A Los Alamos Model 530 Coincidence-Anticoincidence System[3] was used in this study. 2,5-diphenyloxazole[4] in a concentration of 4 g/l was the liquid scintillation solute and *p*-cymene volumes of 30 ml and 40 ml were employed. Five complete experiments were run, each at a different instrumental sensitivity. Absolute counting efficiencies were determined with C^{14}-benzoic acid as an internal standard. Mass spectrographic analyses gave a value of 6.00 ± 0.02 percent C^{14} in the carboxyl carbon of the benzoic acid. This was synthesized[5] from carbon dioxide containing 5.96 ± 0.01 percent $C^{14}O_2$. The benzoic acid specific activity was calculated from the C^{14} abundance and a half-life value of 5568 years.[6]

Experimental gross rates were corrected for background and converted to disintegration rates. The average value, from five experiments, of the specific activity of carbon in contemporary *p*-cymene was 12.9 ± 0.2 disintegrations per minute per gram. This number represents a rather startling departure from the usually quoted value of 15.3 ± 0.1 disintegrations per minute per gram of carbon in wood.[7] Such a difference is not altogether unexpected with both a new counting form and an instrument of twelve times higher efficiency than the Screen Wall Counter used by Libby and co-workers. The accuracy of either number depends on the efficiency calibration of the instrument used and not on the number of samples subsequently counted.

The liquid scintillation counter may soon be routinely used for radiocarbon dating. Further work is being carried out with methanol and methyl ether, both of which can easily be derived from carbon dioxide and which may be incorporated into the solvent system of liquid scintillators. The relatively high background of instruments such as the Los Alamos Model 530 Coincidence-Anticoincidence System is the only serious problem remaining before they can be converted to natural radiocarbon counters. Ways are being explored for lowering counting rates due to thermionic emission, light dark current, natural contamination in the apparatus, and external radiation. These will be reported at a later date.

The authors wish to thank R. L. Schuch for his valuable assistance and R. J. Kandel and W. H. Rogers for the mass spectrographic analyses.

† Work done under the auspices of the U. S. Atomic Energy Commission.
[1] Distillation Products Inc., Stock Number 83.
[2] Synthesized from propylene and toluene by the procedure of Ipatieff, Corson, and Pines, J. Am. Chem. Soc. **58**, 919 (1936).
[3] R. D. Hiebert and R. J. Watts, Nucleonics (to be published).
[4] Hayes, Hiebert, and Schuch, Science **116**, 140 (1952).
[5] D. L. Williams and A. R. Ronzio, Los Alamos Scientific Laboratory report, LADC-1385 (unpublished).
[6] Engelkemeir, Hamill, Ingrahm, and Libby, Phys. Rev. **75**, 1825 (1949).
[7] W. F. Libby, *Radiocarbon Dating* (The University of Chicago Press, Chicago, 1952), p. 16.

Crystal Structure and Nuclear Directional Correlation. I. Pb^{204}†

H. Frauenfelder, J. S. Lawson, Jr., W. Jentschke, and G. DePasquali
University of Illinois, Urbana, Illinois
(Received August 27, 1953)

THE Zürich group has recently shown in a beautiful experiment[1] that the $\gamma-\gamma$ directional correlation of Cd^{111} in a metallic indium single crystal depends markedly upon the orientation of the crystal axes with respect to the counter axes. Their experiment demonstrates that the main contribution to the effect of extranuclear fields on the directional correlation arises from the interaction between the nuclear quadrupole moment and electric field gradients.

Growing and orienting single crystals is, however, so time-consuming that the method is applicable only to nuclides with relatively long half-lives. In the case of Pb^{204} ($T_{\frac{1}{2}}=65$ min), we therefore chose a different approach to investigate the effect of crystal structure and studied the directional correlation of this isomer in Pb—Tl substitutional alloys. Pb and Tl form mixed crystals over a wide range of composition[2] and display at suitable compositions and temperatures face-centered cubic (f.c.c.), body-centered cubic (b.c.c.), and hexagonal close-packed (h.c.p.) lattices.

We determined the directional correlation function $W(\theta)$ of Pb^{204} for two types of sources at various temperatures. The first type (~100 percent Tl) was directly produced by irradiating Tl in the cyclotron with deuterons[3] and constitutes therefore a Pb—Tl crystal with vanishingly small Pb content. Such a crystal possesses an h.c.p. lattice below 235°C and a b.c.c. lattice between 235°C and the melting point 303°C. The second type (~40 percent Tl), prepared by adding Pb to the irradiated Tl, shows the f.c.c. structure of Pb and has a melting point of 365°C.

The measured anisotropies, $A=[W(180°)/W(90°)]-1$, are shown in Figs. 1(a) and 1(b) as a function of the source tempera-

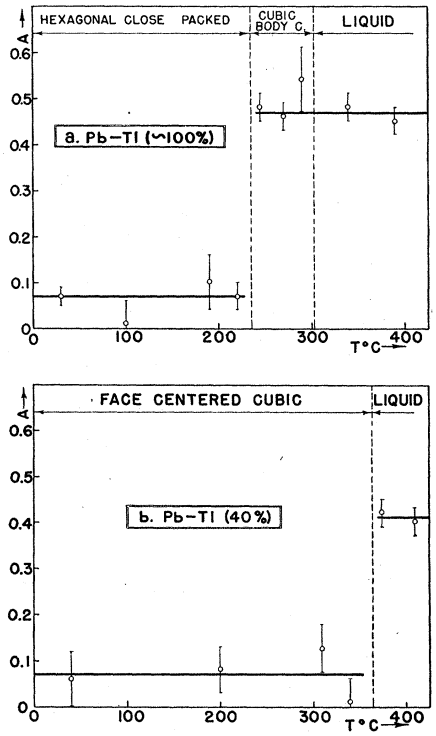

FIG. 1. Anisotropy A of the Pb^{204} $\gamma-\gamma$ cascade in Pb—Tl alloys as a function of the source temperature T. (a) 100 percent Tl. (b) 40 percent Tl.

ture T. (The indicated errors are statistical errors only. The measured anisotropies are corrected for scattering in the source[3] and the absolute values may therefore be in error by as much as a factor 1.1. However, this does not affect the relative values.)

The data in Fig. 1 show the importance of the crystal structure for the directional correlation of Pb^{204}. The highest anisotropy, 0.47 (±0.02) is obtained *both* in the solid b.c.c. lattice and in the liquid of corresponding composition.[3] The discussion of the Pb^{204} decay shows that no greater anisotropy is possible.[3] We therefore conclude that the interaction between the nucleus and its surroundings is vanishingly small in the b.c.c. lattice.

However, problems arise in the explanation of the anisotropies for the h.c.p. and the f.c.c. structures (0.07 ± 0.02, and 0.07 ± 0.03, respectively). These values are considerably lower than the lowest ("hard core") value 0.15 predicted by theory for a static quadrupole interaction in randomly oriented crystals.[4] (Attempts to show that a partial orientation of our crystals caused the low values were unsuccessful.) No value for the magnitude of the quadrupole interaction can therefore be given. The fact that the f.c.c. lattice yields the same low anisotropy as does the h.c.p. lattice is also unexplained.

The present results demonstrate the effect of crystal structure on the directional correlation of Pb^{204}. It is quite evident that more theoretical and experimental investigations are needed to clarify the major deviations from the expected behavior.

† Assisted by the joint program of the U. S. Office of Naval Research and and the U. S. Atomic Energy Commission.

[1] Albers-Schönberg, Hänni, Heer, Novey, and Scherrer, Phys. Rev. **90**, 322 (1953).
[2] Y. C. Tang and L. Pauling, Acta Cryst. **5**, 39 (1952).
[3] Frauenfelder, Lawson, Jentschke, and DePasquali (to be published).
[4] A. Abragam and R. V. Pound (to be published).

Isomerism in Pb^{206}

D. E. ALBURGER,* *Nobel Institute of Physics, Stockholm, Sweden*
AND
M. H. L. PRYCE, *Clarendon Laboratory, Oxford, England*
(Received August 24, 1953)

AN investigation of the energy levels of Pb^{206}, formed in the electron capture decay of 6.4-day Bi^{206}, has been undertaken because of theoretical interest[1] in nuclei near closed-shell lead-208. Previous results[2] were inconclusive since the transition energies could not be determined with sufficient accuracy and a number of unresolved lines were known to be present.

The following measurements on Bi^{206} have been carried out at the Nobel Institute and at the Royal Institute of Technology, Stockholm; conversion line energies and intensities in a double-focusing spectrometer and in an intermediate-image spectrometer; coincidences between pairs of K conversion lines in a two-lens "spectrogoniometer"; and coincidences between conversion lines and gamma rays observed in the intermediate-image spectrometer modified with a NaI crystal detector near the source. The energy measurements, which establish a number of cascade crossovers, together with the coincidence data impose more than 50 restrictions on the level arrangement. Except for a few very weak unassigned gamma rays the tentative level scheme adopted contains 26 transitions and satisfies all of the conditions mentioned above.

Two sets of lines which are of special interest have their relative intensities listed in Table I. Both transitions are thought to

TABLE I. Relative conversion line intensities of $E3$ isomeric transitions in Pb^{206}.

E_γ	K	L_{I+II}	L_{III}	M	N
202.5	0.084	0.57	0.25	0.22	···
516.1	18	10	~1.8	3.3	~0.6

originate from a 7− fifth excited state at 2200.3 kev going to the fourth and third levels at 1998.1 and 1683.8 kev, respectively. The latter are each assigned 4+ not only because the K to L ratios of the two transitions appear to be consistent with $E3$ but because of other features of the decay scheme. It is possible that the 7− level is analogous to that responsible for the 68-min isomer of Pb^{204}.

A measurement of the lifetime of the Pb^{206} isomeric state has been carried out by placing a Bi^{206} source very close to a NaI crystal and EMI photomultiplier detector and allowing the output pulses to trigger the sweep of a Tektronix type 511AD synchroscope. When a sweep is started by the pulse from a gamma ray which is "prompt" with respect to K capture and occurs in the upper part of the level scheme there is a chance of observing a delayed pulse from either the main isomeric transition or gammas in the lower part of the scheme. The efficiency for detecting delayed events is expected to be reasonably high since, on the basis of the assumed level scheme, 10 strong gamma-ray transitions precede the isomeric level and 5 strong ones follow it.

For optimum conditions the integral pulse-height selector was set as low as possible, i.e., to the point where the room background and noise counting rate from the shielded detector was about 150 per minute. A scope sweep of 500 microseconds (10 cm) was used and its calibration and linearity were checked by feeding in a 200-kc/sec sine wave from a G.R. type 805C signal generator and then counting the number of cycles per cm. To record the data a cardboard mask covered the base line and all other parts of the scope face except a 1.5-cm wide slot whose distance from the origin pulse could be adjusted as desired. A large diameter short focal length lens placed at a distance of 50 cm from the scope screen focused the light pulses appearing in the mask opening onto a 931 photomultiplier. The latter was provided with a long-time-constant circuit in order to smooth out the "hashy" pulse arising from the decay characteristics of the scope screen phosphor. Even with this precaution reliable operation was not attained until a one-kick multivibrator having a time constant of several milliseconds was inserted between the discriminator and scalar.

Figure 1 shows the delayed pulse distribution obtained with a source of approximately 5×10^{-4} microcurie which gave a triggering rate of 1500 counts per min. A calculated background of 2.8 counts per min has been subtracted and corrections have been made for nonlinearity of the oscilloscope trace. The slope of the curve corresponds to a half-life of 145±15 microseconds. This value is just above the region 10^{-7} to 10^{-4} sec within which no lifetimes had been observed[2] by means of the more commonly used delayed-coincidence techniques.

The bismuth sources were prepared by cyclotron bombardment of lead with 16-Mev deuterons followed by chemical separation.[2] Measurements on the conversion electron spectrum showed that a small amount of 50-year Bi^{207} and a very minute amount of 14-day Bi^{205} were present. The decay rate of the isomer was therefore studied and its intensity was observed to decrease at approximately the 6-day rate of Bi^{206}.

The 145-microsecond lifetime, which is determined almost entirely by the 516.1-kev transition, appears to be shorter than expected[3] although no multipole order other than $E3$ gives as reasonable a fit. Using the K conversion coefficients[4] of Rose *et al.*, one can compute from the conversion line intensities in Table I that the ratio of the 516.1- to 202.5-kev transition probabilities is 300 (with an error of perhaps 25 percent) if it is assumed that both are $E3$. This leads to a partial half-life of about 0.04 sec for the 202.5-kev transition. The computed ratio of unconverted gamma rays is about 1600. The fact that the ratio would be 700 on the basis of a seventh-power energy dependence[3] seems to be in agreement with the observations.

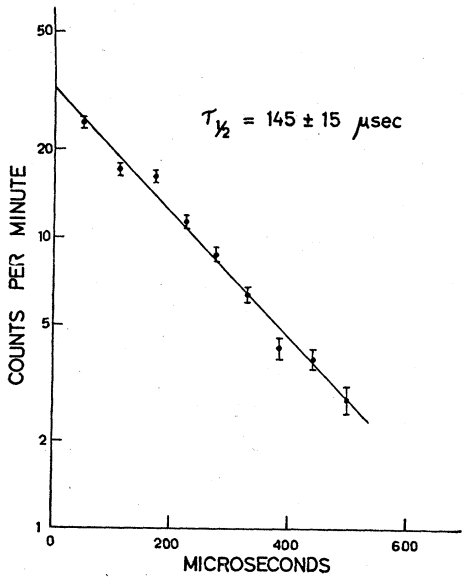

FIG. 1. Delayed pulse distribution resulting from the isomeric state of Pb^{206}.