PERTURBED 208.00−59.54 keV GAMMA-GAMMA ANGULAR CORRELATION IN ^{237}Np

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Abstract: The perturbed directional 208.00 — 59.54 keV gamma-gamma angular correlation was measured in ^{237}Np. The static hyperfine coupling between the nuclear magnetic moment and the angular momentum of the electronic states was found present both in the liquid source and in the polycrystallinic source. The spin sequence for the 208.00 → 59.54 → 0 keV transition was confirmed to be 3/3, 5/2, 5/2 with the mixing ratio $\delta = +0.145$ for the $E2$ admixture in the preferably $M1$ transition. The agreement between directional correlation and internal conversion data was shown to be fairly good. The angular momentum J of the electronic states was determined for both sources: for the polycrystallinic source the $J = 1$ value gives the attenuation factor $G_{22}(t)_{J=1}$ in good agreement with the experimental results, while for the liquid source, the best J value was shown to be $J = 3/2$. The J-dependent hyperfine structure constant a was determined for the polycrystalinic source as $\frac{a}{\hbar} = 83 \pm 3$ MHz and for the liquid source as $\frac{a}{\hbar} = 44 \pm 2$ MHz. The average magnetic field at the place of ^{237}Np nuclei was deduced for the polycrystallinic source $\overline{H(0)_{J=1}} = 54 \pm 14$ [kOe] and for the liquid source $\overline{H(0)_{J=3/2}} = 42 \pm 12$ [kOe].

1. Introduction

The rather long lifetime of the 59.54 keV level suggested a possible strong perturbation for the cascade transition through this level. During the last few years the 208 — 59.54 keV gamma-gamma angular correlation in ^{237}Np has been measured several times in IFJ laboratory with the aim to find out the perturbation mechanism which takes place. Since neptunium is in the region of deformed nuclei one could expect a possible static electric quadrupole perturbation (SEQP). On the other hand, unfilled $5f$ and $6d$ electron shells in neptunium leave a possibility for a large static hyperfine coupling (SHC) of the nuclear magnetic moment and the angular momentum of electronic states. However, static perturbations are not the only possible ones in this case. For a source in an aqueous solution a time-dependent perturbation could take place. Bodensted[1] suggested that an accurate measurement of this cascade could decide between the Abragam-Pounds[2] and the Dillenburg-Maris[3] perturbation mechanism. Unfortunately, we did not succeed in attaining

the desired accuracy in this case, and it was rather difficult to decide upon the type of perturbation present. It was therefore desirable to determine the multipolarities and mixing ratios for the 208.00 and 59.54 keV electromagnetic transitions by an independent measurement, in order to minimize the number of »free« parameters to those only which describe the perturbation. The energy levels of ^{237}Np were recently studied from the alpha decay of ^{241}Am[4] and from the beta decay of ^{237}U[5],
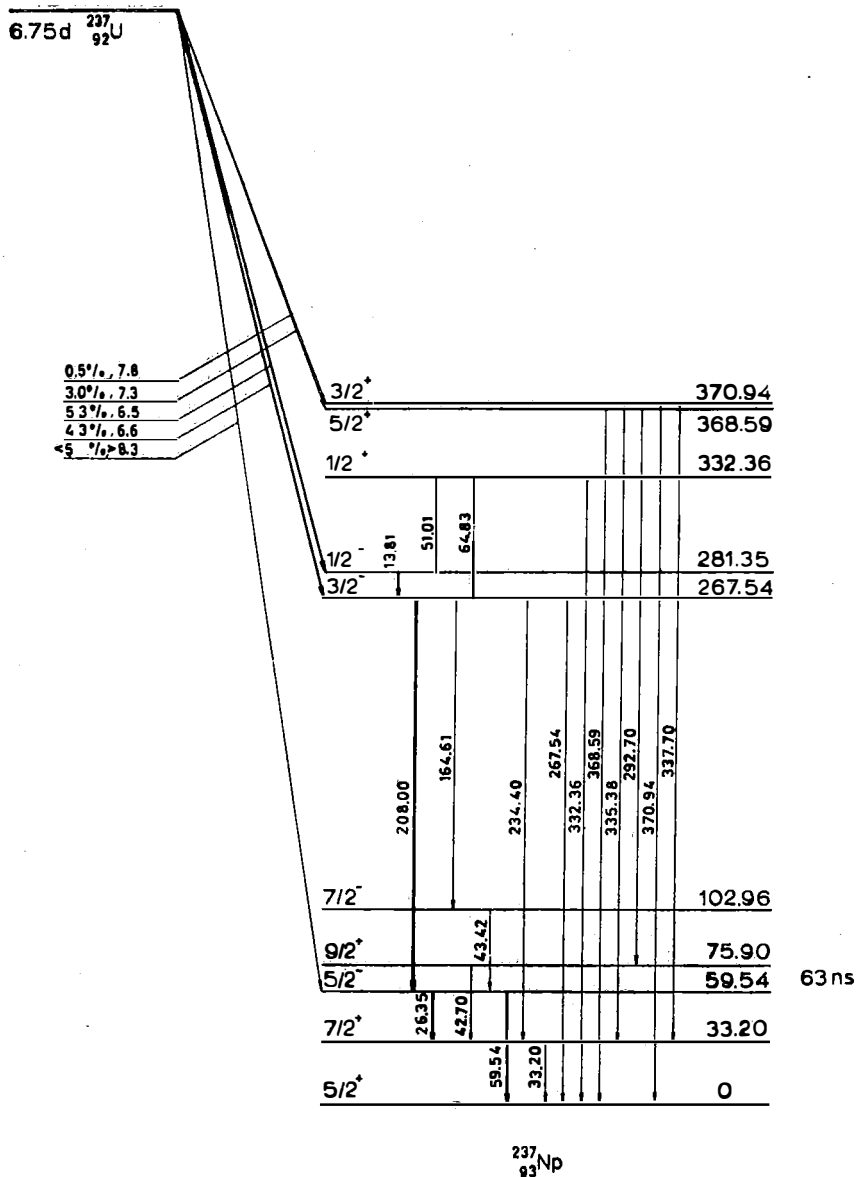

Fig. 1. Decay scheme of ^{237}Np.

and a revised decay scheme was proposed (see Fig. 1). Internal conversion data show that the 208.00 keV transition is mainly $M1$ with a mixture of 2.1% $E2$, while 59.54 keV is a pure $E1$ transition[4,5]. The spin sequence for the cascade 208.00 → 59.54 → 0 keV transition was confirmed to be 3/2, 5/2, 5/2.

2. Source and the experimental set-up

The ^{237}U source was the ^{238}U (fast n, 2n) ^{237}U reaction with fast neutrons obtained from the ^{9}Be (d, n) ^{10}B reaction. From the irradiated uranium metal UO_2Cl_2 was prepared. A single spectrum is shown in Fig. 2. The source was used in two different forms: in the liquid form (aqueous solution) and in the polycrystallinic form obtained by drying the liquid source.

The experimental arrangement contained two $1\frac{1}{2}'' \times 1\frac{1}{2}''$ NaJ(Tl) crystals mounted on the photomultipliers 56 AVP type followed by a standard fast-slow system. The coincidence gated time spectrum was stored in a 100-channel analyzer. The time resolution for prompt coincidences from ^{22}Na and for the energies chosen
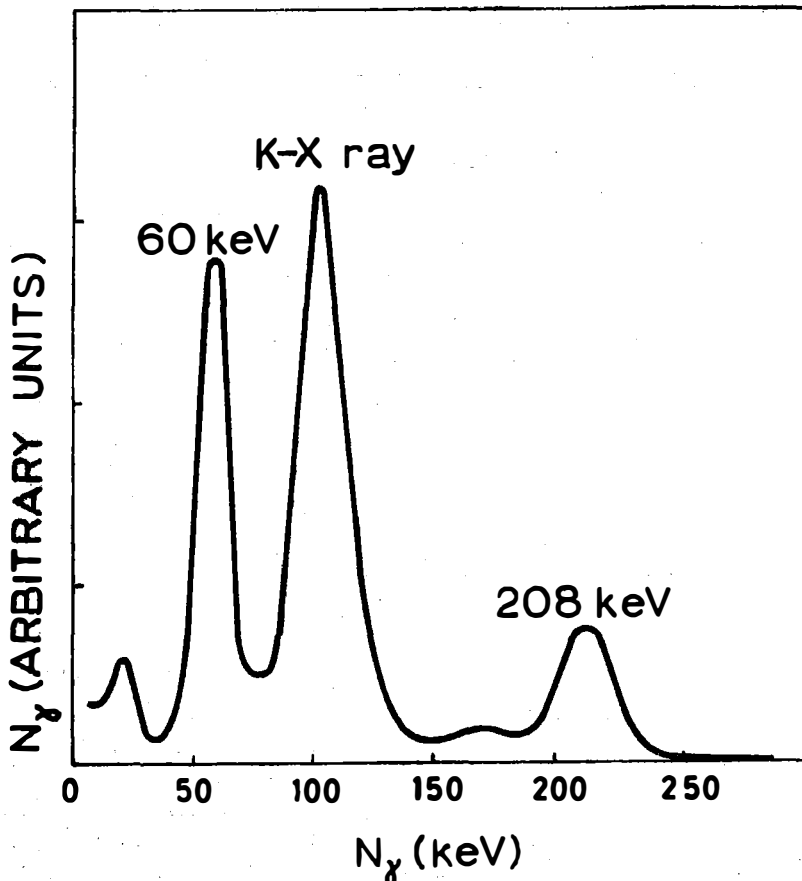

Fig. 2. The single gamma spectrum from the ^{237}U beta decay.

by the single channel analyzers in the 208.00 and 59.54 keV region was $2\tau = 9$ ns at FWHM of the prompt curve. The integral and the differential correlation data were taken simultaneously. The time base used for the integral measurement was 192 ns. The 208 keV photopeak single rate, selected in a movable detector, was used to normalize the coincidence counting rate. Thus the different Compton contributions from higher energy transitions in the 59.54 keV window at $\Theta_1 = 90°$ and $\Theta_2 = 180°$ angles between the detectors were taken into account. Since the higher energy transitions are very weak, their contribution in the 208.00 keV window is negligible and the normalization with the 208.00 keV intensity is justified.

3. Directional correlation measurement

For a pure 59.54 keV $E1$ transition $(5/2^- \rightarrow 5/2^+)$ the directional angular correlation function has the simple form

$$W(\Theta, t) = 1 + A_{22} G_{22}(t) P_2(\cos \Theta), \tag{1}$$

where $G_{22}(t)$ is the attenuation factor determined by perturbation, and A_{22} reads

$$A_{22} = \frac{F_2(5/2\ 3/2\ 1\ 1) + 2\delta F_2(5/2\ 3/2\ 1\ 2) + \delta^2 F_2(5/2\ 3/2\ 2\ 2)}{1 + \delta^2} \times$$

$$\times F_2(5/2\ 5/2\ 1\ 1), \tag{2}$$

where δ is the mixing ratio, and F's are the well-known tabulated angular correlation coefficients. After subtracting the accidental coincidences from the measured time spectra $N(180°, t)$ and $N(90°, t)$ we calculated

$$R(t) = 2 \frac{N(180°, t) - N(90°, t)}{N(180°, t) + 2N(90°, t)} = A_{22} G_{22}(t). \tag{3}$$

In order to improve the statistics in $N(\Theta, t)$, the counting rates in two neighbouring channels were summed. The $N(\Theta, t)$ values obtained were used to calculate the experimental $R(t)$ curve. The experimental points $R(t)$ drawn in Fig. 3 were obtained by taking the arithmetic mean of the two neighbouring $R(t)$ values. The experimental $R(t)$ values in both cases (the polycrystallinic source and the liquid source) show two main characteristics:

   i) the mean level for $R(t)$ is constant in time;
   ii) the $R(t)$ curve has an oscillatory character.

The constancy of the mean level assures that we are not dealing with a time--dependent perturbation. Static perturbations are left as the only possibility. However, the electric quadrupole perturbation can be ruled out even after a rough comparison of $R(t)_{exp}$ with the theoretical $R(t)_{th} = A_{22} G_{22}(t)_{th}$ curve for the spin $I = 5/2$. In a good approximation the equality $\omega_0 \tau \approx 15.7$ is valid. This indicates a limiting value for the attenuation factor $G_{22}(\infty)_{lim} = 0.2$. Using a 2.1% $E2$ admixture in the $M1$ 208.00 keV transition from the internal conversion data[5], from our directional correlation measurement we obtain $\delta = +0.145$ as the only
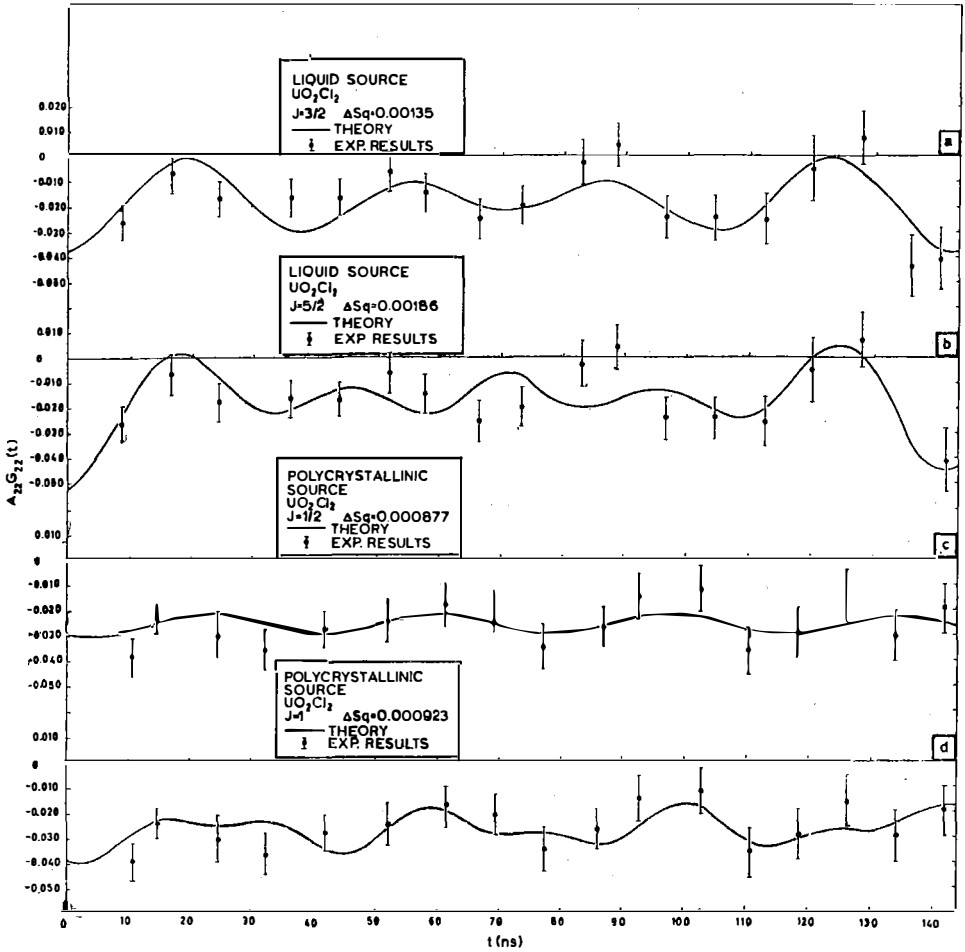
Fig. 3. The experimental and the teoretical $R(t)$ values for the polycrystallinic and for the liquid source for the J values which give the best fit to the theoretical $G_{22}(t)_J$ J-dependent attenuation coeficients.

possibility, and consequently $A_{22} = - 0.040$. This further indicates that the mean value of the experimental results in the case of SEQP should be $A_{22} G_{22}(\infty)_{\text{lim}} = = - 0.008$, which is beyond the mean values of our experimental $R(t)$ values:

$$\text{liquid source} \qquad R(t) = - 0.017 \pm 0.04,$$
$$\text{polycrystallinic source} \quad R(t) = - 0.026 \pm 0.04.$$

Hence, the only remaining perturbation is the static hyperfine coupling (SHC). The attenuation factor for SHC reads[6]

$$G_{kk}(t) = \sum_{F, F'} (2F + 1)(2F' + 1) \begin{Bmatrix} F & F' & k \\ I & I & J \end{Bmatrix}^2 \exp\left(- \frac{i}{\hbar}(E_F - E_{F'})t\right), \quad (4)$$

where I is the spin of the intermediate state and

$$E_F = \frac{a}{2} \left[F(F+1) - I(I+1) - J(J+1) \right] \tag{5}$$

are the eigenvalues of the magnetic $I-J$ hyperfine coupling in the absence of the external magnetic field. Here a is the magnetic hyperfine structure constant

$$a = \frac{g \cdot \mu_N}{I} \frac{\overline{H(0)}_J}{J}, \tag{6}$$

where J is the angular momentum of the electronic states of an atom or of a molecule, and $\overline{H(0)}_J$ the average magnetic field produced by the electrons around the nucleus. The attenuation factor is obviously dependent on the unknown (electron) angular momentum J of the uranil complexes and may possibly be different for the sources used in two different forms. It was therefore decided to compute the SHC attenuation factors with the J-values as free parameters.

To avoid any errors, the treatment which had to give the theoretical $R(t)_J$ curve was made identical with that used in obtaining $R(t)$ from the $N(90°, t)$ and $N(180°, t)$ spectra. For $N(\Theta, t)$ we assumed the general form

$$N(\Theta, t) = C \left[1 + K G_{22}(t) \right] \exp\left(-\frac{t}{\tau} \right) P_2(\cos \Theta). \tag{7}$$

The attenuation factor $G_{22}(t)$ is of the form

$$G_{22}(t)_J = \sum_{F, F'} (2F+1)(2F'+1) \left\{ \begin{matrix} F & F' & 2 \\ 5/2 & 5/2 & J \end{matrix} \right\}^2 \cos\left(\frac{E_F - E_{F'}}{\hbar} t + \varphi \right). \tag{8}$$

We assumed a gaussian distribution for the prompt time resolution curve with FWHM equal to 9 ns.

$$P(t, t')\, dt' = \frac{1}{\Delta \sqrt{2\pi}} \exp\left[-\frac{(t-t')^2}{2\Delta^2} \right] dt', \tag{8}$$

and the average which takes into account the finite time resolution $\overline{N(\Theta, t)_J^{2\tau_\bullet}}$ was calculated from

$$\overline{N(\Theta, t)_J^{2\tau_\bullet}} = \frac{\displaystyle\int_{t-\tau}^{t+\tau} P(t, t')\, N(\Theta, t')\, dt'}{\displaystyle\int_{t-\tau}^{t+\tau} P(t, t')\, dt'}. \tag{10}$$

The computer programme ensured the summation of the neighbouring N values before computing the theoretical $R(t)_J$ values. The subsequent averaging on the $R(t)_J$ curve was performed in the same manner as for obtaining the experimental $R(t)$ values. The theoretical $R(t)_J$ values thus obtained were ready for comparison with $R(t)$ (Fig. 2). The theoretical $\overline{R(t)_J}$ values were computed for the four

J-values: $J = 1/2, 1, 3/2, 5/2$. The goodness of the fit for different J values was measured by a quantity s:

$$s = \sum_i (A_{22} \overline{G_{22}(t_i)_{J,\,th}} - A_{22} G_{22}(t_i)_{exp})^2, \tag{11}$$

where the sum goes over all experimental points.* The results are shown in Table 1·

Table 1

The depedence on s values showing the best J values for the source used

J	s	J	s
1/2	$0.887 \cdot 10^{-3}$	1/2	$0.258 \cdot 10^{-2}$
1	$0.923 \cdot 10^{-3}$	1	$0.253 \cdot 10^{-2}$
3/2	$0.122 \cdot 10^{-2}$	3/2	$0.135 \cdot 10^{-2}$
5/2	$0.195 \cdot 10^{-2}$	5/2	$0.186 \cdot 10^{-2}$

From the tabulated results one can conclude that two J values, i. e. $J = 1/2$ and $J = 1$ give a better fit for the polycrystallinic source and $J = 3/2$ and $J = 5/2$ for the liquid source. The least mean square fit of the experimental $R(t)$ points to the theoretical $R(t)_J$ values is drawn in Fig. 3, and the results are given in Table 2. The numbers in parenthesis indicate the errors. The parameter K introduced through eq. (7) is simply related to A_{22}

$$K = Q_2^{(1)} \, Q_2^{(2)} \, A_{22},$$

where $Q_1^{(1)}$ and $Q_2^{(2)}$ are the finite solid angle correction factors.

It is, therefore, easy to conclude that only $J = 1$ for the polycrystallinic source and $J = 3/2$ for the liquid source can give A_{22} which simultaneously satisfy the directional correlation and internal conversion data. Taking the mean value of K from the two independent measurements, one obtains

$$A_{22} = - (0.046 \pm 0.005).$$

This A_{22} value can be compared with that obtained from the integral measurement. The time integrated attenuation factor defined by eq. (8) was computed in the usual way[6]

$$\overline{G_{22}(t)_J^{192}} = \frac{\int_0^{192} G_{22}(t) \, e^{-\frac{t}{\tau}} \, dt}{\int_0^{192} e^{-\frac{t}{\tau}} \, dt}. \tag{12}$$

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* $\overline{G_{22}(t_i)_{J,\,th}}$ means two subsequent averagings over the finite time resolution and taking the arithmetic mean for the neighbouring values.

Table 2

The values of the constant K, hyperfine structure constant in $\hbar$ units, and phase φ obtained for the best J values for the two sources

Polycrystallinic source				Liquid source			
J	K	$\dfrac{a}{\hbar}$ [ns^{-1}]	φ [rad]	J	K	$\dfrac{a}{\hbar}$ [ns^{-1}]	φ [rad]
1/2	$-0.031(4)$	0.956(2)	$-0.80(5)$	3/2	$-0.041(5)$	0.044(2)	$-0.00(1)$
1	$-0.043(5)$	0.083(3)	$-0.80(5)$	5/2	$-0.058(6)$	0.045(2)	0.37(2)

The integral measurement gave the experimental anisotropy and by calculating $\overline{G_{22}(t)_J}^{192}$ we obtained

$$A_{22} = \frac{2A}{\overline{G_{22}(t)_J}^{192}(3+A)}. \tag{13}$$

The A_{22} values thus obtained are given in Table 3.

The average magnetic field $\overline{H(0)_J}$ at the nucleus can now be easily deduced for both sources by the help of eq. (6). Knowing for $g = 0.8 \pm 0.2$, one has $H(0)_{J=1} = 54 + 14$ [kOe] for the polycrystallinic source and $H(0)_{J=3/2} = 42 \pm 12$ [kOe] for the liquid source.

Table 3

The anisotropy A determined from the integral measurement and the coefficient A_{22} calculated from the integral attenuation factor

Polycrystallinic source			Liquid source		
A	$\overline{G_{22}(t)_{J=1}}^{192}$	A_{22}	A	$\overline{G_{22}(t)_{J=1}}^{192}$	A_{22}
$-0.042(3)$	0.6697	$-0.042(2)$	$-0.028(5)$	0.434	$-0.043(4)$

Acknowledgement

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PERTURBIRANA KUTNA GAMA-GAMA KORELACIJA
208.00 — 59.54 keV U ^{237}Np

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Sadržaj

Mjerena je perturbirana kutna gama-gama korelacija 208.00 — 59.54 keV u ^{237}Np. Pokazano je da perturbacija ove korelacije za vrijeme života ($T_{1/2} = 63$ ns) medijalnog 59.54 keV stanja dolazi od magnetske hiperfine interakcije između magnetskog dipolnog momenta jezgre i kutne količine gibanja elektronskih stanja za tekući i polikristaličan izvor. Potvrđen je niz spinova 3/2, 5/2, 5/2 za prijelaz 208.00 → 59.54 → 0 keV. Omjer miješanja $\delta = + 0,145$ za $E2$ primjesu u 208.00 keV u uglavnom $M1$ prijelazu je u vrlo dobrom slaganju sa mjerenjima konverzionih koeficijenata. Određena je kutna količina gibanja $J = 1$ elektronskih stanja za izvor u polikristalnom obliku i $J = 3/2$ za tekući izvor. Konstanta hiperfine interakcije a (zavisna o kutnoj količini gibanja elektronskih stanja) određena je za oba izvora (u jedinicama $\hbar$): za polikristaličan izvor $\frac{a}{\hbar} = 83 \pm 3$ [MHz], a za tekući izvor $\frac{a}{\hbar} = 44 \pm 2$ [MHz]. To omogućuje izračunavanje prosječnog magnetskog polja na mjestu jezgre za polikristaličan izvor $\overline{H(0)_{J=1}} = 54 \pm 14$ [kOe] i za tekući izvor $\overline{H(0)_{J=3/2}} = 42 \pm 12$ [kOe].