magnetic field. The results for tin and thallium suggested that one should seek other properties that are very nearly alike for these two elements with the hope that some general empirical relation might be obtained. It was found that the ratio of $T_c/\Theta$, where $\Theta$ is the Debye characteristic temperature, was very nearly the same for tin and thallium. Combining this information with the relation previously given by Steele[2]

$$E_0=\gamma T_c^2/3e \tag{1}$$

(where $\gamma$ is the electronic specific heat in the normal state, $e$ is the electronic charge, and $E_0$ is the value of $E$ at the absolute zero of temperature) suggested that

$$E_0\sim(T_c/\Theta)^2 \tag{2}$$

and

$$\gamma\sim\Theta^{-2}. \tag{3}$$

The $\gamma$ appearing in Eq. (1) may not be identical to the $\gamma$ determined from equilibrium phenomena such as specific heat.[1]

The correlation indicated by Eq. (2) can be tested by the recent thermoelectric data. In Table I the pertinent data is given for

TABLE I. Data for the superconductors.

| Element | $E_0\times10^3$ volts | $T_c$°K | $\Theta$°K | $(E_0\Theta^2/T_c^2)\times10^3$ volts |
|---|---|---|---|---|
| Lead | 59.0 | 7.26 | 90[a] | 9.1 |
| Indium | 10.4 | 3.41 | 106[b] | 10.0 |
| Tin | 5.5 | 3.72 | 150[c] | 9.0 |
| Thallium | 5.5 | 2.40 | 96[a] | 8.8 |

[a] J. de Launay and R. L. Dolecek, Phys. Rev. **72**, 141 (1947).
[b] Value calculated from data of J. R. Clement and E. H. Quinnell, Phys. Rev. **79**, 1028 (1950), by assuming that the specific heat in the normal state can be represented by a $T^3$ law plus a linear term in $T$.
[c] Value calculated from the combined data of W. H. Keesom and P. H. Van Laer, Physica **5**, 193 (1938); and W. H. Keesom and J. N. Van den Ende, Proc. Acad. Sci. Amst. **35**, 143 (1932); by assuming that the specific heat can be represented by a $T^3$ law plus a linear term in $T$.

lead, indium, tin, and thallium. It is seen that the values of $E_0\Theta^2/T_c^2$ for the particular elements are less than 10 percent different from the mean value of $9.2\times10^{-3}$ volt. In the light of the experimental limitations on the value of $E_0$ (reported[1] as subject to an error of the order of 10 percent) and the spread in possible values of $\Theta$, it is felt that the data for these four elements substantiates the correlation given by Eq. (2).

Preliminary measurements with mercury ($T_c=4.16$°K) have shown that $E_0$ for that element is about $250\times10^{-3}$ volt. In order that this be consistent with the results for the other elements and the relation given by Eq. (2), it is necessary that $\Theta$ for mercury should be about 25°K. Although this $\Theta$ is very much smaller than the value obtained from specific heat measurements ($\Theta=96$°K), it is in fair agreement with the value given by resistance measurements ($\Theta=37$°K).

Additional thermoelectric measurements are needed to test Eq. (2) further. For example, aluminum, with a $\Theta\sim400$°K, would have $E_0\sim10^{-9}$ volt if Eq. (2) were applicable. Such measurements are now in progress at this Laboratory. The results will be reported at a later date.

[1] M. C. Steele, Phys. Rev. **81**, 262 (1951).
[2] M. C. Steele, Phys. Rev. **78**, 308 (1950).

## Further Search for Nuclear Transitions with Lifetimes between $3\times10^{-9}$ sec and $10^{-7}$ sec*

W. E. WRIGHT† AND M. DEUTSCH
*Laboratory for Nuclear Science and Engineering and Department of Physics Massachusetts Institute of Technology, Cambridge, Massachusetts*
(Received February 26, 1951)

THE method of delayed coincidences has been utilized in a search for nuclear metastable states with lifetimes less than $10^{-7}$ sec. Scintillation counters, using stilbene or anthracene with RCA 5819 photomultiplier tubes, were used with wide band amplifiers of a design due to Elmore and a fast electronic coincidence circuit. Under favorable circumstances a half-life of $3\times10^{-9}$ sec could be detected with this apparatus. In most cases studied, the lifetimes were too short to be measured by this apparatus, and only upper limits could be set. Typical curves of coincidence counting rate *vs* delay and some of the results have been reported previously.[1]

Coincidence absorption studies were undertaken in order that an upper limit on the half-life of a specific transition might be set, even though only prompt coincidences were observed. The detectors were so arranged that both single- and double-channel coincidence absorption experiments could be conducted. Electromagnetic radiation was identified by comparing the observed half-thickness values with the curves published by Glendenin.[2] For the identification of electronic radiation, an empirical range-energy curve was constructed from the observed absorption curves for $Dy^{165}$, $Re^{188}$, and $Au^{198}$. These isotopes were selected because of the simplicity and well-known character of their decay schemes, and because of the wide range in energy of their radiations. In most cases, a determination of the energy and reference to the literature permitted a differentiation between the preceding and succeeding radiations.

The results are summarized in Table I. Column 1 lists the

TABLE I. Observed upper limits on the half-life of excited states.

| Daughter isotope | Energy (kev) | Observed $T_{\frac{1}{2}}\times10^9$ sec |
|---|---|---|
| $Br^{80}$ | 37 | <4 |
| $Xe^{131}$ | 82<br>284 | <4 |
| $Gd^{152}$ | 120<br>370 | <3 |
| $Ho^{165}$ | 91 | <5 |
| $W^{182}$ | 114<br>310 | <4 |
| $Os^{188}$ | 160 | <5 |
| $Pt^{192}$ | 240 | <3 |
| $Hg^{199}$ | 160 | <3 |

daughter isotopes of the decays investigated. Column 2 lists the energy in kev of the transition observed. Where two energies are listed for a single isotope, it has not been possible to differentiate between the preceding and succeeding radiation. Column 3 gives in units of $10^{-9}$ sec the upper limits for the half-life observed for the transitions listed in columns 1 and 2.

The authors wish to thank Professor J. W. Irvine, Jr., and the members of the MIT Cyclotron Group for their advice and assistance in the preparation of the radioactive sources required for these studies.

* This work was supported in part by the joint program of the AEC and ONR.
† Now at ONR Washington, Washington 25, D. C.
[1] M. Deutsch and W. E. Wright, Phys. Rev. **77**, 139 (1950).
[2] L. E. Glendenin, Nucleonics **2**, No. 1, 12 (1948).

## Delayed Coincidence Angular Correlations

STUART P. LLOYD
*Department of Physics, University of Illinois, Urbana, Illinois*
(Received February 26, 1951)

WE consider the directional correlation of successive particles emitted in the nuclear cascade $j_1\rightarrow j\rightarrow j_2$, where the $j$'s are the angular momenta of the nuclear levels, when the source is placed in a constant homogeneous magnetic field. The existence of remarkable nuclear spin phase memory phenomena[1] in nonparamagnetic solutions leads one to believe that it might be possible to determine the gyromagnetic ratio of the intermediate

nucleus by observing the angular correlation of the emitted particles as a function of time.[2] One would expect a correlation to persist to times of the order of the total spin relaxation time $T_2$ of the intermediate nuclei $j$ in the sample, perhaps milliseconds or better.[3] We show here how the theoretical delayed coincidence correlation function describing the effect of the magnetic field is to be obtained from the usual correlation function.

The equations of the Weisskopf-Wigner treatment of the decay[4] can be made to yield a function $W(\Omega_1;\Omega_2;t-t')$ which can be interpreted as the rate (steradian$^{-2}$ sec$^{-1}$) at which particles of the second transition are emitted at time $t$ with direction $\Omega_2\equiv\theta_2,\varphi_2$ when the preceding particle in the cascade was emitted at time $t'$ with direction $\Omega_1\equiv\theta_1,\varphi_1$, the initial nuclei $j_1$ being randomly oriented at time zero. As indicated, this rate comes out to be a function of $t-t'$ only, and we refer to it as the delayed coincidence angular correlation function.[5] If $\omega_0$ is the circular Larmor frequency of the intermediate nucleus $j$ in the external magnetic field (along the $z$-axis), then the delayed coincidence correlation is given by

$$W(\theta_1,\varphi_1;\theta_2,\varphi_2;t)=4\pi\tau_1\exp(-t/\tau)\times\Sigma_{mm'}X_{mm'}(\theta_1,\varphi_1;\theta_2,\varphi_2)\exp(i\omega_0(m-m')t), \quad (1)$$

where

$$X_{mm'}(\theta_1,\varphi_1;\theta_2,\varphi_2)=(2j_1+1)^{-1}(2\pi/\hbar)^2\rho_1\rho_2\mathcal{S}\Sigma_{m_1m_2}\times[(j_2m_2|\Omega_2,\sigma_2|jm)(jm|\Omega_1,\sigma_1|j_1m_1)\times(j_2m_2|\Omega_2,\sigma_2|jm')^*(jm'|\Omega_1,\sigma_1|j_1m_1)^*]. \quad (2)$$

In Eq. (1), $\tau_1$ and $\tau$ are the lifetimes of the initial and intermediate nuclei, respectively, and in Eq. (2), $\mathcal{S}$ is the invariant summation over the spin coordinates $\sigma_1$ and $\sigma_2$ of the emitted particles, and $\rho_1$ and $\rho_2$ are densities-in-energy of the states of the emitted particles. The usual instantaneous angular correlation[6] is

$$W(\theta_1,\varphi_1;\theta_2,\varphi_2)=\tau W(\theta_1,\varphi_1;\theta_2,\varphi_2;0)=4\pi\tau\tau_1\Sigma_{mm'}X_{mm'}(\theta_1,\varphi_1;\theta_2,\varphi_2), \quad (3)$$

normalized to $\int W(\theta_1,\varphi_1;\theta_2,\varphi_2)(d\Omega_1/4\pi)d\Omega_2=1$. It is actually a function only of the angle $\theta$ between $\Omega_1$ and $\Omega_2$, of course.

The transformation properties of the nuclear matrix elements[7] can be used to obtain

$$X_{mm'}(\theta_1,\varphi_1;\theta_2,\varphi_2)=X_{mm'}(\theta_1,\varphi_1;\theta_2,\varphi_2+\alpha)\exp(i\alpha(m'-m))$$

for rotations of $\Omega_2$ by angle $\alpha$ around the $z$-axis. If this is substituted into Eq. (1) with $\alpha=\omega_0t$, one discovers that

$$W(\theta_1,\varphi_1;\theta_2,\varphi_2;t)=(1/\tau)\exp(-t/\tau)W(\theta_1,\varphi_1;\theta_2,\varphi_2+\omega_0t). \quad (4)$$

The delayed coincidence angular correlation pattern is just the instantaneous pattern precessing with the intermediate nucleus around the external field.[8] If the instantaneous correlation is given as a power series in $\cos\theta$, the corresponding delayed coincidence correlation is obtained as a fourier series in $\omega_0t$ by transforming the instantaneous correlation to a Legendre series:[9] $W(\theta)=\Sigma\gamma_lP_l(\cos\theta)$, and then applying the spherical harmonic addition theorem[9] to

$$P_l(\cos\theta)=P_l(\cos\theta_1\cos\theta_2+\sin\theta_1\sin\theta_2\cos(\varphi_1-\varphi_2)).$$

For example, the instantaneous correlation $W(\theta)=1+r\cos^2\theta=1+(r/3)+(2r/3)P_2(\cos\theta)$ gives the delayed coincidence correlation (omitting the decay and normalization factors):

$$W(\theta_1,\varphi_1;\theta_2,\varphi_2;t)=1+(r/3)+(r/6)(3\cos^2\theta_1-1)(3\cos^2\theta_2-1)+(r/2)\sin2\theta_1\sin2\theta_2\cos(\varphi_1-\varphi_2-\omega_0t)+(r/2)\sin^2\theta_1\sin^2\theta_2\cos2(\varphi_1-\varphi_2-\omega_0t). \quad (5)$$

In general, the odd harmonics of $\omega_0t$ drop out if either counter is in the equatorial plane, $\theta_1$ or $\theta_2=90°$; and there are no oscillating terms at all if either counter is on the axis, $\theta_1$ or $\theta_2=0°$ or 180°. The relative amplitudes of the various harmonics are complicated functions of $\theta_1$ and $\theta_2$, in general, and the best position for the counters has to be worked out for each case separately. For the correlation (5), one maximum of the first harmonic amplitude relative to the non-oscillating terms occurs when both counters are on the cone:

$$\cos^2\theta_1=\cos^2\theta_2=(1/2)-(r/(8+6r))+O(r^3).$$

It is to be noted that both the instantaneous correlation and the gyromagnetic ratio of nucleus $j$ can be determined by measuring the distribution of pulses in time in one counter; e.g., if the counter is in the equatorial plane, $W(90°,\varphi;90°,\varphi;t)=(1/\tau)\exp(-t/\tau)W(\omega_0t)$, where $W(\theta)$ is the instantaneous correlation.

If the highest power of $\cos\theta$ in the instantaneous correlation is $\cos^L\theta$, the delayed correlation contains harmonics up to $\cos(L\omega_0t)$, and the total gate width $\Delta t$ should be narrow enough so that these harmonics are not washed out by averaging over the gates: if $L\omega_0\Delta t\geq1$, gate width correction factors must be introduced. For the same reason, the variation $(T_2^*)^{-1}$ of the Larmor frequency due to inhomogeneities in the applied magnetic field should be kept small compared to $(L\tau)^{-1}$.

I wish to thank Professor A. T. Nordsieck for a discussion of the Weisskopf-Wigner method and Dr. C. P. Slichter for discussions on the nature of the fields in material media.

[1] E. L. Hahn, Phys. Rev. **80**, 580 (1950). The spin echo effect involves free Larmor precessions of protons in water of up to $10^6$ revolutions at 30 Mc.
[2] Sunyar, Alburger, Friedlander, Goldhaber, and Scharff-Goldhaber, Phys. Rev. **79**, 181 (1950).
[3] Bloembergen, Purcell, and Pound, Phys. Rev. **73**, 679 (1948).
[4] V. Weisskopf and E. Wigner, Z. Physik **63**, 54 (1930).
[5] G. Goertzel deals essentially with $\int_0^\infty W(\Omega_1;\Omega_2;t)dt$ in his investigation of the effect of hyperfine structure interactions in $\gamma-\gamma$-angular correlations; Phys. Rev. **70**, 897 (1946). We assume that there are no such fields at the nucleus in the liquid sample.
[6] D. L. Falkoff and G. E. Uhlenbeck, Phys. Rev. **79**, 323 (1950).
[7] S. P. Lloyd, Phys. Rev. **80**, 118 (1950).
[8] This was suggested by Professor M. Goldhaber (private communication).
[9] The necessary formulas can be found in W. E. Byerly, *Fourier Series and Spherical Harmonics* (Ginn and Company, 1893), pp. 179, 211, 199.

## Electron Density and Light Intensity Decay in Cesium Afterglows*

P. DANDURAND† AND R. B. HOLT
*Department of Physics, Harvard University, Cambridge, Massachusetts*
(Received February 20, 1951)

THE decay of the electrodeless discharge through purified cesium has been studied by the previously described techniques[1] of microwave electron density, optical spectrographic, and optical light intensity measurements as a function of time after termination of the exciting fields. In the pressure range 0.02 to 0.1 mm, we found that the electron density, $n$, decays exponentially with a time constant, $\tau_1$, increasing with pressure from 0.4 millisecond at 0.02 mm to about 1 millisecond at 0.1 mm. A minimum in the light intensity is found at 40 microseconds after cut-off, followed at about 80 microseconds by a broad maximum. The ratio $I_{max}/I_{min}$ is about 8 at the lowest pressure and decreases with pressure. Higher input power during the discharge moves the maximum to shorter times. After the maximum, the light intensity decay is exponential with an associated time constant $\tau_2$ consistently closely equal to $\tau_1/2$, which shows that the light intensity is proportional to $n^2$. Spectrograms show an intense line spectrum during the discharge, the usual CsI spectrum coexisting with a great number of other lines belonging to the excited cesium ion. In the afterglow, however, the spectrum is practically a pure band spectrum, which was not analyzed in detail, but which shows up in the same wavelength range as the bands found previously in absorption in cesium vapor.[2]

The low pressure electron density data follows the behavior expected for ambipolar diffusion quite well. In order to compare the ion mobility data for cesium ions through cesium vapor with previous measurements[3] of the mobility of cesium ions through rare gases, the electron temperature at moderate times in the afterglow at a pressure of 0.036 mm was estimated by a standard kinetic theory calculation as 1400°K. On this basis, our data gives a value of the mobility, $k_0$, of about 50 cm$^2$/volt-sec calculated for 1-mm pressure and 0°C. This value is rather surprising, since the measured values of the mobility of cesium ions through rare