

with 1/4 μg terbium carrier added before elution. The equipment and method used in the separations were essentially as described previously in the work by Thompson *et al.* on the new element berkelium.² Two column separations were made in order to remove all of the gadolinium, the first column of 1.0 cm diameter, the second of 2 mm diameter. One-fourth μg of samarium nitrate in solution was added to the active terbium fraction before mass spectrographic separation to serve as an internal mass standard. The products of the mass spectrographic separation in the 60° slit-type mass spectrograph, using a thermal ionization source from a tungsten filament, were collected on an Eastman III-0 photographic plate. Before development this plate was left face to face with an Eastman NTA transfer plate for a day. After development the collection plate showed solid lines at the mass numbers corresponding to all the stable samarium isotopes (as both Sm^+ and SmO^+) and to stable terbium (as Tb^+). The transfer plate was searched with a microscope for alpha-tracks. A concentration of alpha-tracks was observed on the transfer plate only in a region corresponding to mass 149. The 4-hr. terbium alpha-activity was the only alpha-activity here present in large enough amount to be detected by this technique. Tb^{149} has 84 neutrons and would thus be expected to have the maximum alpha-decay energy of the terbium isotopes in this mass region.

The suggestion was also made in the previous communication¹ that the long-known natural radioactivity of samarium might be assigned to Sm^{147} or Sm^{148} , rather than to Sm^{149} , since this assignment would be more consistent with decay toward the stable configuration of 82 neutrons (on this basis the most alpha-unstable samarium isotope would be Sm^{146} , and its anomalous absence from nature would thus be explained). This hypothesis has also been tested and it has been found that a major part of this natural radioactivity should indeed be assigned to Sm^{147} . The possibility that there may be also some alpha-radioactivity of nearly the same energy associated with Sm^{148} has not been ruled out.

A sample of isotopically pure Sm^{147} was obtained by ion exchange chemical separation from an 0.8 mg amount of Pm^{147} (a beta-emitter variously reported as 2.26-yr.³ or 3.7-yr.⁴ half-life) which had been allowed to decay for about a year. This samarium sample was analyzed in an optical spark spectrograph to establish its chemical purity, in the mass spectrograph to prove the material was isotopically pure mass 147, and was examined for alpha-activity in an ionization chamber with differential pulse-height analyzer, which affords an alpha-energy determination in addition to detection. Alpha-activity of the same energy as that of the natural samarium alpha-activity was observed. The specific alpha-activity of this small sample of Sm^{147} has not yet been accurately determined, but it is roughly of the order of magnitude to account for the total alpha-activity observed in natural samarium.

Dempster⁵ had previously come to the conclusion that the natural samarium alpha-activity should be assigned to Sm^{147} on the basis of a continuance of his work using isotopic separation with the mass spectrograph together with the photograph technique for detection.

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¹ Thompson, Ghiorso, Rasmussen, and Seaborg, *Phys. Rev.* **76**, 1406 (1949).

² Thompson, Cunningham, and Seaborg, *J. Am. Chem. Soc.* **72**, 2798 (1950).

³ Inghram, Hayden, and Hess, *Phys. Rev.* **79**, 271 (1950).

⁴ J. A. Seiler and L. Winsberg, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York), National Nuclear Energy Series, Plutonium Project Record, Vol. 9B (to be published).

⁵ A. J. Dempster, tentative result given in Argonne National Laboratory Report ANL-4355 (October, 1949), unpublished.

Magnetic Shielding Constant of H_2

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RECENT investigations of the magnetic shielding effects in the H_2 molecule¹⁻³ have shown that, to calculate $-\Delta H/H$, the fractional change of the magnetic field H at a nucleus due to the electrons, it is necessary to know the average value of r^{-1} . Here r is the distance of one of the electrons from the nucleus considered and the average must be taken over the electron distribution of the ground state of H_2 .

The electronic wave function derived previously by the author⁴ is used to determine average r^{-1} as a function of the internuclear distance R . The results are given in Table I.

Averaging these values over the zero-point vibration of the molecule gives average $r^{-1} = 0.904 \pm 0.003$. The error is estimated following the procedure used in reference 4 to determine the error in the electric field gradient. Further assurance of the reliability of the estimated limits of error is obtained by comparison with the known error of the electron energy⁴ calculated from this wave function. Since average r^{-1} is $\frac{1}{4}$ of the potential energy of the molecule excluding the mutual repulsion of the two electrons, it is one of the major contributions to the energy of the molecule. The percentage error given for average r^{-1} is comparable to the percentage error in the energy of the electron distribution, as one would expect it to be.

TABLE I. Average r^{-1} as a function of the internuclear separation R . r^{-1} and R are given in atomic units.

R	Average r^{-1}
1.2	0.982
1.3	0.946
1.4	0.915
1.5	0.886

The formula for the magnetic shielding constant of H_2 as given by Ramsey² is

$$\sigma = -\Delta H/H = \frac{1}{3}\alpha^2(\text{average } r^{-1}) - (\alpha^2 a_0 R^2 / 6\mu_N) [(2Z\mu_N/R^3) - (\mu' H_r / MJ)].$$

The notation is essentially that used by Ramsey (Eq. (24) of reference 2).

A new value $(3.21 \pm 0.01) \times 10^{-5}$ obtained for the first term of σ is to be compared with the value 3.24×10^{-5} given by Anderson¹ using Nordsieck's wave function⁵ and the value 3.16×10^{-5} by Hylleraas and Skavlem.³ The latter two values are those at the equilibrium value of R and are not averages over the molecular vibration. This explains the difference between the present value and Anderson's; the present calculation giving 3.25×10^{-5} at the equilibrium position $R = 1.4$. The Hylleraas and Skavlem value was obtained from a much less accurate wave function.

The second term of σ is given by Ramsey as $(0.56 \pm 0.01) \times 10^{-5}$, the error being only that due to the experimental error in H_r . Anderson⁶ has pointed out that the molecular vibration contributes an additional source of error in this term. A correction for this is less easily made here than in the first term because H_r is not known as a function of R . An experimentally measured value of H_r , which itself corresponds to an average over the molecular vibration, is known, however. A theoretical expression² for H_r is composed of integrals containing the factor r^{-3} . For this reason, we assume that $H_r(R)$ varies as R^{-n} , choose $n \sim 3$, and require that the average of $H_r(R)$ be 13.66 as given by Ramsey.

The average value of the second term of σ calculated in this way becomes 0.55×10^{-5} . Because of the uncertainty of the assumptions made above, the error of this is taken to be 5 percent, which would correspond to an error in n of about ± 1.5 .

The resulting expression for σ is

$$\begin{aligned}\sigma &= (3.21 \pm 0.01) \times 10^{-6} - (0.55 \pm 0.03) \times 10^{-6} \\ &= (2.66 \pm 0.03) \times 10^{-6}.\end{aligned}$$

The error in σ given above is smaller than is necessary in order to correct any experimental data obtained thus far. Greater accuracy than this will undoubtedly be necessary, however, for some future experiments.

- ¹ H. L. Anderson, Phys. Rev. **76**, 1460 (1949).
² N. F. Ramsey, Phys. Rev. **78**, 699 (1950).
³ E. Hylleraas and S. Skavlem, Phys. Rev. **79**, 117 (1950).
⁴ G. F. Newell, Phys. Rev. **78**, 711 (1950).
⁵ A. Nordsieck, Phys. Rev. **58**, 310 (1940).
⁶ H. L. Anderson, private communication.

Scintillation Decay Times*

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THE decay curves for the scintillation of some molecular crystals, excited by nuclear γ -rays, have been determined using a delayed coincidence apparatus. The results are given in Fig. 1 and Table I.

In the experimental arrangement, one or two 1P21 photo-multipliers were used to detect the photons emitted by the crystals. The output from the ninth dynode of the multipliers was fed through RG 7/U coaxial cables (used as delay lines) and amplified in distributed amplifiers before being applied to a germanium diode coincidence circuit. The photo-multipliers were either excited by separate crystals or by one crystal placed between them. The decay curves were measured by varying the relative length of the RG 7/U cables in the two channels.

By varying the bias on the coincidence discriminator, one can select coincidences due to a superposition of one, two, three, etc., photon pulses in the non-delayed channel with the pulses in the delayed channel. For delays exceeding the width of the pulses at the input of the coincidence circuit, the coincidence rate is then given by $\sum_{n \geq 1} A_n \exp(-nt_d/\tau)$. τ is the decay time of the scintillation, t_d is the time delay, n is essentially the number of single photon pulses contributing to the recorded coincidence pulse from the non-delayed channel, and A_n are constants independent of t_d . The present experiment was so arranged as to record coincidences due to three or more photons only. This was done in order to obtain an effective discrimination between the true coincidence pulses and large single pulses feeding through the coincidence circuit. Furthermore, this procedure leads to an increased resolving power in delayed coincidence experiments. The half-width of the delayed coincidence curve for radiations following each other in instantaneous succession is independent of the pulse sizes in the two channels, and the coincidence rate decays with a time constant which is three times the lifetime of the crystal. The contribution from coincidences due to relatively large pulses being generated by one or two photons in the non-delayed channel or pulses due to four or more photons was observed for lower, respectively higher discriminator bias settings. In Fig. 1, the measurements corresponding to the term $\exp(-3t_d/\tau)$ are shown. The crystals and the photo-multipliers were operated at room temperature ($\pm 24^\circ\text{C}$). In order to obtain an estimate of the width of the pulses due to single electrons released from the cathode (curve F, Fig. 1), self-coincidences of thermal noise pulses from one multiplier were recorded or, alternatively, a strong Co^{60} γ -ray source was inserted between the multipliers.

The 1,4-diphenylbutadiene and the *para*-terphenyl were used in the form of microcrystalline powder¹ in Lucite containers. The luminescence of the Lucite was negligible. The *trans*-stilbene,² anthracene,³ and naphthalene⁴ were available as transparent crystals.

The average lifetimes of the excited crystals, being three times the decay periods deduced from the curves in Fig. 1, are listed in Table I.

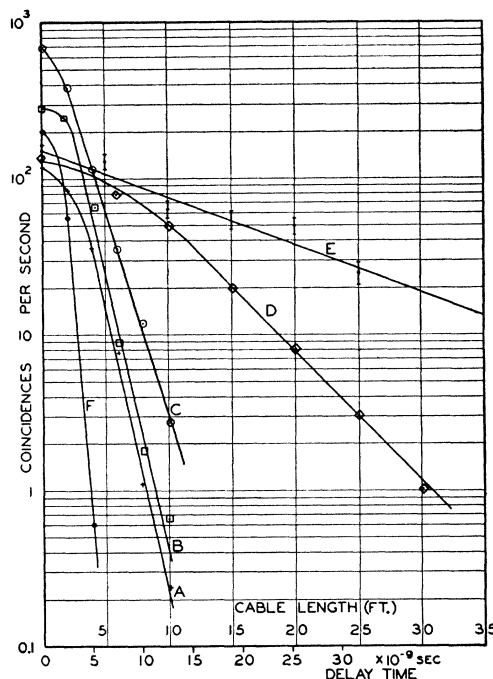


Fig. 1. The decay curves $\exp(-3t_d/\tau)$ of scintillations from: A: 1,4-diphenylbutadiene, B: *p*-terphenyl, C: *t*-stilbene, D: anthracene, and E: naphthalene. Curve F represents noise pulses. The ordinates of curve E have been multiplied by a factor of 1000. This curve also runs through a point with coordinates (55 ft., 0.004 ± 0.001 sec.⁻¹).

TABLE I. Decay times of scintillation crystals.

Crystal	$\tau(1/e)$ in 10^{-9} sec.
<i>p</i> -terphenyl	4.2
1,4-diphenylbutadiene	4.2
<i>t</i> -stilbene	5.7
anthracene	24
naphthalene	60

The curvature of the decay curves near the ordinate axis in Fig. 1 is caused either by saturation of the multipliers or by a superposition of pulses due to several photons. This was demonstrated by recording triple coincidences between multiplier anode pulses of different integral charge and the fast double coincidences. For the large pulses, the curvature was much more predominant while for the small pulses it disappeared.

The width of the pulses due to single electrons liberated from the cathode of the photo-multiplier (curve F, Fig. 1) is apparently determined by the rise time of the distribution amplifiers (2.6×10^{-9} sec.). This was borne out by experiments without amplifiers preceding the coincidence circuit, giving a pulse width $\lesssim 10^{-9}$ sec., which is in general agreement with the value calculated by Sard⁵ and with the experimental results of Post and Shiren.⁶ The decay constants reported here are all in the expected range of agreement with the values calculated by Kasha⁷ from optical data.

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¹ Obtained from Larco Nuclear Instrument Company.

² Grown by E. Tajima using stilbene purified from that obtained from Eastman Kodak Company.

³ Obtained from the Harshaw Chemical Company.

⁴ Reagent Merck's naphthalene recrystallized about seven times from ethyl alcohol. Obtained from M. Kasha.

⁵ R. D. Sard, J. App. Phys. **17**, 768 (1946).

⁶ R. F. Post and N. S. Shiren, Phys. Rev. **78**, 80 (1950).

⁷ M. Kasha, *Scintillation Counters. Qualitative Treatment of the Molecular Optical Parameters* (to be published).