Fluorescence, Phosphorescence, and Photostimulation of NaCl(AgCl) with High-Energy Irradiation*

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Quantitative results are presented on the light-emitting and storage properties of silver chloride activated sodium chloride crystals excited with high-energy radiation. The characteristic of the strong light emission, mostly in the ultraviolet range, upon stimulation by longer wavelength light after pre-excitation with high-energy radiation, is extensively investigated, as well as the fluorescence and phosphorescence. A practically linear stimulated light response as a function of the roentgen dose over a range from about 10^{-2} to 10^{4} roentgens is found. Such a measurement of stimulated light can be performed a considerable time after the exposure to the high-energy radiation and still provide a determination of the dose. Both the ordinary phosphorescence and the stimulated light phosphorescence under suitable conditions may decay very slowly and show many other similarities. The measurements show that the amount of energy stored by the crystal which can later be released as emitted light is greater than 3 percent of the energy totally absorbed. After a sufficiently long stimulation by visible light the rate of de-excitation becomes very small; further stimulation with light of shorter wavelengths produces a rise in the amount of stimulated light and an increase in the de-excitation rate. The amount of stored energy which can be stimulated as light decays slowly with storage time. This effect is closely correlated with the coloration well known with pure NaCl crystals; bleaching a crystal results in a reduced stimulation. Most of the experiments were carried out with a radium gamma-ray source, but measurements with x-rays and fast electrons show very similar results. Experiments with fast electrons were performed with doses up to 500 000 rep; saturation then occurs, but after de-excitation (bleaching) the crystal could be re-excited and be used again with no appreciable change in properties. The counting properties of these crystals are rather poor because of small peak heights. A rather complete explanation of these properties can be given by a model assuming a continuous distribution of traps over a small range. The energy released from these traps goes to the activator, from which the energy is emitted as light.

I. INTRODUCTION

THE more qualitative aspects of the fluorescence, phosphorescence, energy storage, and release of the stored energy as light in AgCl-activated NaCl crystals when excited by high-energy radiation have been presented in two letters.[1,2] Here quantitative results of more complete experiments and some further findings with such crystals will be given. Alkali-halide crystals with and without special activation have been previously investigated and found to exhibit long-time phosphorescence, and energy storage and release by light,[3–8] though very little quantitative data on stimulation properties has been published. The emitted light intensities of activated crystals were found to be very much greater than for pure NaCl; in fact NaCl(AgCl) is one of the most efficient stimulable phosphors, which has not been fully recognized and investigated previously. It is the large amount of light (see Sec. IV), which can be released by stimulation which makes these systems particularly interesting.

The present investigation has been carried out with NaCl crystals activated with 1 percent of AgCl[9] since, of the crystals so far studied, these were found to have the best energy storage properties. The fluorescence (integrated intensity) of these crystals under gamma-ray excitation was found to be about the same as that of an anthracene crystal of the same mass. Upon removal of the excitation source the crystal showed a rather long-time phosphorescence, especially if the excitation time was sufficiently extended. After the decay of the phosphorescence to a low level and subsequent irradiation of the crystal with light in a range from the near ultraviolet to the near infrared (designated below as stimulating light), the crystal exhibited a strong light emission mostly in the ultraviolet region (stimulated light). No such stimulation occurs if the crystal is not previously excited with high-energy radiation. This shows that a portion of the energy absorbed from the gamma radiation can be stored in the crystal and be released by light. This storage of energy is found to be associated with the presence of color centers which have been so extensively studied by Pohl[3] and many

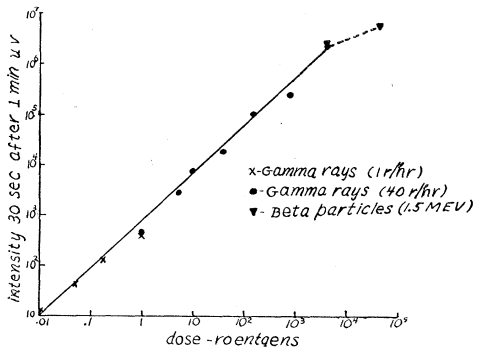

FIG. 1. Light-stimulated intensities for various doses.

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[1] M. Furst and H. Kallmann, Phys. Rev. **82**, 964 (1951).
[2] H. Kallmann and M. Furst, Phys. Rev. **83**, 674 (1951).
[3] R. W. Pohl, Proc. Phys. Soc. (London) **49**, extra part, 3 (1937).
[4] K. Przibram, Z. Physik **20**, 196 (1924); **107**, 709 (1937).
[5] D. Glasser and I. E. Beasely, Phys. Rev. **47**, 570, 789 (1935).
[6] M. Kato, Sci. Papers Inst. Phys. Chem. Research Tokyo **41**, 113 (1943); **42**, 35, 95 (1944).
[7] Weyl, Schulman, Ginther, and Evans, J. Electrochem. Soc. **95**, 70 (1949); Schulman, Ginther, and Klick, J. Appl. Phys. **22**, 1479 (1951).
[8] F. Seitz, Revs. Modern Phys. **18**, 384 (1946).
[9] Most of them provided by the courtesy of Harshaw Chemical Company.

other investigators. We have not attempted to work out the full relationship between these two effects since more careful study of the influence of the activators on the behavior of the color centers must first be made. The stimulated light also exhibits a fairly slow decrease after removal of the stimulating source, similar to the phosphorescence after gamma excitation. The initial intensity of the stimulated light after sufficient excitation was many times greater than the fluorescent intensity with the identical gamma-ray source.

Of practical interest is the essentially linear light stimulation with high-energy radiation doses (Fig. 1) within the range of 10^{-2} to about 10^4 roentgens (see Sec. IV). Also the amount of high-energy radiation absorbed by a crystal can be measured at a considerably later time.

II. FLUORESCENCE AND SPECTRAL DISTRIBUTION

The measuring arrangement was essentially the same as that used in previous measurements with solutions and crystals.[10]

Since the total fluorescent light output of these activated NaCl crystals is of the order of 10 percent of the total absorbed energy,[11] their fluorescence cannot stem from the direct excitation of the activators alone because their concentration is not more than 1 percent; instead the fluorescence must at least partly originate in a transport of energy from the bulk material to the activators (see Theoretical Section).

The fluorescent spectral distribution under gamma-ray excitation is depicted in Fig. 2. Two bands occur, one ranging from about 2300A to 2600A and a somewhat weaker band ranging from about 3200A to 4500A, essentially the same as those found with light excitation.[6] With the use of filters, it was found that more than 75 percent of the fluorescent emission lies in the ultraviolet below 4000A. The stimulated light emission also occurs in practically the same spectral regions, but the far ultraviolet band is relatively weaker. The gamma-ray-excited emission spectra of crystals from different portions of a large crystal from the melt show characteristic differences; those portions from the bottom (cone portion) having a greater relative intensity in the shorter ultraviolet band. Figure 2 also shows the stimulating light spectrum that was most often used; this source is designated as a 3600A source. The spectrum of the stimulated light is essentially independent of that of the stimulating light; thus the stimulated band below 4000A also appears under stimulation with longer wavelengths. The stimulation band is found to be broader, especially toward the ultraviolet, than the absorption band of the color centers of the pure NaCl crystal.[12]

[10] H. Kallmann and M. Furst, Phys. Rev. **79**, 857 (1950); Broser, Kallmann, and Martius, Z. Naturforsch. **4a**, 204 (1949).
[11] Furst, Kallmann, and Kramer, Phys. Rev. **89**, 416 (1953).
[12] N. F. Mott and R. W. Gurney, *Electronic Processes in Ionic Crystals* (Oxford University Press, London, 1948), p. 111.

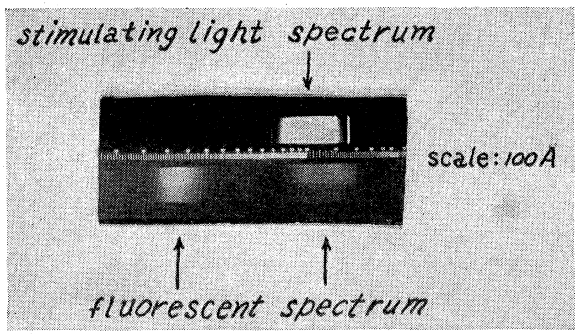

FIG. 2. Fluorescent spectrum of NaCl activated with 1 percent AgCl under gamma-ray excitation.

III. PHOSPHORESCENCE AFTER HIGH-ENERGY EXCITATION

Starting with a de-excited crystal (see below), it was observed that the fluorescent light emission under gamma excitation did not rise almost instantaneously as with organic crystals; instead a continuous rise was found which became more and more gradual (see Fig. 3). The rise time is independent of the excitation intensity in the range 0.05 r/hr to 3.7 r/hr. Upon the removal of the exciting source an almost immediate drop to about one-third of the fluorescence occurs; then there is a more gradual decrease in phosphorescent intensity. Figures 4 and 5 show the decay of phosphorescence after various doses of gamma radiation. The values there are referred to the reading 1 minute after removal of the exciting source; this period is considerably longer than the time of the initial very fast decay. The essential results are that the decrease in phosphorescent light is slower than exponential. However, the shape of the curve is practically the same for different doses when applied for equal periods of time; thus it is the duration of excitation which mainly determines the shape of these curves. These results cannot be correlated to those found by Kato.[6] Explanations for these effects will be given in the Theoretical Section.

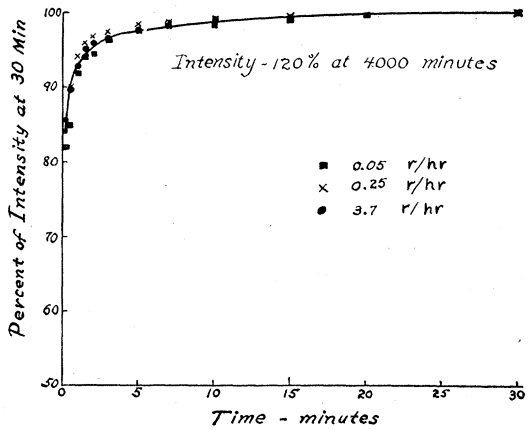

FIG. 3. Rise in fluorescent intensity for gamma-ray excitation—dose rate 1 roentgen per hour.

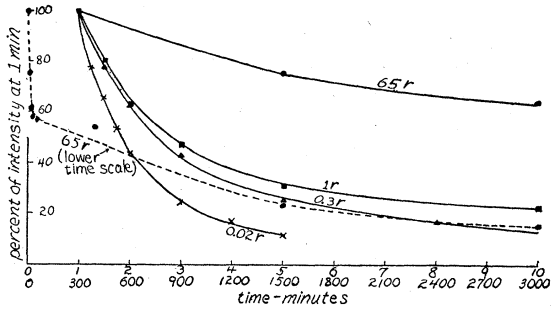

FIG. 4. Phosphorescent decay for various doses after excitation, at rate of 1 roentgen per hour.

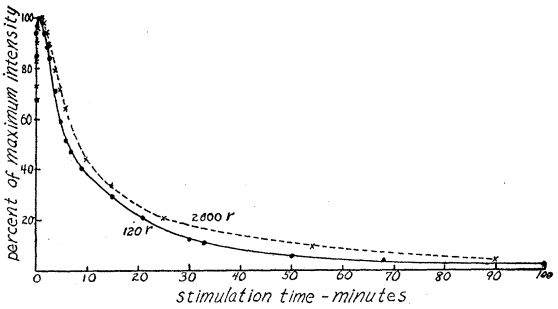

FIG. 6. Stimulated intensity under continuous visible stimulation.

IV. ENERGY STORAGE AND RELEASE BY LIGHT

The amount of light which can be released by stimulation is considerably larger than the total amount of light which is emitted as phosphorescence in several days at room temperature. The band of stimulating light lies in the range from about 3000A to about 5000A, though light outside of this range is also somewhat effective for stimulation (see also Sec. VI). With light of about 2500A, however, direct excitation already occurs.

In determinations of the intensity of the stimulated light it is not necessary to observe the stimulated light while the stimulating light is acting, since the light emitted after removal of the stimulating source also decays slowly; thus two types of experiments were performed. In one series the excited crystal was illuminated with the stimulating light (usually a 3600A source) for a definite time period; then the crystal was placed in the instrument and measurements were begun within 30 seconds. In the other series of experiments the crystal was irradiated with visible light and its light emission was measured simultaneously by screening of the stimulating light by suitable filters. This could not be accomplished for the ultraviolet, which is one of the reasons for always using intermittent stimulation for these wavelengths.

A maximum in the stimulated light emission was not immediately reached upon the application of the stimulating light, a behavior similar to that with the fluorescent light. Figure 6 shows as examples the relative stimulated light intensities under continuous visible irradiation after 120 and 2600 roentgen doses were applied; these curves are remarkably similar (see Theoretical Section). The time when the maximum is reached depends on the intensity of the stimulating source, so that, for example, with the stimulating ultraviolet intensity reduced by a factor of 10, the time for maximum stimulated intensity was increased from 3 minutes to 20 minutes. A similar shift was found with visible stimulation.

The curves of Fig. 6 further show that the stimulated light intensity does not decrease exponentially, but at a much smaller rate.[13] A similar effect is found with ZnS phosphors. This may also be seen from Table I, which presents the stimulated intensities for intermittent stimulation with varying dark periods in between.

From these continuous-stimulation curves, a lower limit for the entire stimulated light (light intensity multiplied by time) can be calculated by integrating the curve. This can then be compared to the total amount of fluorescent light emitted under simultaneous gamma excitation, which is essentially given by the maximum fluorescent intensity multiplied by the time

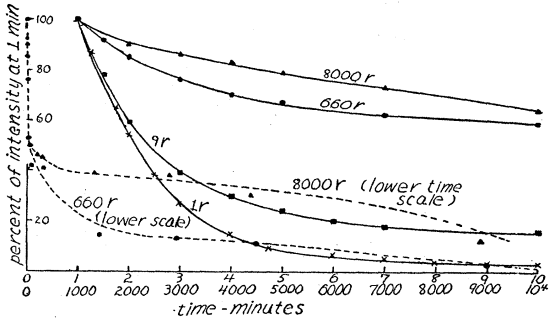

FIG. 5. Phosphorescent decay for various doses after excitation, at rate of 40 roentgens per hour.

TABLE I. Decrease of stimulated intensity with ultraviolet stimulation dose 8000 roentgens.

Irradiation	Time	Intensity 30 sec after irradiation
Gamma, at rate of 40 r/hr	200 hr[a]	5000[b]
No irradiation	503 hr	170[b]
Ultraviolet	1 min	185 000
No irradiation	25 hr	130[c]
Ultraviolet	1 min	47 500
No irradiation	4.5 hr	550[c]
Ultraviolet	1 min	32 000
No irradiation	65 hr	29[c]
Ultraviolet	1 min	26 500
No irradiation	36 min	330[c]
Ultraviolet	1 min	21 000
No irradiation	24 hr	...
Ultraviolet	2 min	7000
No irradiation	17 hr	24[c]
Ultraviolet	1 min	5700

[a] Excitation time.
[b] Remaining phosphorescence after high-energy excitation.
[c] Remaining phosphorescence from previous stimulation.

[13] It may be noted that these curves do not represent a spontaneous decay but a decrease while the stimulating light acts.

for long exposures. These calculations show that the total amount of stimulated light (released by visible light) is about 30 percent of the total fluorescent light. However, since there is considerable additional stimulation possible by means of light in the near ultraviolet, the total amount of stored energy which can be emitted as light may still lie above this figure. Since about 10 percent of the absorbed gamma-ray energy is emitted as light as shown by comparing with anthracene,[11] the portion of absorbed energy which is stored and can later be emitted as light in NaCl(AgCl) turns out to be greater than 3 percent.

TABLE II. Stimulated light intensities.

Irradiation	Time	Intensity[a]	Percent of first ultraviolet reading
Gamma, at rate of 40 r/hr	20 hr	...	...
Ultraviolet	1 min	210 000	100
Ultraviolet	2 min	84 000	41
No irradiation	30 hr		
Ultraviolet	1 min	36 600	18
No irradiation	72 hr	...	...
Ultraviolet	1 min	20 000	10
Gamma, at rate of 1 r/hr	1 min	440[b]	...
Ultraviolet	1 min	12	100
Ultraviolet	1 min	7	60
Gamma, at rate of 1 r/hr	5 min	530[b]	...
Ultraviolet	1 min	17	37
Gamma, at rate of 1 r/hr	18 min	630[b]	...
Ultraviolet	1 min	130	100
Ultraviolet	1 min	130	100
Ultraviolet	1 min	65	50
Gamma, at rate of 40 r/hr	8 min	...	...
Ultraviolet	1 min	2600	100
Ultraviolet	1 min	1200	46
Gamma, at rate of 40 r/hr	15 min	...	...
Ultraviolet	1 min	5500	100
Ultraviolet	1 min	1900	35
Ultraviolet	1 min	1050	19
Gamma, at rate of 40 r/hr	60 min	...	...
Ultraviolet	1 min	19 500	100
Ultraviolet	1 min	9000	46

[a] Stimulated readings were obtained 30 seconds after stimulation.
[b] Fluorescent value.

After application of the stimulating light for a short interval and its subsequent removal, the light does not completely decay instantaneously; instead a decrease in intensity similar to that of the phosphorescence after high-energy excitation is found. Examples of such curves under various conditions are given in Figs. 7 and 8. All of the curves in the figures are similar in shape to each other (percentage curves) if the same light stimulation is used, though the high-energy excitation conditions are considerably different. However, an increase in the length of the stimulation period (with a weaker light source) produced a considerable slowing down of the phosphorescent decay of the

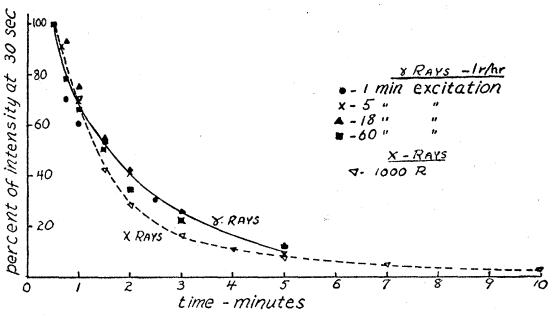

FIG. 7. Decay after 1 minute of ultraviolet stimulation with gamma-ray and x-ray excitations.

stimulated light (see Fig. 8); this is analogous to the slowing down after prolonged gamma-ray excitation (Figs. 4 and 5).

The stimulated light intensities produced during short stimulation periods using the 3600A source after various intervals without stimulation are shown in Tables I and II. The crystal used for the measurements in Table I was cut from the edge of a large crystal grown from the melt and shows poorer storage qualities than other pieces of the same large crystal (see Sec. IX). Table II shows results with more efficient crystals under different excitation and stimulation periods. The periods of darkness were usually long enough for the phosphorescence of the stimulated light to decay to a relatively small portion (about 1 percent) of its value at 30 sec. The ratio of the stimulated intensity after the first stimulation to that after the second one was usually between 2 and 3 (except with long periods of darkness, when the factor was about 4). The ratio between successive stimulations then usually decreases and approaches nearly one after a large number of stimulations are applied. This behavior is similar to that observed with continuous stimulation by visible light, where the slowing down of the decrease in the stimulated light intensities with increasing stimulation time was still more pronounced.

The dependence of the amount of stimulated light on the applied dose and dose rate of excitation are described in Table III; similar doses were applied but at different rates, corresponding to different periods of

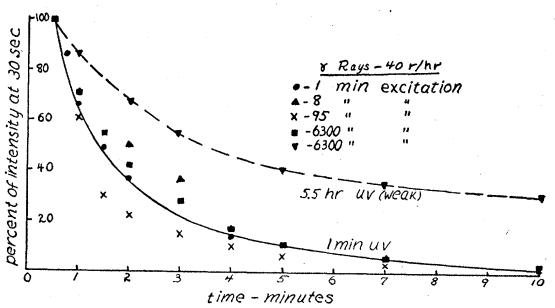

FIG. 8. Decay after 1 minute of ultraviolet stimulation with gamma-ray excitation.

TABLE III. Intensities for different excitation rates after ultraviolet stimulation.

Rate r/hr	γ irradiation Time	Dose, roentgens[a]	Intensity 30 sec after 1 min of uv stimulation
0.01	2 hr	0.020	24
1	1 min	0.017	22
1	60 min	1	380
60	1 min	1	420

[a] Different crystals were used with the two doses.

excitation. The results show that the intensity of the stimulated light depends essentially only on the total dose of radiation rather than on the length of excitation time for a given dose. For very much larger differences in exposure time than described in the table, the effect of the decay of the stored energy becomes noticeable (see Sec. V).

In Fig. 1 the relationship between stimulated light intensities and dose are described. The smallest dose of gamma radiation measurable with accuracy by stimulated light was about 10 milliroentgens; somewhat smaller doses could be detected, but the readings were close to the small residual stimulated intensities present even with unexcited crystals. For softer high-energy radiations, especially x-rays, the minimum detectable dose would be smaller because of the greater amount of energy absorbed by the same crystal volume for the same roentgen dose. The curve of Fig. 1 shows that from about 0.01 to 4200 roentgens, a factor greater than 10^5 in dosage range, the intensity of the stimulated light was essentially proportional to the dosage. There are fluctuations in the curve, possibly due in some cases to the differences in the periods of excitation (see also next section); however, no systematic deviations were found. High-energy electrons (~1.5 Mev)[14] were also used as an excitation source, and the point corresponding to 4000 r with this source fits rather well in the curve obtained for gamma rays. With 40 000 r, a deviation from linearity becomes noticeable, and at 5×10^5 roentgens definite saturation was observed. (Part of this saturation effect may be due to the increased opacity of the crystal due to the strong coloration.) An extensive check of the relation between the stimulated light intensity and the previous roentgen dose shows that these crystals can be used for quantitative dose measurements, especially if the relatively small decay of the stimulated light intensity with storage time is taken into account. A crystal which has been highly excited and then de-excited can be re-excited with no noticeable change in its properties even after having received a total dosage of millions of roentgens.

TABLE IV. Stimulated intensities with various doses and periods of darkness.

Dose, roentgens	Rate, r/hr	Excitation time, min	Dark period	Intensity 30 sec after 1 min uv
		Crystal No. 1		
0.13	0.53	15	2 min	49
0.14	0.53	16	80 min	51
0.14	0.53	16	17.5 hr	37
0.13	0.53	15	162 hr	17
0.53	0.53	60	5 min	190
0.55	0.53	62	75 min	160
0.53	0.53	60	16.5 hr	120
11	44	15	2 min	4200
11	44	15	80 min	3750
12	44	16.5	18.5 hr	2700
11	44	15	64 hr	1800
11	44	15	960 hr	800
44	44	60	5 min	15 000
44	44	60	80 min	11 000
44	44	60	16 hr	10 500
		Crystal No. 3		
0.13	0.53	15	2 min	46
0.15	0.53	17	90 min	28
0.14	0.53	16	17.5 hr	10
0.13	0.53	15	162 hr	3
0.53	0.55	62	5 min	165
0.53	0.53	60	5 min	180
0.55	0.53	62	65 min	130
0.53	0.53	60	15.5 hr	34
0.54	0.53	60	64 hr	13
11	44	15	2 min	1400
11	44	15	17.5 hr	600
56	44	77	2 min	9600
44	44	60	17 hr	1600

V. DECAY OF STIMULATED LIGHT INTENSITY WITH STORAGE TIME

The stimulated light intensities are only comparatively slightly diminished if the energy is stored in the crystal for a considerable length of time. Results using two different crystals are given in Table IV. Crystal No. 1, typical of crystals from the center of a large crystal grown from the melt, exhibits a decrease of stimulated light intensity with a storage time of 160 hours by a factor of at most 3. Crystal No. 3, which was obtained from the edge of the same large crystal grown from the melt as was crystal No. 1, exhibits a somewhat faster decrease in stimulated intensity with storage time for these doses. Experiments with other crystals bear out the somewhat faster decay with crystals near the edge (see below).

The slow decrease of energy storage and its localization are also shown by an experiment using fast electrons (beta particles) for exciting a small area of the crystal. Subsequent stimulation produced a sharp luminescent spot which gave no visual evidence of any broadening when the stimulation was repeated after the lapse of two weeks.

[14] The fast electron generator of the Electronized Chemical Company, Brooklyn, New York, was used (see Sec. VII).

VI. DE-EXCITATION

De-excitation of the crystal is shown by the decrease in stimulability.[15] Figure 6 and Tables I and V represent also such de-excitation. With increasing de-excitation, further de-excitation becomes increasingly difficult.

This decrease in the rate of de-excitation is not determined by the absolute amount of residual stored energy but rather by the percentage of the initial stored energy that remains (Fig. 6); it occurs with near-ultraviolet light (~3600A) stimulation as well as with visible stimulation, but the slowing down occurs with a different degree of stimulability. With shorter wavelengths, of about 2500A, the de-excitation process is somewhat faster, but with such wavelengths there is also some direct excitation, and an equilibrium is reached under prolonged irradiation. As a result of these measurements, the method of de-exciting strongly excited crystals usually consisted of exposing the crystal to ultraviolet light in the region of 2500A and then to light with longer wavelengths for final de-excitation.

Even after such a procedure the crystals still retain a small amount of residual excitation equivalent to about an exposure of the order of 1 milliroentgen (a reading of about 5 in Tables I and IV) which could not be removed by this method. This small excitation was present even with crystals that were not previously exposed to high-energy radiation except cosmic rays. Preliminary experiments with cosmic rays have been inconclusive in deciding whether the residual effect is due to them or to a small direct excitation with the stimulating source. This will be investigated further.

Experiments carried out alternately with different wavelengths of stimulating light have shown that they influence each other in producing stimulation. Typical results with intermittent stimulation are shown in Table V. The light sources used for quenching did not all have the same total energy; thus the source of visible light had an energy greater by a factor of 50 than the 3600A source, while the source in the region of 2800A was the weakest. This table shows that in spite of these different strengths the shorter wavelengths de-excite more efficiently. Initially, with the above-mentioned sources, the stimulated light intensity decreases for each consecutive shot of stimulating light from the same source. Periods of darkness of the order of a day do not appreciably accelerate the decrease in stimulated intensity. If a partially de-excited crystal is stimulated for a longer period of time (e.g., 10 min or longer) and then followed by a dark period of the same duration, a subsequent short stimulation (30 sec) of the same wavelength (for visible and ultraviolet) produces a greater stimulated intensity than the preceding long stimulation. This is due to a heating of the crystal during the prolonged stimulation and a consequent faster decay of the emitted light (see below).

Once the crystal is considerably de-excited, an

[15] By stimulability we mean the capability of the crystal to emit light induced by irradiation of the crystal with a visible or near-ultraviolet light source. It depends mainly on the previous excitation of the crystal, for instance by gamma radiation. It is essentially zero if the crystal is completely de-excited and not again excited by high-energy radiation.

TABLE V. Stimulation and de-excitation with different light sources.

Stimulation			Emission intensity after 30 sec		
Visible	uv, region of 3600A	uv, region of 2800A	Visible	uv, region of 3600A	uv, region of 2800A
30 sec	...	...	2 030 000	...	...
...	30 sec	...	...	3 330 000	...
...	...	30 sec	...	...	1 530 000
30 sec	...	...	850 000	...	...
30 sec	...	...	535 000	...	...
...	30 sec	...	...	1 000 000	...
...	30 sec	...	...	900 000	...
...	30 sec	...	...	775 000	...
...	...	30 sec	...	...	450 000
...	...	30 sec	...	...	370 000
...	...	30 sec[a]	...	...	280 000
...	30 sec	...	...	370 000	...
30 sec	...	...	170 000	...	...
30 sec	...	...	124 000	...	...
2 min	...	...	40 000	...	...
2 min	...	...	12 000	...	...
30 sec	...	...	13 500	...	...
...	2 min	...	...	125 000	...
...	2 min	...	...	100 000	...
30 sec	...	...	36 500	...	...
...	...	30 sec	...	...	32 000
...	...	2 min	...	...	33 000
...	...	2 min	...	...	29 000
...	...	30 sec	...	...	25 000
30 sec	...	...	14 000	...	...
...	30 sec	...	...	44 000	...
10 min	...	...	750	...	...
30 sec	...	...	1250	...	...
...	10 min	...	...	8700	...
...	30 sec	...	...	15 000	...
30 sec	...	...	4000	...	...
...	...	14 min	...	...	2900
...	...	30 sec	...	...	3100
30 sec	...	...	2200	...	...
...	30 sec	...	...	6800	...
...	30 sec	...	...	7700	...
...	30 sec[a]	...	...	7100	...
30 sec	...	...	2600	...	...
45 min	...	...	170	...	...
30 sec	...	...	180	...	...
30 sec	...	...	165	...	...
...	30 sec	...	...	2400	...
30 sec	...	...	980	...	...
30 sec	...	...	680	...	...
30 sec	70 min[b]	...	180	...	...
30 sec	...	...	130	...	...
...	30 sec	...	...	390	...
...	30 sec	...	...	470	...
...	30 sec	...	...	480	...
30 sec	...	...	250	...	...
30 sec	...	...	145	...	...
8.5 min	...	...	9	...	...
30 sec	...	...	19	...	...
30 sec	...	...	22	...	...
...	...	30 sec	...	...	72
...	...	30 sec	...	...	82
30 sec	...	...	39	...	...
30 sec[c]	...	...	33	...	...
...	30 sec	...	...	260	...
...	30 sec	...	...	380	...
...	30 sec	...	...	430	...

[a] The crystal had been in the dark for 16 hours.
[b] Visible light was applied directly after 70 minutes of ultraviolet.
[c] The crystal had been in the dark for 64 hours; the decay after stimulation was slower than with the previous shot.

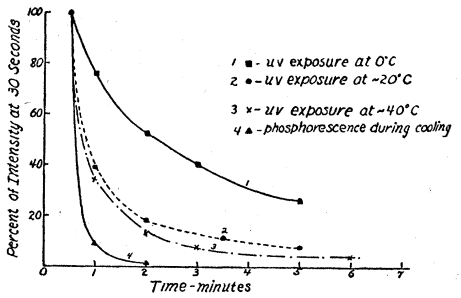

FIG. 9. Effect of temperature on stimulated emission.

interaction in the de-excitation by different wavelengths occurs which can be seen in Table V. After ultraviolet stimulation (3600A) the succeeding stimulation by visible light produces a considerably higher value than the previous similar visible stimulation. There are also indications, not shown in this table, that in a similar manner stimulation with shorter ultraviolet wavelengths induces an increase in stimulation with longer ultraviolet wavelengths. The effect is due to the presence of electron traps of different depths (see Theoretical Section).

In order to provide more information about the phosphorescent properties of these crystals a small number of experiments at different temperatures were performed. Figure 9 presents curves of phosphorescent decay after stimulation is removed for different temperatures (curves 1–3), while curve 4 gives the light emission as function of time for a warm crystal during its period of cooling off towards room temperature. The curves show how the decay of light emission becomes more rapid with increasing temperature. Figure 10 depicts something like a glow curve: an excited crystal cooled down to −78°C was stimulated by ultraviolet at this temperature and then slowly warmed up. The first part of the curve shows the phosphorescent decay at the low temperature; after about two minutes the warming up prevents the further decrease of the phosphorescent emission and the light emission increases to a maximum followed by another decrease as a consequence of a depletion of the phosphorescent traps (see Theoretical Section). However, no effort to obtain accurate glow curves was made because of appreciable retrapping and the lack of saturation of the traps; both of these conditions are necessary for the usual interpretation given to glow curves.

VII. OTHER EXCITATION SOURCES

The experiments described above are concerned almost entirely with excitation by gamma rays. Fast electrons (beta particles) also produce a similar excitation of the crystal, and the results described above are also valid with this type of excitation (see also Fig. 1). Though some experiments with fast electrons were performed using a 10-millicurie strontium 90 source, most of the quantitative results with fast electrons were obtained by using pulses from a discharge tube.[16] The equipment provided a pulse of 1.5-Mev electrons for a period of about 10^{-5} second. The radiation intensity transferred in one pulse to the crystal was of the order of 7×10^5 erg/cm^2 [10 000 rep (roentgen-equivalent-physical)] at about 120 cm from the source as determined by calorimetric measurements.

The amount of light stimulated after the electron excitation was essentially the same as that with gamma-ray excitation for the same amount of absorbed energy if the decay of the stored energy in the interval between excitation and stimulation was taken into consideration. The decay of the light emitted after stimulation followed practically the same curves for both types of excitation; also the same type of variability among the different crystals was obtained, although in the case of the electron pulse the density of excitation is 10^{10} times larger than in the case of gamma-ray excitation. With large doses (500 000 rep) either obtained by changing the position of the crystal with respect to the discharge tube or increasing the number of pulses, a definite saturation of the light emitted after stimulation was obtained; thus the light emitted after 30 seconds of ultraviolet stimulation with 500 000 rep and 30 000 rep were found to be about equal. However, de-excitation of the more excited crystals occurred at a slower rate.

With alpha particles[17] the main features of the phenomenon were the same, but the observed fluorescent and stimulated light intensity was smaller for the same amount of absorbed energy for alpha-particle excitation than it is for gamma-ray and fast-electron excitation. Because of the observed deterioration of crystals under alpha-particle excitation,[18] extensive experiments with the alpha-particle source were not carried out.

X-ray excitation also produces results similar to gamma-ray excitation, as can be seen from Fig. 7; also the stimulated light intensity for a 1000-r x-ray dose fits rather well with the results shown in the curve of Fig. 1. The tremendous storage and stimulation properties of these crystals can be demonstrated by the ability

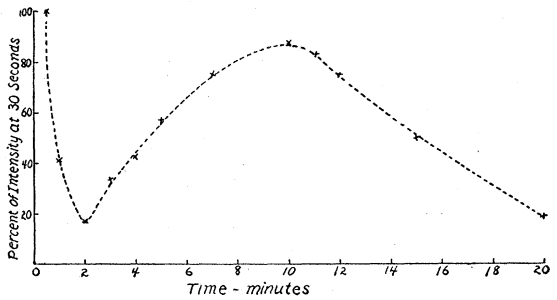

FIG. 10. Light emission of warming crystal, stimulated at −78°C.

[16] Electronized Chemical Company.
[17] C. E. Mandeville and H. O. Albrecht, Phys. Rev. **90**, 25 (1953).
[18] I. Broser and H. Kallmann, Z. Naturforsch. **5a**, 381 (1950).

to depict an object by means of x-rays. The latent image of the object in a crystal can later be photographically recorded by first stimulating the crystal and then placing it directly on a photographic plate. Such a picture was made with as little as 0.1 r from a 60-kv x-ray source. The resolution on these photographs was good. Using a 1-r dose, two clear pictures of the same latent image were made after a lapse of about 30 minutes after the x-ray exposure, during which time the crystal was exposed to sunlight for several minutes. With about 100 r, even after a lapse of 5 days, the latent image was still made visually observable on the crystal by stimulation with an ultraviolet lamp, although by far the greater portion of the emitted light is in the invisible ultraviolet.

VIII. COUNTING PROPERTIES

Some experiments on the counting properties of these crystals have been carried out by L. Bittman and N. Linson at this laboratory. The peak heights of the fluorescent light flashes emitted under gamma-ray excitation were found to be smaller than those of anthracene when measured with a circuit with short time constant, in spite of the high fluorescent efficiency of these crystals. An explanation will be given in the Theoretical Section.

IX. ENERGY STORAGE WITH DIFFERENT CRYSTALS[19]

As previously remarked, the light emission properties of these crystals depend on the degree of activation and on the location in the original melt. Investigation has yielded the following results: Crystals with activations of 0.1 percent and 0.2 percent AgCl were found to be inferior to those with 1 percent activation with respect to fluorescent, phosphorescent, and stimulation efficiencies; but their fluorescence is not weakened as much as the storage efficiency. Those with 5 and 10 percent activation showed a slightly smaller fluorescent and storage efficiency than the 1 percent crystal, but the peak heights of the 5 percent activated crystals have been found to be slightly higher than the 1 percent activated crystals.[20] Investigation of specimens taken from different portions of a quite large crystal grown from the melt made by Harshaw Chemical Company has revealed characteristic differences in the storage and fluorescent properties of the various sections of the large crystal. Table VI indicates the type of variation for the various portions of this large crystal. Specimens from the bottom (cone portion) of the large crystal have smaller stimulabilities than those from the central portion, but the phosphorescence of the stimulated light decreases more slowly with the crystals from the cone portion. The fluorescence of specimens from the cone portion are, however, somewhat higher than those from the top section. Similar results were obtained with portions from other large crystals grown from the melt. These results indicate—and this is the reason for their presentation—that the different traps which are so important for all the light-emitting phenomena of these crystals are not purely a function of the activator (its concentration) but depend considerably on the way the crystal is grown. This indicates that the fluorescent and storage effects are not completely correlated to each other. Crystals with higher concentrations of silver chloride (5 percent and 10 percent) seem to be inferior to the 1 percent crystals in both fluorescent and stimulation properties.

TABLE VI. Response of specimens from different portions of a large NaCl(AgCl 1 percent) crystal.

Crystal No.	Section	Thickness	I_{f160}[a]	$I_{f160}/I_{f1\frac{1}{2}}$[b]	I_{st30}[c]	I_{st30}/I_{st300}[d]	I_{f160}/I_{st}
20	Bottom	0.75 cm	235	1.25	80	3.1	0.34
21	Bottom	0.8 cm	310	1.1	120	4.6	0.39
22	Center	0.85 cm	300	1.3	240	8	0.8
23	Top	1 cm	215	1.07	270	68	1.3
24	Top	0.8 cm	195	1.2	190	21	1

[a] I_{f160} = fluorescent intensity after 60 minutes of irradiation.
[b] $I_{f1\frac{1}{2}}$ = fluorescent intensity after $\frac{1}{2}$ minute of irradiation.
[c] I_{st30} = stimulated intensity 30 seconds after termination of stimulation.
[d] I_{st300} = stimulated intensity 300 seconds after termination of stimulation.

X. COLORATION AND BLEACHING

In experiments with considerable excitation, the NaCl(AgCl) crystal became colored, as is well known for alkali halide crystals;[3,21] the stimulation was found to be fairly closely correlated with the coloration. Those crystals which exhibited high fluorescence but smaller stimulation capabilities were also found to have less coloration for the same dosage than crystals with better stimulation qualities. These experiments show further that the fluorescent efficiency is partially independent of the storage and stimulation efficiencies.

Coloration and stimulability have the following properties in common: (1) Both proceed parallel to each other until the crystal becomes quite opaque and saturation sets in. Considerable stimulability remains, however, when the crystal appears transparent, most likely because the stimulability measurements are more sensitive than those of coloration. (2) As with color centers, where the action of light in any part of the F band bleaches all the rest of the band, de-excitation with one wavelength also decreases the stimulability by other wavelengths; however, it is not completely removed. (3) The growing of the crystal appears to be important for both the coloration of the crystal and the storage and stimulation qualities, as indicated by the influence of position of the specimens in the crystal grown from the melt. (4) The decay of the stored energy as determined by the amount of stimulation is similar to the decay of the degree of coloration as reported by Alger.[21] The number of trapped electrons as determined

[19] Some of the crystals were supplied by the courtesy of Squier Laboratory, Signal Corps.
[20] Bittman, Furst, and Kallmann, Phys. Rev. **87**, 83 (1952).
[21] R. S. Alger, J. Appl. Phys. **21**, 30 (1950).

from the saturation of light emission ($\sim 10^{17}$ per gram) is about the same as the number of vacancies associated with the color centers.[3] There are, however, features occurring with color centers and stimulable energy storage which are not correlated with each other (see Theoretical Section).

XI. THEORETICAL CONSIDERATIONS (DYNAMICS OF PHOSPHORESCENCE, STIMULATION, AND DE-EXCITATION)

The experiments described above on the slow rise of the fluorescent light emission, on the phosphorescent decays after excitation and stimulation, and on the stimulability and the de-excitation of the crystal can all be explained by using the relatively simple model depicted in Fig. 11. The valence band and the lowest excited band of the bulk material are shown, and it is assumed that practically the entire excitation of the crystal by high-energy radiation takes place from the valence band to various higher excitation bands of the bulk material. The electrons excited to these bands will return to the valence band mostly by means of passing through the band of lowest excitation energy shown in the figure. In the energy gap between these two bands there are a number of trap levels distributed over a range of energy; these traps are assumed to be much less numerous than the number of atoms of the bulk material. These are divided into two types, designated by p and s for reasons discussed below. In addition there are levels of the activator atoms. It is assumed that most of the deep traps are not directly associated with the activators, since strong trapping also occurs without activators being present. The amount of direct excitation from the valence band to activator or trap levels is assumed to be negligible. The electrons or excitation energy which reach the excited band of the bulk material will go either to the traps or to the activators from which light may be emitted. From the curve of fluorescent light as a function of time, it can be concluded that a considerable amount of excitation energy goes to the traps.

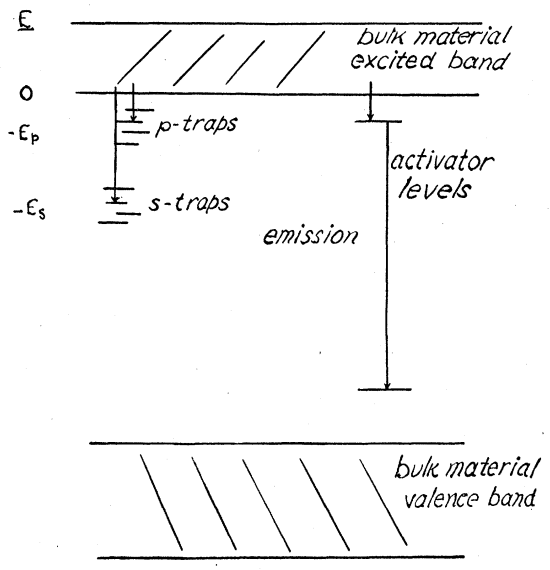

FIG. 11. Energy level scheme for emission and storage with activated NaCl crystals.

The less deep traps, p, are assumed to be responsible for the phosphorescence, whereas the deeper traps, s, are associated with the storage phenomenon. The number of s traps is calculated from the saturation of stimulation at about 5×10^5 roentgens to be about 10^{17} per gram of the bulk material. Two types of traps are assumed because it was found that the energy storage exhibits only a very small short time component, smaller than expected from the appreciable intensity of the short time component found in the phosphorescence even after prolonged periods of high-energy excitation. The stimulable stored energy, on the other hand, decays only very slowly with time and is still considerable after 1000 hours. Nevertheless, it is not completely certain that it is necessary to discriminate between two different types of traps. The experiments are not conclusive enough to exclude the possibility of postulating a continuous distribution of similar traps from the highest p traps to the deepest s traps. Certainly some of the observed long time phosphorescence is due to the spontaneous release from s traps.

It was indicated in Fig. 3 that the fluorescent light emission reaches a rather steady value after about 20 minutes and then rises only very slowly within days. This rise of the fluorescent radiation is due to the fact that not all of the electrons in the excited level of the bulk material reach the emission centers; instead a considerable portion are trapped in the p and s levels. Only if the number of electrons released from these traps by collisions with the lattice becomes equal to the number of electrons falling from the excited state into these traps, will the net consumption of electrons by the traps stop and the light emission increase. For an energy level i lying below the excitation band by an amount E_i, the time τ_i when equilibrium (between filling and release) will be reached for this level (τ_i also gives the decay time of the trap) is given by the following well-known equation:

$$1/\tau_i=\gamma_i \exp(-E_i/kT), \qquad (1)$$

where $\gamma_i=\epsilon_i\nu_i$, ν_i is the frequency factor of the average number of vibrations made per second by the electrons occupying these traps, ϵ_i is a factor which is a measure of the retrapping and structure, and the exponential is the Boltzmann factor.

A rise in the fluorescent curve and its practical leveling off after a certain time could perhaps also be explained by the assumption that the saturation occurs not because of an equilibrium between the number of electrons trapped and those released by heat, but rather because all of the available traps are filled to capacity (saturated). This is very often observed in ZnS phosphor but is not observed for these crystals under the condi-

tions of the investigation, since it was found that the rise time curve of the fluorescent light is practically independent of the intensity of the exciting radiation (Fig. 3), clearly indicating an equilibrium between filling and emptying of the traps but no trap saturation.[22] According to Eq. (1), the rise with time t of the fluorescent intensity I should be given by

$$I=A+B\ln t, \tag{2}$$

where the constants A and B are dependent upon the special model which is chosen for the light emitting processes. Equation (2) holds as long as equilibrium is not reached for all traps. It is valid for a time period extending between a time equal to the time constant of the shallowest trap to that of the longest living traps. After a time τ_m given by Eq. (1), where E_i is replaced by the energy of the deepest trap, no further rise occurs. The experimental curve (Fig. 3) rises over a period of days and can be represented rather well by Eq. (2). This formula is derived under the assumptions that the number of electrons in the excitation levels are proportional to the excitation intensity, and that they contribute either to light emission or filling the traps. The filling of the traps is assumed to be proportional to the number of traps, and the trapping cross section to be the same for all depths of traps. It is further assumed that the retrapping probability does not vary considerably during the period of observation; this is borne out by the similarity of curve shapes in Fig. 3 for different dose rates. Finally, the crude simplification is made that for a given time of excitation t, all traps of an energy E_p, where $E_p/kT=\log(\gamma_p t)$, are filled to equilibrium while all others of greater energy are not filled at all.

If the factor γ_p is assumed to be of the order of 10^{10} per second, a value of $\frac{3}{4}$ ev for the center of the E_p band can be obtained from the phosphorescence experiments. This value is only a crude estimate, since γ is not known accurately enough. Without retrapping and with strong coupling of the traps with the lattice, a factor of 10^{12} per second is usually assumed. For the energy levels involved here, a change of γ from 10^{10} to 10^{12} produces a change of only 15 percent in the calculated energy. A more accurate determination of the trap-level energies can be made if the experiments are performed at a series of different temperatures; this is contemplated for the future.

The strong probability of trapping by these levels can be concluded from various experiments. It was observed that the fluorescent efficiency of these substances is of the order of that of anthracene (10 percent absolute yield). At the same time, in counting experiments the peak heights of light flashes produced by single gamma quanta or alpha particles are found to be smaller than in anthracene. This indicates that a considerable portion of the excited electrons do not immediately lead to light emission but first fill traps. Investigations are under way to show this effect more directly. The very large number of available traps is further shown by the small peak heights from alpha particles, since even the strong excitation density produced by alpha particles is not enough to *saturate* the available traps around an alpha-particle track. In ZnS the situation is mostly reversed.

The experiments show that the phosphorescent effects both after excitation as well as after stimulation are quite similar. It is, therefore, probable that the directly excited electrons and those released by stimulation populate the same excited band since they are subject to being trapped by the same trap levels. If the shapes of the decay curves in Figs. 4 and 5 are considered, it is seen that they deviate considerably from being exponential and that their "decay times" vary with the time of excitation and extend from minutes to hours. This dependence of the decay time on the period of excitation can be explained as follows. According to Eq. (1) the lifetime of the electrons trapped in these levels depend strongly on their energy; thus, if this energy covers a range of about 0.2 ev, the lifetime of these traps would vary from minutes to more than a day. If now the excitation is extended over a certain period T_R, all traps which have a lifetime τ_i smaller than T_R are filled practically to equilibrium; a further extension of the excitation will not increase the population of these traps, but only those traps with lifetimes greater than T_R will have their population increased. Thus, if the same dose is given once within a very short period of excitation and then with a very long time of excitation, the percentage decay such as shown in Figs. 4 and 5 should be more gradual with long excitation time than with short excitation time, since with short excitation time the trap levels with longer lifetimes have the same population as those with shorter lifetimes but contribute less to emission. This is precisely what was found under both excitation and stimulation.

To account for these effects it is sufficient to assume a relative small energy range (of the order of 0.2 ev) with a rather uniform and continuous distribution. This does not necessarily include the deeper s traps (see below). With this assumption and simplifications such as those used with Eq. (2), one obtains an expression for the light intensity I as a function of time t of the type

$$I=K\int_{\delta_R}^{\infty}\frac{e^{-t\delta}}{\delta}d\delta, \tag{3}$$

where $\delta=1/\tau$ of Eq. (1), δ_R is the reciprocal of the irradiation time T_R, and K is a constant.

This equation can be used to compare the amount of light emission obtained after removal of the source for different times of T_R and to compute the relative decay curves. Such comparisons are shown in Table VII, and a fair agreement is found. The largest deviations occur

[22] The possibility still remains that local saturation occurs in the "channel" of the electron; however, pulse measurements appear to show that this is not the case.

TABLE VII. Comparison of phosphorescent intensities with those calculated from Eq. (3).

Irradiation time T_R minutes	Phosphorescent intensity $t=1$ minute[a] Measured	Calculated	$t=5$ minutes Measured	Calculated
1	0.04	0.02	0.002	0.001
13	0.39	0.25	0.09	0.08
1000	0.71	0.72	0.48	0.53
12 000	1	1	0.79	0.81

t,[a] minutes	Phosphorescent decay[b] Measured	Calculated
1	1.0	1.0
5	0.79	0.82
10	0.64	0.74
200	0.46	0.40
1000	0.39	0.23
9000	0.12	0.03

[a] t is the time after removal of the exciting source.
[b] $T_R=12\,000$ minutes.

when the decay time approaches the irradiation time; the values calculated from the equations are then too small. This is expected from the complete neglect of the filling of traps with lifetimes greater than the irradiation time. It may be noted that the same reasoning would give an hyperbolic decay law if the traps were filled to saturation rather than equilibrium.[23] Equations (2) and (3) are derived from the basic equations of trap dynamics.[24]

The light emission stimulated by visible or ultraviolet light reaches a maximum only several minutes (up to 20 minutes in the experiments performed) after the stimulation is initiated. This rise comes about because some electrons released from the s traps are first utilized for filling p traps instead of going to the emission level. Thus the stimulated light intensity would be expected to show a rise similar to that of the fluorescent light intensity if depletion of the electrons in the s traps by the stimulating light did not take place. If the stimulating light is very weak, the depletion in the s traps is small and a rise curve for the stimulated light rather similar to that of the exciting light is obtained. If the stimulating light intensity is strong, the s traps are rather quickly depleted and the light emission decreases long before the nearly steady value of the emission curve can be measured.

Since the stored energy which can be stimulated has a long lifetime and still amounts to 20 percent after 60 000 minutes, the s levels associated with the stimulation must have [from Eq. (1)] a depth below the excitation level of at least 1 ev. The long lifetime explains the linearity of the stimulated light with dose for quite different dose rates. From the preferable stimulation by light of energy greater than 5000A, one might be inclined to assume that the energy E_s of these traps is considerably more than 1 ev; the noticeable decay of the stored energy, however, indicates that this is not the case.

To explain this difference in the energy required by the stimulating light and the thermal release, one resorts to the often assumed shift in the curves of potential *vs* configurational coordinates between the s trap levels and the first excited band (Franck-Condon principle).[25] A similar effect occurs also in ZnS phosphors, but here for similar trap depths the wavelength of the stimulating light lies in the infrared.

Although this assumption not only explains the difference between the energy $h\nu$ of the observed stimulation band and the thermal de-excitation energy, but additionally explains the stimulation and de-excitation curves as functions of time (see below), it is felt that this explanation needs considerably more exploration. It is noteworthy that such a difference between the energy associated with the stimulation frequency and the thermal de-excitation energy occurs for trapped and stimulable electrons of many other materials. However, it is rather surprising that this effect is so marked in the NaCl(AgCl), where one assumes that the electrons are relatively free in the excited level, which would indicate a flat rather than a steep potential curve. A similar assumption about the potential curves also explains the shape of the de-excitation curves of Fig. 6 and the de-excitation shown in Tables I and V for visible and ultraviolet light. With progressing de-excitation the decrease is more gradual than exponential and is slower with visible than with ultraviolet light. If the energy range of the s levels were very narrow, an exponential de-excitation curve would be expected since all electrons absorb the stimulating light equally strongly. A deviation from an exponential law such as found would also be expected if retrapping decreased with increasing excitation of the crystal. Since, however, it was found that these de-excitation curves are essentially independent of the dose (Sec. IV), one must assume that retrapping is constant to a first approximation. If the s traps cover a wider energy range, light of frequency ν can empty only those traps with energies larger than a certain energy E_i with maximum efficiencies. The release of electrons from trap levels deeper than E_i will be much less probable, since the $h\nu$ of the stimulating light is not large enough to raise the electrons above the upper potential curve. On account of the space extension of the wave function, some releasing will still take place but with a very small probability. Thus one finds that at the beginning of stimulation de-excitation is rapid, but that it slows down considerably as soon as the electrons in the favorable energy range are exhausted; however, it does not stop completely since also the electrons in deeper traps are still released. This explanation also makes understandable the more pronounced slowing down with visible light.

From Table IV, which shows that after 40 days a

[23] H. W. Leverenz, *An Introduction to Luminescence of Solids* (John Wiley and Sons, Inc., New York, 1950), p. 269 ff.
[24] H. Kallmann and B. Kramer, Phys. Rev. 87, 91 (1952).
[25] P. Pringsheim, *Fluorescence and Phosphorescence* (Interscience Publishers, Inc., New York, 1949), p. 147.

major portion of the stored energy has decayed thermally, it can be estimated that the *s* traps have an energy depth around 1 ev and a width of not more than several tenths of an ev. On the other hand, conclusions about the range of the *s* trap band cannot be made from the width of the stimulation band, since the decay of the storage energy indicates that the *s* traps have a considerably smaller range than the width of the stimulation band. The width of the stimulation band must already be inherent to a single *s* level; this can also be understood from the potential curve picture and the considerable space extension of the wave function.

If the stored energy is already depleted to a certain extent by stimulation, it is observed that stimulation by visible light following one with ultraviolet light exhibits a larger stimulated intensity than a similar stimulation with visible light before the ultraviolet stimulation (Table V); these effects together with the arguments expressed previously in this section also indicate that considerable retrapping occurs. The traps easily emptied by visible light are depleted first. If then ultraviolet stimulation is applied, electrons in deeper traps are raised to the excitation level, and some are retrapped in those levels easily emptied by visible light, especially during the period of phosphorescence after the ultraviolet stimulation is discontinued. In this way the traps which are easily emptied by visible light are partially refilled, so that stimulability by such light is increased. An analogous argument explains the increase in ultraviolet stimulability after different short shots of ultraviolet stimulation (see Table V). It also makes understandable the observation with certain crystal specimens that after a long period of stimulation, a long dark period increases the stimulability with the same light.

The *s* traps are associated with the traps responsible for the coloration of the crystal, as was shown in Sec. X. We have, however, not investigated this relationship precisely enough to determine how the introduction of the activators modify the properties of the traps responsible for the coloration.

Another problem which we would like to mention is the following: It is generally assumed that the electrons in the excited band behave like free electrons hampered in their motion by traps. This would imply that the probability of recombination of the electrons with the positive charges accompanied by light emission should depend on the total strength of the excitation as is also assumed in ZnS. (Light emission there is proportional to the number of electrons in the conduction band multiplied by the number of positive charges.) Thus, as a consequence, the light emission curves (both fluorescent and phosphorescent) should depend on the degree of excitation. However, the experiments described above show no pronounced effect of this kind. This may be due to the production of excitons rather than free electrons.

The results derived from the small number of heating and cooling experiments are also in agreement with the presented model. Since the *s* traps are considerably deeper than the *p* traps, it was found that the phosphorescence was completely altered by changing the temperature, whereas heating up to about 70°C had only slight effects on the storage capability of the crystal. The rapid decrease in phosphorescent light emission, while the crystal was being cooled, is due to the fact that the *p* traps release much fewer electrons per unit time at lower temperatures.

The above considerations deal mostly with the dynamics of trap filling, emptying, and refilling, and with only roughly determined positions of the energy levels of the traps. They are sufficient to show that the situation is quite different from those found discussed in the theoretical papers of Williams and Johnson which deal mainly with thallium-activated potassium chloride.[26]

[26] F. E. Williams, J. Chem. Phys. **19**, 457 (1951); P. D. Johnson and F. E. Williams, J. Chem. Phys. **20**, 124 (1952).

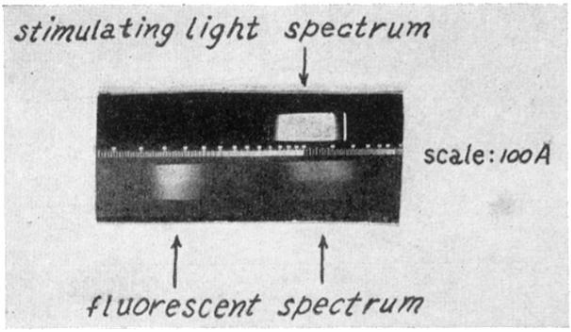

FIG. 2. Fluorescent spectrum of NaCl activated with 1 percent AgCl under gamma-ray excitation.