

The resulting specific activities in terms of the time-off intensity of the 28.7-hour activity per milligram of bombarded barium were in the ratios 1.00:0.73:0.54:0.03:0.05 for the samples enriched in Ba^{134} , Ba^{132} , Ba^{135} , and Ba^{136} , and natural barium, respectively. Similar bombardments of the enriched barium isotopes in carbonate form with fast neutrons from $\text{Li}+d$ produced the 28.7-hour activity with specific activities in the ratios 1.00:0.42:0.42:0.09:0.04 for the samples enriched in Ba^{135} , Ba^{132} , Ba^{134} , and Ba^{136} , and natural barium. Bombardments of Hilger CsCl with 10-Mev deuterons yielded the well-known 38.5-hour activity⁴ of Ba^{133} in the barium fraction but no 28.7-hour activity, precluding assignment of the 28.7-hour activity to Ba^{133} or Ba^{134} as a result of the reaction $\text{Cs}^{133}(d, 2n)$ or $\text{Cs}^{133}(d, n)$. Bombardment of Hilger CsCl with 20-Mev alpha-particles did not produce the 28.7-hour activity in the barium fraction, ruling out assignment to Ba^{136} as a result of the reaction $\text{Cs}^{133}(\alpha, p)$.

On the basis of these data the 28.7-hour activity in barium is assigned to an isomeric state of Ba^{135} as a result of the reactions $\text{Ba}^{134}(d, p)$ and $\text{Ba}^{135}(n', n)$. Calculations of the cross sections for the reactions will be communicated when the isotopic analyses of the enriched barium isotopes are available.

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Upper Limit for the Lifetime of the 411-Kev Excited State of Hg^{198}

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THE β - γ -coincidences arising from the Au^{198} - Hg^{198} β -disintegration have been examined with a coincidence circuit of short resolving time in an effort to measure the mean life of the 411-kev γ -emitting state of Hg^{198} . Using the method of delayed coincidences and resolving times $2\tau_0$ less than 10^{-8} sec., the mean life is found too short to detect, in contradiction to MacIntyre's recent result¹ for the same measurement.

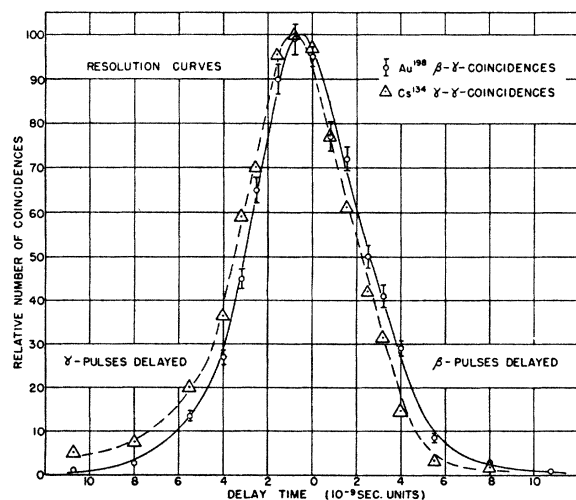


FIG. 1. Resolution curves of the coincidence unit for Au^{198} β - γ -coincidences and Cs^{134} γ - γ -coincidences.

The coincidence unit employs anthracene-1P21 scintillation counters operated with electrode voltages so high (~ 2000 v) that their output pulses do not require amplification. The pulses are equalized in Type 6AK5 pentodes and led along 95-ohm matched coaxial cable to a 50-ohm shorted coaxial cable clipper which determines their length, and thence to a 1N34 diode coincidence mixer. The pulses from either counter can be delayed by accurately known amounts by lengthening the appropriate signal cable. Checks have been made to see that the extra lengths of cable used to delay the pulses do not cause any loss of coincidences through attenuation. The selection of pulse heights and determination of counting rates in the individual counters is accomplished by feeding the individual counter pulses by separate cables into two ordinary linear amplifiers equipped with amplitude discriminators. The output pulses of these amplifiers are fed into a relatively slow (~ 2 μ sec.) triple coincidence circuit with the unselected fast coincidence pulses, and the desired coincidence rate is counted at the triple coincidence output. In this way only those fast coincidences are recorded which correspond to pulses larger than a fixed size from the individual counters, and the difficulties of very fast pulse height discrimination are avoided.

The resolution curve of the coincidence unit was measured by observing the coincidence rate as a function of delay of the pulses from each counter in turn. Results have been recorded for Co^{60} γ - γ -coincidences, Cs^{134} γ - γ -coincidences, and Au^{198} β - γ -coincidences at two different resolving times, $2\tau_0 = 9 \times 10^{-9}$ sec. and $2\tau_0 = 5.5 \times 10^{-9}$ sec. To clarify the diagrams, only the Cs^{134} and Au^{198} results at the shorter resolving time are shown. (The Co^{60} results are indistinguishable from those for Cs^{134} .) Figure 1 shows the resolution curves observed for Cs^{134} γ - γ - and Au^{198} β - γ -coincidences. The two curves are identical within experimental error and both are highly symmetrical. If the Hg^{198} 411-kev excited state had a mean life τ long enough to be detectable in this experiment, the right-hand (β -pulse delay) portion of the Au^{198} resolution curve would have a tail falling off as $\exp(-T/\tau)$, where T is the delay time.² No such tendency is visible in Fig. 1. The slight shifts ($\sim 4 \times 10^{-10}$ sec.) of the maxima of the curves of Fig. 1 with respect to each other and to the zero of time represent fixed delays due to differences in transit time of the electrons in the photomultiplier tubes at slightly different electrode voltages. As a check on this explanation, the voltage on one photo-multiplier was lowered by 30 percent: the resolution curve then moved, without change of shape, in the direction corresponding to a lag of 3.4×10^{-9} sec. in the pulses from that tube. A resolution curve was plotted for β - α -coincidences from $\text{ThC}(\beta)\text{ThC}'(\alpha)\text{ThD}$ to

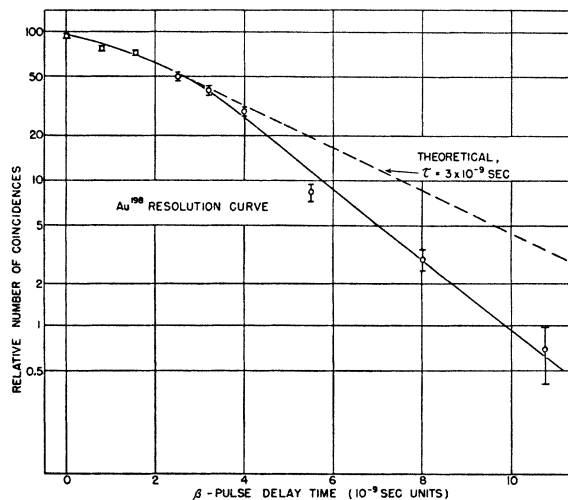


FIG. 2. β -pulse delay portion of Au^{198} resolution curve plotted on a logarithmic scale.

show that the apparatus is, in fact, capable of detecting delayed coincidences due to a finite lifetime. The shape of the resulting curve (not shown) was compatible with the 0.304- μ sec. half-life³ of ThC'.

In order to set an upper limit on the mean life τ of the 411-kev excited state of Hg¹⁹⁸, the β -pulse delay portion of the Au¹⁹⁸ resolution curve has been plotted on a semilogarithmic scale in Fig. 2. The roughly exponential fall-off of the experimental curve is common to the results for Co⁶⁰, Cs¹³⁴, and Au¹⁹⁸ and thus is instrumental. A dashed line in Fig. 2 indicates the shape the curve would have if the mean life of the Hg¹⁹⁸ excited state were as long as 3×10^{-9} sec. Figure 2 demonstrates that the mean life τ of the 411-kev excited state of Hg¹⁹⁸ is less than 3×10^{-9} sec.

An incidental result is that the excited states of Ni⁶⁰ and Ba¹³⁴ which are concerned in γ - γ -coincidences from Co⁶⁰ and Cs¹³⁴, respectively, all have mean lives shorter than 3×10^{-9} sec.

MacIntyre's result for the mean life ($2.3 \pm 0.2 \times 10^{-8}$ sec.) can probably be explained by the fact that he compared β - γ -coincidences from Au¹⁹⁸ with those from Na²⁴. The pulses from Na²⁴ γ -rays (1.38 and 2.76 Mev) would be many times larger than those from Au¹⁹⁸ γ -rays (0.411 Mev): in a coincidence system using amplifiers of finite rise time the result would be a greater lag in the detection of the Au¹⁹⁸ γ -ray pulses and consequently an apparent finite lifetime of the Hg¹⁹⁸ excited state.

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On the Spatial Alignment of Nuclei

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CONSIDERABLE interest centers on the problem of producing a system containing nuclei in a reasonably well-ordered state of alignment in space. Such a system makes possible studies of angular dependence of the scattering and absorption of polarized nucleons and of angular distributions in radioactive decay, relative to the spin axis of the nuclei.^{1,2}

The most direct method of achieving a net alignment is to place the substance in a magnetic field H_0 . If thermal equilibrium exists between the spin system and its surroundings, a Boltzmann distribution of the spins over the $2I+1$ equally spaced energy levels results, with the lowest level, corresponding to the spin nearly parallel or antiparallel to the field, becoming most heavily populated. Practical difficulties are presented by the fact that the nuclei must be put into an extremely strong field and at the same time must be in thermal contact with a paramagnetic substance in a weak field, because a reasonable degree of orientation is achieved only at the lowest temperatures so far attainable by adiabatic demagnetization, with the nuclei in the strongest field obtainable. A somewhat simpler method may be available for nuclei of paramagnetic ions, which may be oriented by the intense fields of the oriented ions themselves, at low enough temperatures.^{3,4} In both cases the sample containing the nuclei must be kept in a field while the other experiments are performed lest random orientations be resumed.

The purpose of this note is to outline an experimentally less difficult method by which a partial alignment of nuclei might be achieved. It is known that the $2I+1$ -fold degeneracy of a nucleus can be partially lifted through an interaction between the nuclear electric quadrupole moment and a crystalline electric field having axial symmetry.^{5,6} The component m_I of angular momentum along the axis becomes a good quantum number, in the absence of a strong magnetic interaction, although levels differing only in

sign of m_I are not separated. So far, because of difficulties attendant on finding suitable samples, only moderate quadrupole splittings have been found, such as the 720 kc/sec. in Al₂O₃.⁶ On the other hand, it must be expected that very much larger splittings exist in, for instance, crystals composed of molecules having p -type covalent bonds.⁷ One would expect, in fact, splittings of the order of hundreds or thousands of megacycles as in gaseous molecules⁸ because the gradient of the electric field at the nucleus is determined by the other nuclei and the electrons of the molecule itself (in contrast to the situation in ionic crystals where the ion containing a nucleus is primarily in a singlet S state and probably contributes little to the gradient of the electric field at the nucleus).

A crystal having quadrupole splittings of this order of magnitude need only be brought into thermal equilibrium with a substance that has been adiabatically demagnetized to a temperature of a few hundredths of a degree and the nuclei will fall into the lowest energy levels. This would mean either the highest or lowest absolute values of the component of the nuclear spin along the major axis of the tensor grad \mathbf{E} at the nucleus would predominate. Either case could be found depending upon the sign of the quadrupole coupling constant $eQ(\partial^2 V/\partial z^2)$. Therefore, the nuclei would be aligned, respectively, either simultaneously almost parallel and antiparallel to this axis in the molecule, or precessing nearly in a plane perpendicular to this axis. If the molecules could be crystallized in such a way that this axis was alike for all, one achieves a certain kind of nuclear orientation. A fairly sizable single crystal or some technique of orienting small crystals would give the desired result if the crystal does not contain non-equivalent orientations of the molecules, with respect to the crystal axes.

Although the kind of alignment resulting is not as unambiguous as in the magnetic alignment, several advantages are apparent. In radioactive emission, anticipated angular distributions involve even powers of $\cos \theta$,¹ for which, if true, the ambiguity is not a disadvantage. The thermal contact with the paramagnetic substance may be easier to achieve because the nuclear sample can be put into the magnetic field while the paramagnetic magnetization and adiabatic demagnetization are performed. This is so because the magnetic splittings are only small perturbations on the electric ones. If the crystal containing the nuclear sample itself could be made also to contain the paramagnetic ions, the thermal contact could be very strong, and the paramagnetic ions, by their relaxation processes, could help to bring about thermal relaxation of the nuclear system. Once thermal equilibrium is established the sample may be completely removed from the magnetic field, simplifying some of the experiments on the aligned nuclei.

To find such a high frequency splitting and to demonstrate the temperature of the spin system, ordinary nuclear paramagnetic resonance experiments at moderate frequencies can be of assistance. With an odd half-integral spin, one could observe the resonance absorption in the transition between the still degenerate levels $m_I = \pm \frac{1}{2}$, when the degeneracy is removed by a magnetic field. If the axis of symmetry of the electric field lies along the magnetic field, one expects to find a resonance line at the frequency corresponding to the normal gyromagnetic ratio, but, if the quadrupole splitting is strong, the frequency would be very sensitive to a small angular displacement from this position. By a perturbation calculation, regarding the Zeeman energy as a small perturbation on the large quadrupole energy, one can find the amount of the quadrupole splitting.

If the intensity of the line is observed as the temperature is reduced to such a degree that $kT < h\nu$ where ν is the quadrupole splitting frequency, either of two things will occur, determined by the sign of the quadrupole coupling constant. If the $m_I = \pm \frac{1}{2}$ levels are the lowest, then the line would get more intense as $1/T$ until a critical temperature is reached where a fairly pronounced increase in intensity would accompany the preferential population of these states. If $m_I = \pm I$ are the lowest, the observed line would go through a maximum and then decrease as the $m_I = \pm \frac{1}{2}$ levels become depopulated.