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ELECTRON CAPTURE AFTER-EFFECTS IN Cd^{111}

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Abstract: The integral and differential angular correlation of the 173-247 keV gamma cascade in Cd^{111} was investigated at different temperatures in In_2O_3 polycrystalline sources. At 280° C a static electric quadrupole interaction is present with an interaction frequency $\omega_0 = 180$ MHz. At room temperature the static perturbation is mixed with a time dependent perturbation probably due to the ionized states in which Cd atoms are left after electron capture in In^{111}.

1. Introduction

The angular correlation of two successive nuclear transitions may be perturbed by external fields in different ways. Static magnetic and electric quadrupole perturbations have been carefully studied and a summary of theoretical [1]) and experimental [2]) results may be found in the literature. The experimental method of differential angular correlation has turned out to be very important for measurements of static perturbations, not only for the determination of the interaction frequency, but also for more detailed information about the interaction as time dependent perturbations in liquids [3]), or asymmetries in the electric field gradient in crystals, or variations of the electric field gradients at the different sites of the nuclei in an imperfect crystal lattice [4]).

Another type of perturbation in angular correlation is resulting from the ionized states in which the atoms are left after electron capture or internal conversion [5]). In both cases one electron is removed from the inner shells after which the created hole is filled by electrons from outer shells by means of radiative and Auger transitions. The last process is dominant in the outer shells, and as each Auger process creates a new hole, the final ionization of the atom may be multiple. A summary of these processes and the resulting ionization was given recently by Carlson [6]). Experiments performed in free atoms of Xe^{131}, which has a Z value close to that of Cd, show that due to internal conversion, the atom becomes ionized with a maximum probability of loosing 8 electrons [7]). This value will probably change for an atom in a crystal, considering that the wave functions of the outer shells are distorted, thus changing the transition probabilities. The electron hole moves from the inner to the outer shells in a time shorter than 10^{-13}s. The lifetime of the final ionized state depends on the characteristics of the surrounding medium.

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In metals free electrons from the conduction band are available, and the filling process is rather fast, probably shorter than 10^{-12} s, thus the perturbation induced by the ionized atoms in the angular correlation is very small. In ionic crystals electrons may move only because of thermal excitation or impurity conduction and the lifetime of the ionized state in this case is longer, and thus will have a stronger influence on the angular correlation.

The after-effects from electron capture and internal conversion have been studied by means of integral as well as differential angular correlation methods [2,8-10]. The main problem is to demonstrate that for a certain nuclear transition cascade a time dependent perturbation is present in one kind of environment, whereas the same perturbation decreases or vanishes when the character of the environment is changed. Earlier experiments have considered changes in environment due to different chemical compounds. In this experiment, however, it was chosen to change the character of the environment by changing the temperature in a polycrystalline structure.

2. Experimental Arrangements

The experiments were carried out with the well-known 173-247 gamma cascade in Cd^{111} following the decay from In^{111} by means of electron capture. The half-life of the intermediate state of the cascade is 85 ns and the half-life of the upper state is shorter than 1 ns [11,12]. The active In was produced by a (p, n) reaction on natural metallic Cd and was chemically separated by disolving the target in nitric acid and 4.5 N hydrobromic acid. The In solution was evaporated to dryness and then diluted with a sulphate solution, from which the active indium was electroplated onto an indium metal rod. The deposit was removed from the rod and finally oxidized and ground several times. The active In was thus in the form of In_2O_3 polycrystalline powder. According to crystallographic data the In_2O_3 structure is stable up to 850°C, at which temperature it decomposes to lower oxidation states. In_2O_3 is a cubic crystal and corresponds to the T_h^7 type (Schoenflies notation), with 16 molecules per elementary cell and with a lattice constant [13]) of 10.12 Å. The crystal does not contain two or more n-fold rotation axes with $n > 2$. Thus an electric field gradient may be present producing a static electric quadrupole interaction. Both the differential and integral angular correlation of the 173-247 keV gamma cascade were measured with the equipment described in ref. [3]). The total time range in the differential measurement was 90 ns and the full width at half maximum of the prompt peak (taken with Na^{22} with the energy settings for the Cd^{111} cascade) was 4.3 ns. NaI(Tl) crystals and RCA 6342A photomultipliers were used.

3. Experimental Results

The time dependent angular correlation was measured at 90° and 180° (A_4 terms are negligible in the first approximation for this cascade). Considering the angular

correlation expressed as:

$$W(\vartheta t) = 1 + A_2 G_2(t) P_2(\cos \vartheta),$$

$G_2(t)$ was evaluated together with the statistical errors with the help of an IBM 1620 computer.

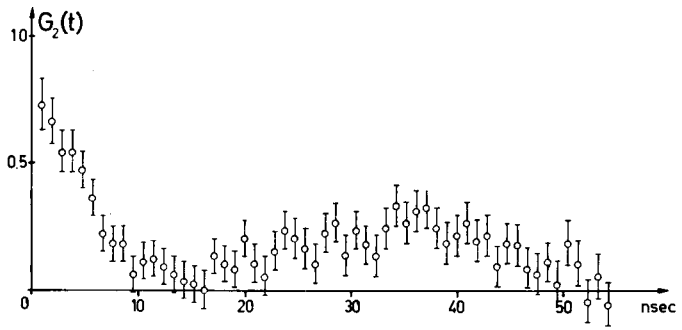

Fig. 1. Experimental value of $G_2(t)$ in In_2O_3 polycrystalline source at room temperature.

The first experiments were carried out at room temperature. A typical result from one of the measurements is shown in fig. 1. All the other measurements agree with this one within the statistical errors. The apparently non-periodic behaviour of $G_2(t)$ may be explained in one of the following ways:

(a) a time dependent perturbation produces a fast wipe out of the correlation;

(b) a static quadrupole interaction is present, but with a wide frequency distribution [4]) as a result of a non-uniform electric field gradient;

(c) a static quadrupole perturbation with high interaction frequency is present which cannot be resolved by the equipment ($\omega_0 \gtrsim 500$ MHz).

A second experiment was carried out at room temperature and at 280°C. It was not desirable to go to higher temperatures in order to avoid phase changes in the crystal structure. Further, the relative variations of the lattice constants should be small [14]). In order to control that no phase changes or other chemical changes should appear the experiment was actually carried out in the following sequence: $\vartheta = 90°$ (280°C), 180°(20°C), 180°(280°C), 90°(20°C), etc. The data were added separately and $G_2(t)$ was calculated for both temperatures. The results of the room temperature data as well as the 280°C data are shown in fig. 2 where the full line represents the result of a theoretical quadrupole interaction of an intermediate spin $I = \frac{5}{2}$, a Gaussian frequency spread [4]) of 10%, the correction for finite time resolution being included. The frequency of the interaction obtained from fig. 2 for 280°C is:

$$\omega_0 = 180 \pm 14 \text{ MHz}.$$

From this result the possibility of a very high interaction frequency must be excluded because it is very unlikely that the frequency could change from more than 500 MHz to 180 MHz in the actual temperature range. Integral correlation measurements

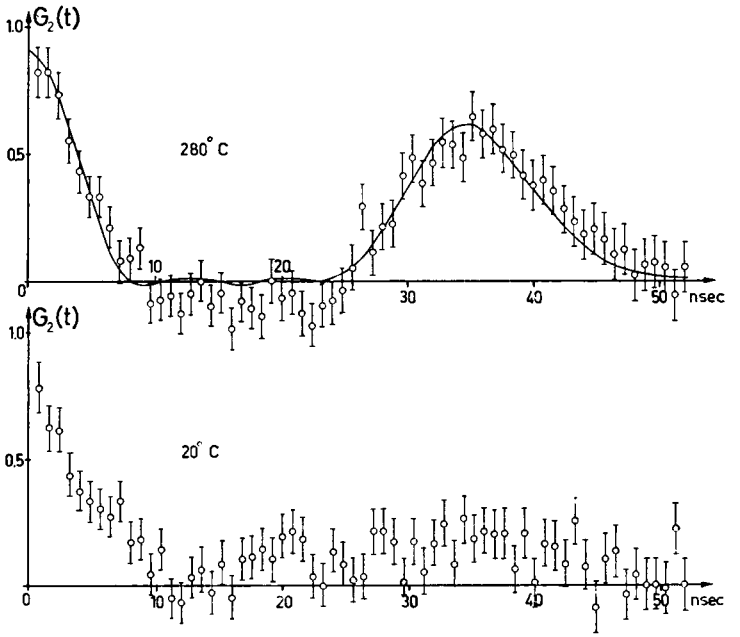

Fig. 2. Experimental results for $G_2(t)$ at 20°C and 280°C. The full line is the theoretical G_2 for a frequency of 180 MHz, a frequency spread of 10% and corrected for finite time resolution.

were performed with the same source as used in the measurements described above. The time resolution was set at 0.8 μs and the measurements were done at three different temperatures. The results after correction for solid angle are shown in table 1.

TABLE 1

Results of the integral angular correlation measured at different temperatures

Temperature	$A_2\bar{G}_2$	$\bar{G}_2$
20°C	-0.023 ± 0.007	0.13 ± 0.03
120°C	-0.028 ± 0.008	0.16 ± 0.04
280°C	-0.037 ± 0.008	0.21 ± 0.04

The integral attenuation coefficients $\bar{G}_2$ were calculated assuming $A_2 = -0.180$. The values of $\bar{G}_2$ at 20°C and 120°C are below the hard core value ($\bar{G}_2$(h.c.) = 0.20). These facts reveal that a time dependent perturbation exists in In_2O_3 at room temperature and, somewhat reduced, at 120°C.

At 280°C, however, the integral attenuation coefficient becomes slightly higher than the hard core value, indicating that the perturbation is solely static. The $\bar{G}_2$ coefficient can be calculated knowing the interaction frequency and the lifetime of the intermediate state [1]). For spin $I = \frac{5}{2}$

$$\bar{G}_2 = \frac{1}{5}\left[1 + \frac{\frac{13}{7}}{1+(\omega_0\tau_N)^2} + \frac{\frac{10}{7}}{1+(2\omega_0\tau_N)^2} + \frac{\frac{5}{7}}{1+(3\omega_0\tau_N)^2}\right]$$

and for $\omega_0 = 180$ MHz and $\tau_N = 122.6$ ns the value of $\bar{G}_2$ is

$$\bar{G}_2 = 0.2010 \pm 0.0003,$$

which agrees with the value found in the integral angular correlation experiment at 280°C. A possible frequency spread introduces a slight increase in the theoretical value of $\bar{G}_2$, but as a whole it is not very sensitive to frequency variations in this frequency region.

4. Discussion

From the measurements carried out one can conclude that in connection with the 173-247 keV gamma cascade in Cd^{111} in an In_2O_3 polycrystalline source there exist two types of perturbations:

(a) a static electric quadrupole perturbation with an interaction frequency $\omega_0 = 180$ MHz, and

(b) a time dependent perturbation that is mixed with the static one at room temperature and becomes almost negligible at about 300°C.

It is possible that this time dependent perturbation is due to the ionized state in which Cd atoms are left after the electron capture, The recovery time is then strongly temperature dependent, and at 20°C is longer than 10^{-8} s. This effect may be understood, considering that for higher temperatures the electronic interaction between atoms in the crystal is stronger, which decreases the recovery time.

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Note added in proof: The half-life of the upper level of the cascade (419 keV state) was recently measured by Sparrman *et al.* [15]) and found to be $(1.2 \pm 0.3) \cdot 10^{-10}$ s. Thus the correlation may be perturbed if the ionized states of the atoms have a longer life-time than 10^{-20} s.

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