zahlreiche Messungen wurden an Fe-Salzen und Fe-Komplexverbindungen vorgenommen [2]. Die Meßergebnisse der einzelnen Autoren fallen unterschiedlich aus, wobei die Abweichungen größer sind als die angegebenen Meßfehler.

In der vorliegenden Arbeit werden Mössbauer-Messungen ausgeführt an Komplexverbindungen des Eisens, die sich durch Additions- oder Substitutionsreaktionen vom Natrium-Nitrosylprussiat herleiten. Sie werden allgemein als Prussiate bezeichnet. Obwohl ein Teil der betrachteten Reaktionen seit langem bekannt ist, konnten in einigen Fällen die festen Verbindungen zum ersten Male in reiner Form gewonnen und untersucht werden. Es gelang, aus den Mössbauer-Spektren Rückschlüsse zu ziehen auf die Strukturen und die Bindungsstärken der betrachteten Verbindungen.

2. Meßanordnung

Zur Ausführung der Mössbauer-Experimente wurde eine Bewegungsapparatur gebaut, die auf dem Prinzip

* D 7 (gekürzt).