Harwell 200-kev $D-D$ source. Because of the low values of the fission cross sections concerned, long irradiations ($\sim$8 hours) were necessary and, as both types of emulsion were used under conditions where they recorded protons of 2.5-Mev energy, a large background of recoil protons and natural radioactive α-particles was present in the processed emulsions. A high rate of chance superposition of tracks was therefore to be expected. However, since the maximum range of the recoil protons was 56μ while the maximum α-particle range in the uranium case was 39μ and in the thorium case 48μ, it was certain that any fission events having light fragments of range in excess of 60μ could not be due to a chance juxtaposition of tracks.

As normal isotopic uranium was used in the experiment, the plates loaded with this material were wrapped in thin cadmium foils to reduce the chance of slow neutron fission in the U-235 component resulting from the slight background of scattered neutrons. The fission cross sections of U-235 and U-238 at 2.5 Mev are such that fission events resulting from the rarer isotope could not be more than a percent or two of those resulting from the U-238. Search of the uranium loaded emulsions yielded 8 cases of ternary fission with the emission of a light, charged fragment of range greater than 60μ among 12,000 binary fissions. The greatest range observed was 202μ. In addition there were 8 events having light particles lying in the range interval 20 to 60μ and which, in the light of experience gained in earlier experiments,[1,4,5] appeared to be ternary fission.

In the case of the thorium-loaded emulsion 14,000 fission events were examined; all of these were fast neutron induced since the threshold is at $1\cdot 1$ Mev. Seven events were found having light fragments of range 60μ and five where the range of the light fragment lay between 20 and 60μ. The longest range observed in this case was 243μ.

The general character of the events in all cases was similar to those observed in the slow neutron[1] and photofission experiments[4,5] i.e., the light particle appears to be an α-particle and is emitted preferentially near to 90° from the heavy fragments.

Deductions as to the relative frequency of binary and ternary fission in these experiments cannot be drawn with confidence since, with such a high background of tracks, it is not certain that the observers would find all ternary events. However, it appears that the frequency of ternary fission is similar to that observed in the slow neutron fission of U-235.

These experiments therefore throw further doubt on the theory advanced by Tsien[6] which suggests that the emission of the long-range fragments is connected with the excitation of the compound nucleus, being less favored the greater the value of the excitation energy.

---

* Part of this work was carried out while the author was still at AERE, and thanks are due to the Director, Sir John Cockcroft, for permission to use this material.

[1] E. W. Titterton, Nature (to be published).

[2] Tsien, Ho, Chastel, and Vigneron, J. phys. et radium **8**, 165 (1947).

[3] Tsien and Faraggi, Compt. rend. **225**, 294 (1947).

[4] E. W. Titterton and F. K. Goward, Phys. Rev. **76**, 142 (1949).

[5] E. W. Titterton and Brinkley, Phil. Mag. **41**, 500 (1950).

[6] S.-T. Tsien, J. phys. et radium **9**, 6 (1948).

Energy Storage and Light Stimulated Phosphorescence in Activated NaCl Crystals Induced by Gamma-Rays*

HARTMUT KALLMANN AND MILTON FURST
New York University, New York, New York

(Received June 6, 1951)

IN a previous paper[1] experiments on fluorescence, phosphorescence, and light stimulation in sodium chloride crystals activated with 1 percent silver chloride were described. Three different decay times occur with these crystals. First, immediately after excitation by high energy radiation, the emitted light decays quite slowly with a period (designated by lifetime of instantaneous phosphorescence) of minutes, hours, or even days, depending on the period, rate, and type of high energy radiation. Second, if such a crystal is then irradiated with light of the near ultraviolet or the visible region, even after its phosphorescent light emission has considerably decreased, a new light emission is stimulated mostly in the ultraviolet region around 2400 to 3000A which again decays rather slowly after the stimulating light has been switched off (designated by lifetime of stimulated phosphorescence). Third, if the stimulation of the crystal is deferred for a considerable period of time it should be expected that the amount of stimulated light decreases since the storage qualities of the crystal may not be perfect (designated by lifetime of storage).

This letter is concerned principally with experiments trying to link these processes to the phenomenon of color centers in alkali-halide crystals. It was found that the lifetime of instantaneous phosphorescence and that of stimulated phosphorescence are closely connected to each other and probably originate from the same energy levels. The lifetime of the instantaneous phosphorescence increases considerably with increasing time of irradiation applied at the same rate. This lifetime is also considerably longer if the same total amount of high energy radiation is applied during a long period than during a short period of irradiation. The same is true for the phosphorescence of the stimulated light. If the same total amount of stimulating light is applied during a short period. the decay of the stimulated light is shorter than the decay when the same stimulation is applied during a longer period. For instance, with a short-time gamma-irradiation (5 minutes) the instantaneous phosphorescence decreased to 14 percent of its original value within eight minutes, with a long-time irradiation (12,000 minutes) applied at the same rate the same percentage decrease was reached only after eight thousand minutes.

These increased lifetimes with extended periods of irradiation indicate that electron traps of different depths are responsible for this phosphorescence. The deeper traps which give a longer lifetime are filled to an equilibrium value only after the time of irradiation is extended for a time longer than the lifetime associated with these traps. Thus, with increased times of irradiation the deeper traps are filled to a higher degree and the lifetime is thus extended, whereas the shallower traps are already filled to an equilibrium level after a shorter time of irradiation.

It is noteworthy that the amount of stimulated light, however, is essentially independent of the time of irradiation by high energy radiation but depends mostly on the total dosage. This indicates that in those traps responsible for the observed phenomenon of long-time decay only a relatively small part of the energy which can be released as light is stored.

The third lifetime, namely, that of storage could not yet be determined very accurately. The amount of energy stored, which is indicated by the intensity of the stimulated light, does not decrease by more than two or three percent during one day under normal laboratory conditions according to our observations. This means a lifetime of the order of weeks or longer.

The idea presents itself that this long-time storage is in some way connected to the formation of color centers in alkali-halide crystals since it is known that these persist for long periods of time. It was observed that the crystals of high storage qualities show a considerable coloration after a gamma-dose of 1000 roentgens and more. The color was in this case brownish; the spectrum is similar to the absorption spectrum of the pure colored centered sodium chloride crystals.[2] This coloration increased with increasing doses and was thus as indicated above parallel to the stimulability of the crystal, and the color disappeared under constant irradiation with visible light as did the stimulability. Other crystals of the same kind but made differently, which also show a considerable fluorescence but a much smaller stimulability, exhibited a much smaller coloration. Repeated gamma-irradiation of the crystal up to 10,000 roentgens makes the crystal quite brown, but this color can be made to disappear by continued irradiation by the stimulating light, at which time the light stimulated intensity is very small.

As a result of these experiments the following tentative explanation for the process involved may be proposed The electrons removed from the negative ions in the crystal by high energy radiation are partly trapped (about 5 percent according to our experiments)[3] in lattice vacancies just as in normal alkali-halide crystals. From these it can be concluded that an energy of 2 to 3 electron volts is needed to remove the electron from color centers to the conductivity band. This large energy of more than 2 volts provides the reason for the stability of these excited states (color centers) for long periods of time. The stimulating light brings these electrons into the conductivity band from where either they fall down to a lower state which is connected with the activator and which transition produces the light emission, or they are trapped in electron traps close to the conductivity band (perhaps identical with the F' centers).[2] These traps are responsible for the instantaneous phosphorescence and for the phosphorescence of the stimulated light. Since the lifetimes for both decay processes are essentially the same, these lifetimes are both attributed to electron traps near the conductivity band. The stimulated light and the high energy radiation both bring the electrons in the conductivity band and their further behavior is independent of whether the electrons get into this band by stimulation or by high energy excitation.

Thus the color centers are responsible for the observed storage and explain its long persistence. The stimulating light produces an effect equivalent to the bleaching effect known to occur in alkali-halide crystals. The difference of the crystals investigated from the unactivated ones lies in the ability of their electrons in the conductivity band to go over, with a relatively high probability, to a special ground state (activator) with emission of light instead of recombining without light emission with the normal ground state.

The process which gives rise to the ultraviolet light emission in the silver activator has still to be explored in more detail. It may be that the activators create states near the valence band, which trap, as in the case of the zinc sulfides, the holes produced in the valence band.

* This work was sponsored by the Signal Corps Engineering Laboratory, Fort Monmouth, New Jersey.

[1] M. Furst and H. Kallmann, Phys. Rev. **82**, 964 (1951).

[2] N. F. Mott and R. W. Gurney, *Electronic Processes in Ionic Crystals* (Oxford University Press, England, 1940), Chapter IV.

[3] To be published soon.

A Comparison of the Experimental and Computed Fast Neutron Efficiencies of a Scintillation Counter*

GEORGE E. OWEN, JOHN NEILER, AND WILLIAM RAY
University of Pittsburgh, Pittsburgh, Pennsylvania

(Received May 21, 1951)

SCINTILLATION counters employing hydrogenous crystals or fluorescent hydrogenous liquids have been used by many investigators to detect fast neutrons.[1] Because the proton cross sections for neutrons are well known,[2] the efficiency of such neutron counters can be calculated.

The purpose of the following experiment was to compare the calculated counting rate for a one-inch cube of anthracene with the experimental counting rate. To perform this comparison a standarized PoBe neutron source was employed.

The neutrons from PoBe have a continuous energy distribution extending from zero to 11 Mev. The theoretical analysis in the following discussion utilizes the neutron energy distribution of Whitmore and Baker.[3]

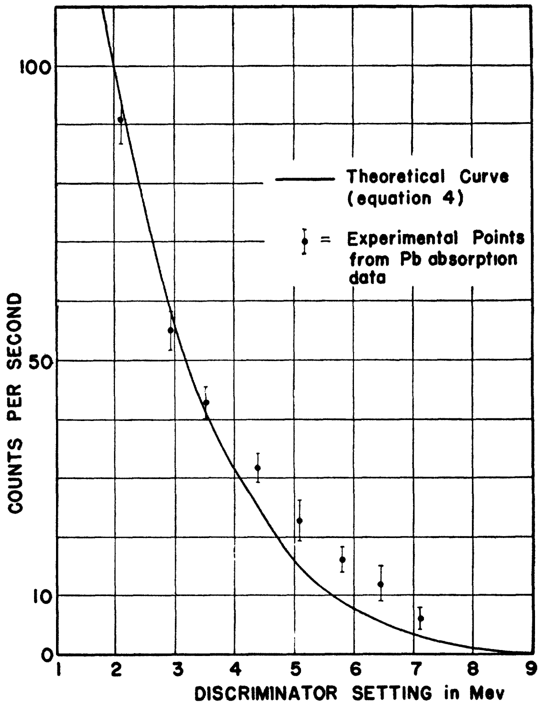

FIG. 1. Integral bias curves for the neutrons from PoBe. The statistical variations shown for the experimental points illustrate the limits of accuracy on the gamma-ray subtraction.

If the total neutron intensity of the standard PoBe source is N', the number of neutrons incident upon the counter crystal is $N'\Omega=N$, where Ω is the fractional solid angle which the crystal subtends at the source. Then

$$N=C_1\int_0^\infty N(E)dE, \tag{1}$$

where $N(E)$ is the energy distribution of the PoBe neutrons.[3]

If the discriminator setting of the amplifier is E_D,[4] the number of neutron interactions in the counter crystal corresponding to neutron energies greater than E_D is

$$n=C_1\int_{E_D}^\infty Kl\sigma_H(E)N(E)dE; \tag{2}$$

l is the length of the crystal in cm, K is the number of hydrogen atoms per cm per barn,[5] and σ_H is the hydrogen cross section in barns.

Equation (2) does not give the number of neutrons which are counted. Consider a neutron interacting with a proton at an energy, $E>E_D$. The recorded pulse is produced by recoil proton, and all energies from 0 to E are possible for this proton. Therefore, the probability that an interaction will record is given by the ratio of the solid angle in which the protons have energies, $E_P>E_D$, to the total solid angle.

The solid angle which will not record has angular limits given by $E_P=E_D=E\sin^2\phi_0$; and the fraction of the interactions which will be counted is

$$(4\pi)^{-1}\left\{4\pi-2\pi\int_0^{2\phi_0}\sin\phi_{CM}d\phi_{CM}\right\}=(E-E_D)/E. \tag{3}$$

This assumes that the scattering is isotropic in the center-of-mass system; therefore, one would expect small deviations from Eq. (3) at energies greater than approximately 10 Mev.

The number of counted interactions, N_R, is therefore given by a combination of Eqs. (1), (2), and (3):

$$N_R=N\int_{E_D}^\infty Kl\sigma_H(E)[(E-E_D)/E]N(E)dE\Big/\int_0^\infty N(E)dE. \tag{4}$$

Equation (4) was computed by graphical methods as a function of E_D. The results of this computation for the specific solid angle, Ω, used in the experiment are shown in Fig. 1.