# Perturbation of the (1063.62-569.67) keV gamma-gamma directional correlation in $^{207}$Pb

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$\gamma$-$\gamma$ directional correlation studies have been carried out on the (1063.62-569.67) keV $13/2^+ \xrightarrow{M4+E5} 5/2^- \xrightarrow{E2} 1/2^-$ cascade in the decay of $^{207}$Bi using the time differential perturbed angular correlation method. The measurements show time dependent magnetic perturbation of the correlation. From the plot of $A_2(\bar{t})$ vs $\bar{t}$ (finite time resolution corrected), the value of the relaxation parameter $\lambda_2$ and the unperturbed $A_2(0)$ coefficient (solid angle corrected) obtained are $(2.4 \pm 0.3) \times 10^9$ sec$^{-1}$ and $0.259 \pm 0.016$, respectively. From the plot of $A_0(t)$ vs $t$, the unperturbed value of $T_{1/2}$ and therefrom $\lambda_0$ for the 569.67 keV level are determined to be $(130.7 \pm 1.3)$ ps and $(5.30 \pm 0.06) \times 10^9$ sec$^{-1}$, respectively. Using the $\lambda_0$ and $\lambda_2$ values, the integral attenuation coefficient $\bar{G}_2(\infty)$ has been estimated to be $0.69 \pm 0.03$. The differential data at various angles were integrated and the solid angle corrected $A_2(\infty)$ coefficient thus obtained is $0.204 \pm 0.005$. The perturbation mechanism is discussed in the light of the spin-spin interactions between the $6s$ electrons of the neighboring paramagnetic $Pb^{+++}$ ions. The results are found to be consistent with the theory of Abragam and Pound.

[RADIOACTIVITY $^{207}$Bi, measured $\gamma\gamma(\theta,t)$, $^{207}$Pb level deduced $\lambda_0$, $\lambda_2$, $A_2(0)$, $\bar{G}_2(\infty)$, $A_2(\infty)$, $\delta$ (1063.62 $\gamma$), $B(M4)\downarrow$ and $B(E5)\downarrow$ values.]

## I. INTRODUCTION

Several integral correlation measurements[1-7] have been performed on the (1063.62-569.67) keV $\gamma\gamma$ cascade in $^{207}$Pb. The $A_2$ and $A_4$ values are nearly in agreement with the theoretical values for the spin sequence $\frac{13}{2}^+(M4+E5)\frac{5}{2}^-(E2)\frac{1}{2}^-$, assuming $\delta(1063.62\gamma) = -0.03$. Körner *et al.*[5] have also performed time differential correlation measurements on this cascade using a $^{207}$Bi source in aqueous solution of $BiCl_3$. No perturbation of the correlation was observed by them. However, it is not understandable why their $A_2(0) \approx 0.125$, as can be seen from their plot of $A_2(t)$ vs $t$. Even if it is corrected for the solid angle (not given) it would be very low compared to $A_2 = 0.232$ obtained by them from an integral measurement.

We, therefore, considered it worthwhile to re-examine the question of perturbation, if any, of this correlation using the time differential perturbed angular correlation (PAC) method. Our measurements show an exponential decrease of the anisotropy. The results are discussed in the light of time-dependent magnetic perturbation.

## II. EXPERIMENTAL DETAILS

The $^{207}$Bi source was obtained from New England Nuclear and used in the dried-up form $Bi(NO_3)_3 \cdot 5H_2O$ in a cylindrical perspex vial of dimensions 3 mm diam × 4 mm. A singles $\gamma$ spectrum recorded with a Ge(Li) detector showed no impurity.

A timing coincidence spectrometer (Fig. 1) using 2.54 cm diam × 1.27 cm and 2.54 cm diam × 2.54 cm NE-111 plastic scintillators with XP-1021 photomultipliers and constant fraction pulse height triggers was employed. Aluminum absorbers of appropriate thickness were used in front of the scintillators to cut out the $K$ and $L$ conversion electrons. The energy gates for the 569.67 and the 1063.62 keV transitions were 50% and 40% of their respective Compton distributions. The time calibration of the time-to-pulse-height convertor (TPHC) was carried out by observing the shift of the prompt resolution curve using a $^{60}$Co source with the introduction of an accurately calibrated GR874-L30 air dielectric coaxial line of $(1.0036 \pm 0.0002)$ ns. The time calibration thus obtained was $(35.7 \pm 0.4)$ ps/channel.

The differential nonlinearity (DNL) curve for the system was recorded. The nonlinearity was estimated to be < 1% and hence no correction was applied.

The full width at half maximum (FWHM) of the prompt curve at the above energy settings using a $^{60}$Co source was 310 ps with a slope ($\frac{1}{2}$) of 50 ps. The solid angle correction factor $Q_2$ for the correlation geometry and at the energy settings mentioned above was determined experimentally by performing $^{60}$Co correlations under identical conditions ($Q_2 = 0.816 \pm 0.002$). The delayed time coincidence spectra were recorded at 90, 135, 180, 180, 225, and 270°. The data from corresponding angles in the two quadrants were stored in a 4 × 1024 mode of a 4096 channel analyzer. Simultaneously, the singles counts from both detectors were also measured for normalization purposes.

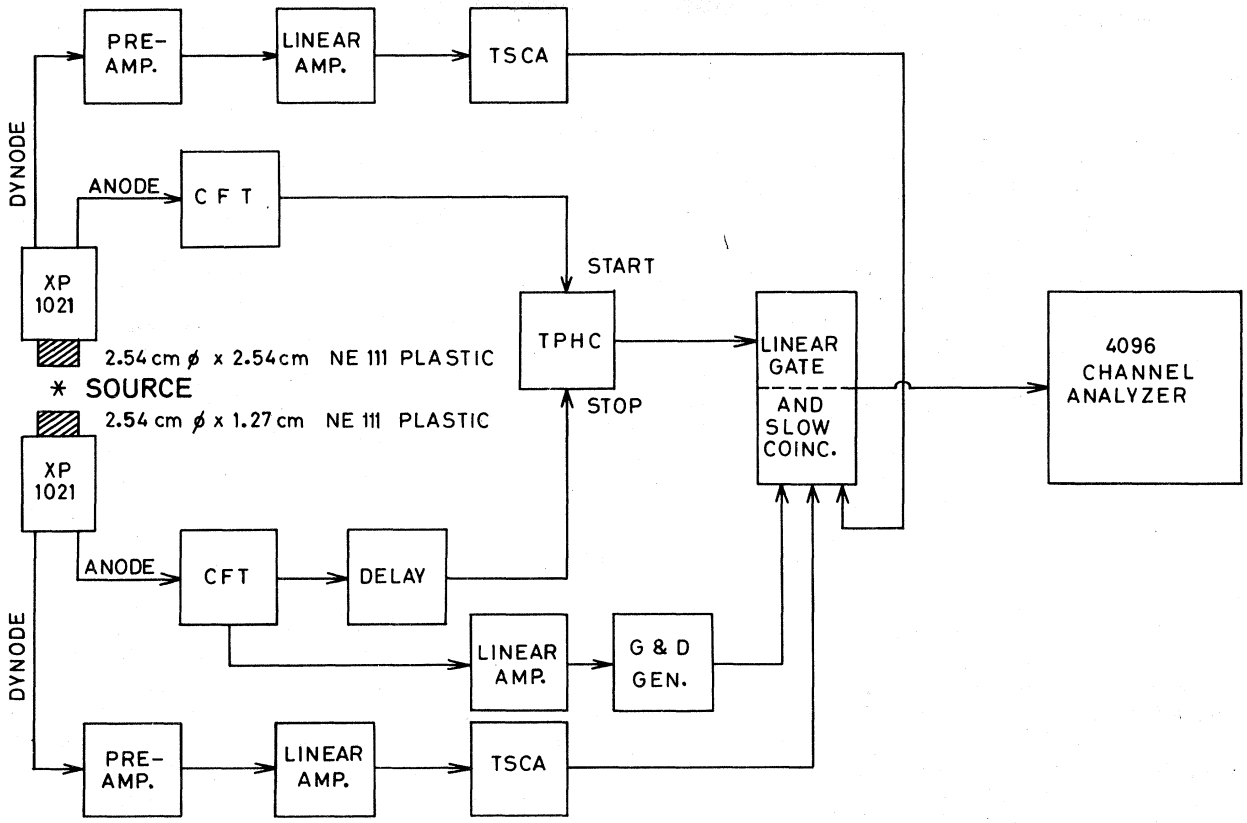


FIG. 1. Block diagram of the timing spectrometer used for $\gamma$-$\gamma$ time differential PAC measurement.

## III. MEASUREMENTS AND RESULTS

Figure 2 shows the delayed time coincidence spectra recorded at 90 (III), 135 (II), and 180° (I). After subtracting chance coincidences and normalizing with the gate counts, the data at each corresponding channel in the delayed region were analyzed by the weighted least squares method for $A_0(t)$ and $A_2(t)$ coefficients. After applying a finite time resolution correction,[8] a plot of $A_2(\bar{t})$ vs $\bar{t}$ is shown in Fig. 3 along with the relevant part of the decay scheme. This clearly indicates the exponential behavior of the perturbation. The $A_2(\bar{t})$ values were least squares fitted to the equation:

$$A_2(\bar{t}) = A_2(0)e^{-\lambda_2 \bar{t}} , \qquad (1)$$

yielding the relaxation parameter $\lambda_2 = (2.4 \pm 0.3) \times 10^9$ sec$^{-1}$ and the unperturbed expansion coefficient $A_2(0) = 0.21 \pm 0.01$. After solid angle correction, $A_2(0) = 0.259 \pm 0.016$. The contribution of the $\frac{7}{2}^- \xrightarrow{1770\cdot 2\gamma} \frac{5}{2}^- \xrightarrow{569\cdot 67\gamma} \frac{1}{2}^-$ interfering cascade ($A_2 = -0.0087 \pm 0.0089$[9]) was estimated to be $\leq 2\%$. No correction for this was therefore applied. The $A_2(0)$ of the present work is on the higher side of all the integral values reported so far[1-7] and is also in sharp disagreement with $A_2(0) \simeq 0.125$

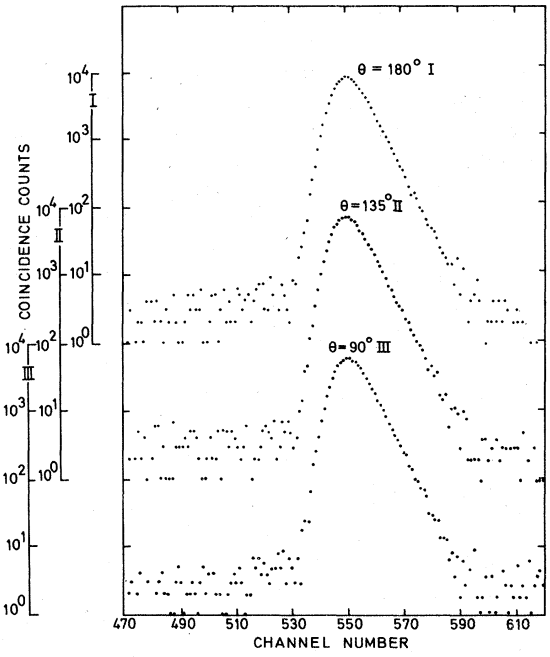


FIG. 2. Delayed time spectra for (1063.62 $\gamma$) (569.67 $\gamma$) coincidences in $^{207}$Pb recorded at $\Theta = 180$ (I), 135 (II), and 90° (III).

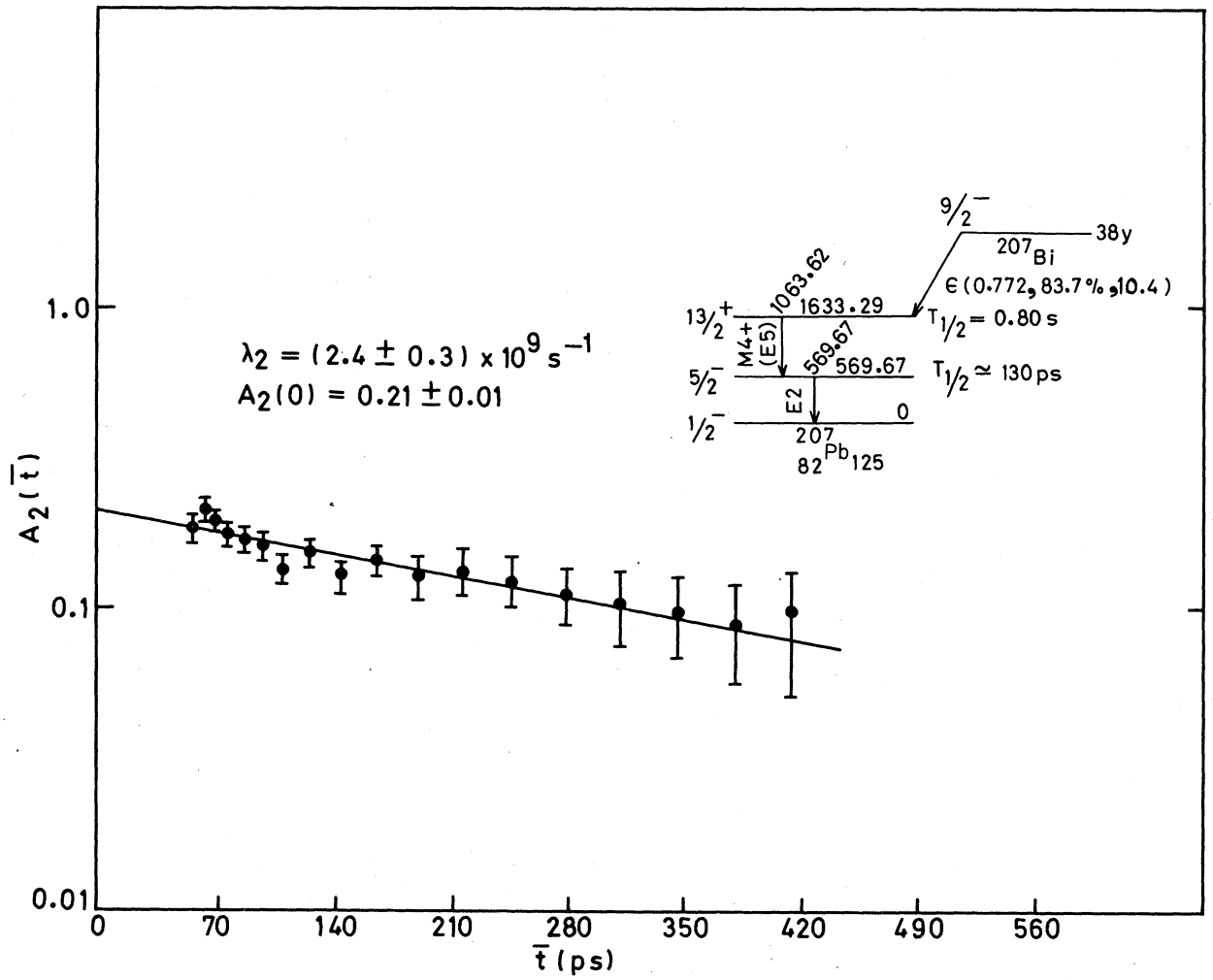


FIG. 3. Plot of $A_2(\bar{t})$ (uncorrected for solid angle) vs $\bar{t}$ from the time differential measurement in $^{207}$Pb for a dried-up $Bi(No_3)_3 \cdot 5H_2O$ source.

(solid angle uncorrected) obtained by Körner *et al.*[5] from their time-differential measurements using an aqueous $BiCl_3$ source. Surprisingly, Körner *et al.*[5] conclude that there is no perturbation of the correlation, yet their $A_2(0)$ value turns out to be very small as compared to their integral value of $A_2 = +0.232 \pm 0.007$ obtained in a separate experiment. This is inconsistent because for no perturbation is $A_2(0) = A_2(\infty)$.

The $A_0(t)$ values were also least squares fitted to the function $A_0(0)e^{-\lambda_0 t}$ as shown in Fig. 4. This gave $T_{1/2}$(569.67 keV level) = (130.7 ± 1.3) ps or $\lambda_0 = (5.30 \pm 0.06) \times 10^9\ s^{-1}$ which is in good agreement with the value obtained by Körner *et al.*[5] Knowing $\lambda_2$ and $\lambda_0$ and using the relation:

$$\bar{G}_2(\infty) = \frac{\lambda_0}{\lambda_2 + \lambda_0} \qquad (2)$$

for a time dependent perturbation, the integral attenuation coefficient $\bar{G}_2(\infty) = 0.69 \pm 0.03$. Using solid angle corrected $A_2(0) = 0.259 \pm 0.016$, the product $A_2(0)\bar{G}_2(\infty)$ yields $A_2(\infty) = 0.18 \pm 0.01$, which is in fair agreement with the solid angle corrected $A_2(\infty) = 0.204 \pm 0.005$ obtained by integrating the differential data. The $A_2$ values obtained from our measurements are compared with those of earlier investigations in Table I. Using our $A_2(0)$ value, the mixing ratio analysis yields $\delta(1063.62\gamma) = -0.12^{+0.06}_{-0.08}$ which is substantially higher than the earlier values[2,3,5-7] ($\approx -0.03$) for the $M4 + E5$ admixture.

The delayed time coincidence spectra (Fig. 2) were also analyzed for half-life values at each angular position. The values obtained are $T_{1/2}(90°) = (133.8 \pm 1.6)$ ps, $T_{1/2}(135°) = (129.7 \pm 1.5)$ ps, and $T_{1/2}(180°) = (125.9 \pm 1.4)$ ps. A critical ex-

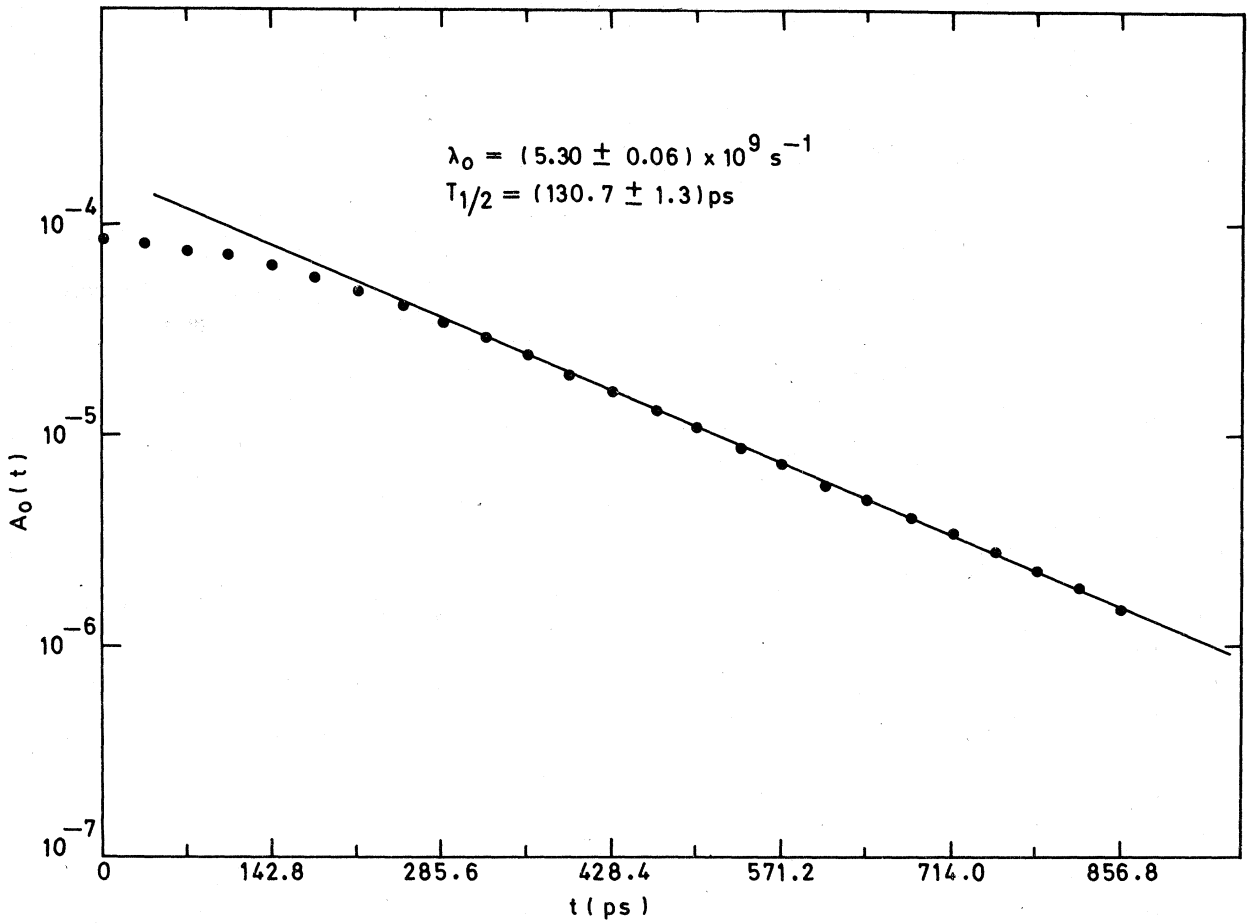


FIG. 4. Plot of $A_0(t)$ vs $t$ from the differential data.

amination of these values shows that the extranuclear perturbation does exist. Further, using these values, the unperturbed value of $T_{1/2}$ was calculated by the method of quadratic interpolation[10]: $T_{1/2}(125°40') = (130.5 \pm 1.5)$ ps. This is in good agreement with $T_{1/2} = (130.7 \pm 1.3)$ ps as obtained from our $A_0(t)$ vs $t$ plot (Fig. 4). These values are compared with those of earlier investigations in Table II.

## IV. DISCUSSION

Bismuth exists in the source $[Bi(NO_3)_3 \cdot 5H_2O]$ in the triply ionized form $Bi^{+++}$. Following electron capture, it decays to the 1633.29 keV isomeric state in $^{207}Pb$. This brings about the following change in the electron configuration:

$$Bi^{+++}(5d^{10}6s^2) \xrightarrow{\epsilon} Pb^{+++}(5d^{10}6s^1)\ . \qquad (3)$$

Therefore, $Pb^{+++}$ is expected to exhibit paramagnetic relaxation behavior due to its nonstationary and partially filled $6s$ shell. This is mainly a consequence of the spin-lattice interaction in solid sources. Further, on account of the spin-spin interaction between the $6s$ electrons of the neighboring paramagnetic ions, the $6s$ electron presumably undergoes a spin-flip. This reverses the direction of the hyperfine field $H_0$, thereby producing a wildly fluctuating magnetic field at the site of the decaying nucleus. This would bring about a strong time dependent perturbation of the correlation.

The following calculations have been carried out in the light of the theory of Abragam and Pound.[11] Kopfermann (Ref. 12, p. 132) has estimated the value of the magnetic field ($\langle H_0 \rangle = 6.5 \times 10^7$ Oe) due to the unpaired $6s$ electron in $^{207}Bi$ in its fifth stage of ionization.

For the time dependent magnetic interaction:

$$\lambda_2 = a'[1 - (2I+1)W(I12I\,|\,II)]\ , \qquad (4)$$

where

TABLE I. $A_2$ and $A_4$ expansion coefficients for the (1063.62–569.67) keV $\gamma\gamma$ cascade in $^{207}$Pb.

| Reference | Source form | Integral $A_2$ | Integral $A_4$ | Differential $A_2$ (Unperturbed) | Differential $A_4$ |
|---|---|---|---|---|---|
| 1 | $^{207}$Bi attached to the resin in 0.5 $N$ HCl | +0.2168<br>+0.2114<br>+0.2260 | −0.0097<br>−0.0328<br>−0.0209 | ...<br>...<br>... | ...<br>...<br>... |
| 2 | Liquid<br>solid | +0.219±0.003<br>+0.224±0.006 | −0.020±0.005<br>−0.031±0.007 | ...<br>... | ...<br>... |
| 3 | Solid $BiCl_3$<br>(5 $\mu$Ci)<br>(40 $\mu$Ci) | <br>+0.227±0.005<br>+0.232±0.005 | <br>−0.028±0.009<br>−0.025±0.008 | <br>...<br>... | <br>...<br>... |
| 4 | Dried up | +0.220±0.003 | −0.038±0.004 | ... | ... |
| 6 | Liquid $BiCl_3$ in dil. HCl | +0.232±0.002 | −0.022±0.002 | ... | ... |
| 7 | 25 $\mu$Ci of $^{207}$Bi in 0.5 $N$ $HNO_3$ | +0.235±0.003 | −0.029±0.004 | ... | ... |
| 5 | Liquid $BiCl_3$ in 3 $N$ HCl | +0.232±0.007 | −0.022±0.003 | +0.125 (Solid angle uncorrected) | ... |
| Present work | Dried up $Bi(NO_3)_3\cdot 5H_2O$ | +0.204±0.005 | −0.004±0.003 | +0.259±0.016 (Solid angle corrected) | ... |

$$a' = \tfrac{2}{3}\tau_{s'}\omega_{s'}{}^2 I(I+1)J(J+1) \qquad (5)$$

and

$$\omega_{s'}{}^2 = \frac{g^2\langle H_0\rangle^2\mu_N{}^2}{\hbar^2 s'(s'+1)} .$$

$\tau_{s'}$ denotes the spin relaxation time of the 6$s$ electron shell, $\omega_{s'}$ is the magnetic interaction frequency, and $\mu_N$ is the nuclear magneton. Using our experimental $\lambda_2$ value, $\langle H_0\rangle = 6.5\times10^7$ Oe, $I=\frac{5}{2}$, $g=0.26$,[5] and $s'=\frac{1}{2}$ we obtain $\omega_{s'}=1.4\times10^{11}$ s$^{-1}$ and $\tau_{s'}=(0.78\pm0.09)\times10^{-13}$ s. The mean life for the 569.67 keV level is $\tau_N=1.87\times10^{-10}$ s. Thus for a time dependent magnetic hyperfine interaction, the conditions: $\tau_{s'}\ll\tau_N$ and $\omega_{s'}\tau_{s'}\ll 1$ are well satisfied.

### A. Reduced transition probabilities for the 1063.62 keV transition in $^{207}$Pb

The $M4$ and $E5$ reduced transition probabilities for the $i_{13/2}{}^{-1}\rightarrow f_{5/2}{}^{-1}$ 1063.62 keV $\gamma$ transition

TABLE II. $T_{1/2}$ values for the 569.67 keV level in $^{207}$Pb.

| Reference | Sandwich geometry $T_{1/2}$ (ps) | $T_{1/2}$ (ps) from differential experiment $\theta=90°$ | $\theta=135°$ | $\theta=180°$ | $\theta=125°\,40'$ (By quadratic interpolation) | From $A_0(t)$ vs $t$ plot |
|---|---|---|---|---|---|---|
| 17 | 90±30 | ... | ... | ... | ... | ... |
| 18 | 110±11 | ... | ... | ... | ... | ... |
| 19 | 110±11 | ... | ... | ... | ... | ... |
| 20 | 100±1 | ... | ... | ... | ... | ... |
| 21 | 134±9 | ... | ... | ... | ... | ... |
| 22 | 128±5 | ... | ... | ... | ... | ... |
| 5 | 129±3 | ... | ... | ... | ... | 128 ±3 |
| Present work | ... | 133.8±1.6 | 129.7±1.5 | 125.9±1.4 | 130.5±1.5 | 130.7±1.3 |

have been deduced using our $\delta(E5/M4) = -0.12^{+0.06}_{-0.08}$, the known $T_{1/2} = 0.80$ s,[13] and the theoretical[14] $\alpha(M4) = 0.128$ and $\alpha(E5) = 0.0577$. The values obtained are $B(M4)\downarrow = (0.2585 \pm 0.0005) \times 10^4\ e^2\,\mathrm{fm}^8$ and $B(E5)\downarrow = (161 \pm 2) \times 10^6\ e^2\,\mathrm{fm}^{10}$.

Bohr and Mottelson[15] have evaluated the single particle $M4$ transition probability $[B(M4)_{\mathrm{s.p.}} = 1.7 \times 10^4\ e^2\,\mathrm{fm}^8]$ by using the wave functions given by Blomqvist and Wahlborn.[16] This is ~6 times the experimental value obtained by us. If one assumes this transition to be pure $M4$, then $B(M4)_{\mathrm{obs.}} = 2.8 \times 10^3\ e^2\,\mathrm{fm}^8$ as quoted in Ref. 15. Thus for the single hole configurations, the observed retardation may indicate $M4$ polarization effects of the same magnitude as for the $M1$ components (Ref. 15, p. 340).

Further, the $E5$ enhancement factor is found to be $\simeq 44$ with $B(E5)_{\mathrm{s.p.}} = 3.64 \times 10^6\ e^2\,\mathrm{fm}^{10}$ (assuming statistical factor unity).

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[1]F. K. McGowan and E. C. Campbell, Phys. Rev. 92, 523 (1953).
[2]S. Gustafsson, K. Johansson, E. Karlsson, and A. G. Svensson, Phys. Lett. 10, 191 (1964).
[3]P. Kleinheinz, R. Vukanovic, L. Samuelsson, D. Krmpotic, N. Lindström, and K. Siegbahn, Nucl. Phys. A93, 63 (1967).
[4]H. E. Bosch, L. F. Gato, N. Behar, and G. J. Garcia, Phys. Rev. C 1, 242 (1970).
[5]H. J. Körner, K. Auerbach, J. Braunsfurth, and E. Gerdau, Nucl. Phys. 86, 395 (1966).
[6]C. Bargholtz, L. Eriksson, and L. Gidefeldt, Phys. Scr. 7, 254 (1973).
[7]M. Kaplan and E. J. Wilson, Phys. Rev. C 9, 1653 (1974).
[8]E. Bodenstedt, H. J. Körner, C. Günther, and J. Radeloff, Nucl. Phys. 22, 145 (1961).
[9]N. H. Lazar and E. D. Klema, Phys. Rev. 98, 710 (1955).
[10]U. Bäverstam and H. Höjeberg, Nucl. Instrum. Methods 95, 611 (1971).
[11]A. Abragam and R. V. Pound, Phys. Rev. 92, 943 (1953).
[12]H. Kopfermann, *Nuclear Moments* (Academic, New York, 1958).
[13]M. R. Schmorak and R. L. Auble, Nucl. Data B5, 3 (1971), $A = 207$.
[14]L. A. Sliv and I. M. Band, in *Alpha-, Beta-, and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland, Amsterdam, 1965), Vol. 2, Appendix 5.
[15]A. Bohr and B. R. Mottelson, *Nuclear Structure* (Benjamin, New York, 1969), Vol. 1.
[16]J. Blomqvist and S. Wahlborn, Ark. Phys. 16, 545 (1960).
[17]T. R. Gerholm, Ark. Phys. 10, 523 (1956).
[18]S. Gorodetzky, R. Manquenouille, R. Richert, and A. Knipper, J. Phys. Rad. 22, 699 (1961).
[19]Y. K. Lee and C. S. Wu, Phys. Rev. 132, 1200 (1963).
[20]A. W. Sunyar, private communication, quoted in Ref. 5.
[21]R. Rougny, J. J. Samueli, and A. Sarazin, J. Phys. Rad. 25, 989 (1964).
[22]S. Gorodetzky, N. Schutz, and A. Knipper, Compt. Rend. 262B, 1158 (1966).