

The Inception of Photoelectric Scintillation Detection Commemorated after Three Decades

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The modern photoelectric scintillation detector emerged just 30 years ago out of the rubble of World War II. Kallmann in Germany had the original idea of combining an organic fluor (naphthalene), which is transparent to its fluorescent light, with an electron multiplier phototube for the detection of single scintillation events. Hofstadter at Princeton discovered that the inorganic thallium-activated sodium iodide scintillator has superior gamma detection efficiency and a high photoelectric yield for gamma spectrometry.

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The year was 1947 and Geiger-Müller (G-M) counters were widely used to detect the radiation emitted from radioactive tracers such as P-32, Fe-59, and I-131. With their thin mica windows placed close to the planchet containing a sample, a high counting yield was achieved. Following the work of earlier in-

vestigators, Marinelli and Quimby positioned special G-M counters over the patients' necks a day or so after they had been given a small oral dose of iodine-131, and thus obtained information about the state of thyroid function. When radionuclides became more readily available in 1946, adequate in-



Hartmut Kallmann



Robert Hofstadter

strumentation seemed to be on hand to perform many desirable *in vitro* and *in vivo* measurements. Biomedical scientists, therefore, were not searching for new ways to detect nuclear radiations. Nuclear physicists, engaged in more demanding studies, found the slow G-M counter, with its less than 1% efficiency for gamma-ray detection, a serious limitation. They were therefore constantly seeking improved radiation detectors.

HARTMUT KALLMANN AND THE FIRST PHOTOELECTRIC SCINTILLATION DETECTOR

In the March 1948 issue of *Nucleonics*, Martin Deutsch of MIT referred to work carried out in Berlin, where Professor Hartmut Kallmann had developed and reported on "Naphthalene Counters for Beta and Gamma Rays." Papers published in German journals were not generally available in the United States at that time. Whereas the conventional inorganic fluors such as zinc sulfide and calcium tungstate are largely opaque to their own fluorescent radiation, Kallmann found that most of the light produced in organic fluors, particularly naphthalene, escaped from layers as thick as 1 cm. Furthermore, by placing the fluor on the photocathode surface of an early electron-multiplier phototube he could detect individual scintillation pulses and relate their pulse heights to the energy of the incident radiation. With large blocks of naphthalene, a high gamma-counting efficiency could be achieved. Moreover, the scintillation pulses from naphthalene were of extremely short duration, which made the naphthalene detectors much faster than the G-M counters.

The difficulties endured in starting this new line of research were described to me recently in a letter from Kallmann, who celebrated his 80th birthday in 1976. Because of his opposition to the Third Reich, Kallmann's academic activities had been suspended in the early 1930s, but the I.G. Farben Company made a laboratory available to Kallmann in what had formerly been a horse stable. There, together with a co-investigator, Kallmann developed neutron photography, but he could not publish his results at that time because no journal would accept his papers. His discoveries were filed as patent claims by I.G. Farben, and today these patents disclose the bases for modern neutron photography.

These investigations made Kallmann aware that photography was possible with the aid of fluors, and also gave him the idea that an electron-multiplier phototube would be much more effective than a G-M counter for the detection of individual scintillations caused by neutron interactions with the fluor. At that time Kallmann had no access to an electron-multiplier phototube. Moreover, he did not want to announce his

idea because he clearly recognized that his findings might significantly facilitate nuclear investigations. He therefore locked it up in his desk drawer without mentioning it to anyone and waited for the end of the Third Reich.

After the war, late in 1945, Kallmann returned to his old laboratory at the University in the American sector of Berlin. He found that the advancing Russian armies had completely stripped it of all equipment. His only asset was a young and enthusiastic collaborator, now Professor Broser of Berlin. How could a fresh start be made? Equipment was not available in the totally bombed-out city except sometimes on the black market—and then only in trade for other goods, mainly American cigarettes.

First, Kallmann had to find a source of radioactivity. That seemed simple to him. Some walls were painted with lead paint which he realized might contain traces of radioactivity; so he and his co-worker set out to scrape the paint from the laboratory walls. The next problem was how to measure its radiation. Again Kallmann found a solution. Luckily, because there was no general use for them, moth balls or flakes were still to be found. Kallmann hunted for as many as he could find, melted them, and cast them into large transparent blocks of naphthalene. Finally, he was able to secure some photographic plates. On these he placed the naphthalene blocks and on the blocks he put the lead paint. In this way, he achieved more intensive blackening of the film than when he placed the paint directly on the plates. There was much laughter at such primitive experimental methods. When someone from the American Military Government visited the laboratory, he also found such simple efforts ludicrous. But Kallmann suggested that instead of laughing, he should help by supplying cigarettes. The American officer was noncommittal, but the next day 10,000 American cigarettes appeared on Kallmann's desk. That same afternoon Kallmann bought the long-sought electron-multiplier phototube on the black market. Additional instruments were obtained similarly, and thus the first photoelectric scintillation detector came into existence. Kallmann, who is a passionate nonsmoker, fondly recalls today that the brand name of the cigarette was—prophetically—*Lucky Strike*, and indeed it provided a lucky strike for science in general and nuclear medicine in particular.

Scientific publication in Germany was still very limited, but Kallmann demonstrated his scintillation detector at a special colloquium in Berlin-Dahlem in June of 1947, and a short note about the colloquium appeared in the July 1947 issue of *Natur und Technik*. A complete report of Kallmann's research reached the Massachusetts Institute of Technology, and in October 1947 Professor Martin Deutsch made it available, in translation, to the American scientific

community. Its title was:

“The Counting of High-Energy Particles and Quanta by Photoelectric Detection of Individual Light Flashes in Fluorescent Materials.”

In his introduction Kallmann wrote: “Very recently, a new method of detection has been developed for heavy charged particles as well as for electrons, beta particles, and gamma-ray quanta. It is based on the fact that they produce more-or-less pronounced light flashes in various fluorescent materials. In the early days of nuclear physics much alpha-particle counting of these scintillations was done visually, especially in Rutherford’s laboratory. This early counting was performed by visual observation of the very faint light

flashes generated by the impingement of alpha particles on zinc sulfide.

“In the method to be described, single scintillations are recorded automatically. This is achieved by letting phototube in which they initiate a current pulse of electrons. These pulses may be observed on an oscilloscope (Fig. 1) or they may actuate a thyratron counting circuit after only moderate amplification. Not only is the number of pulses recorded equal to the number of incident particles or gamma rays, but the height of the pulses permits conclusions to be drawn concerning their nature and energy. For any one kind of particle or photon the intensity of the light flashes is proportional to the expended energy. Furthermore, there is a very characteristic difference in the

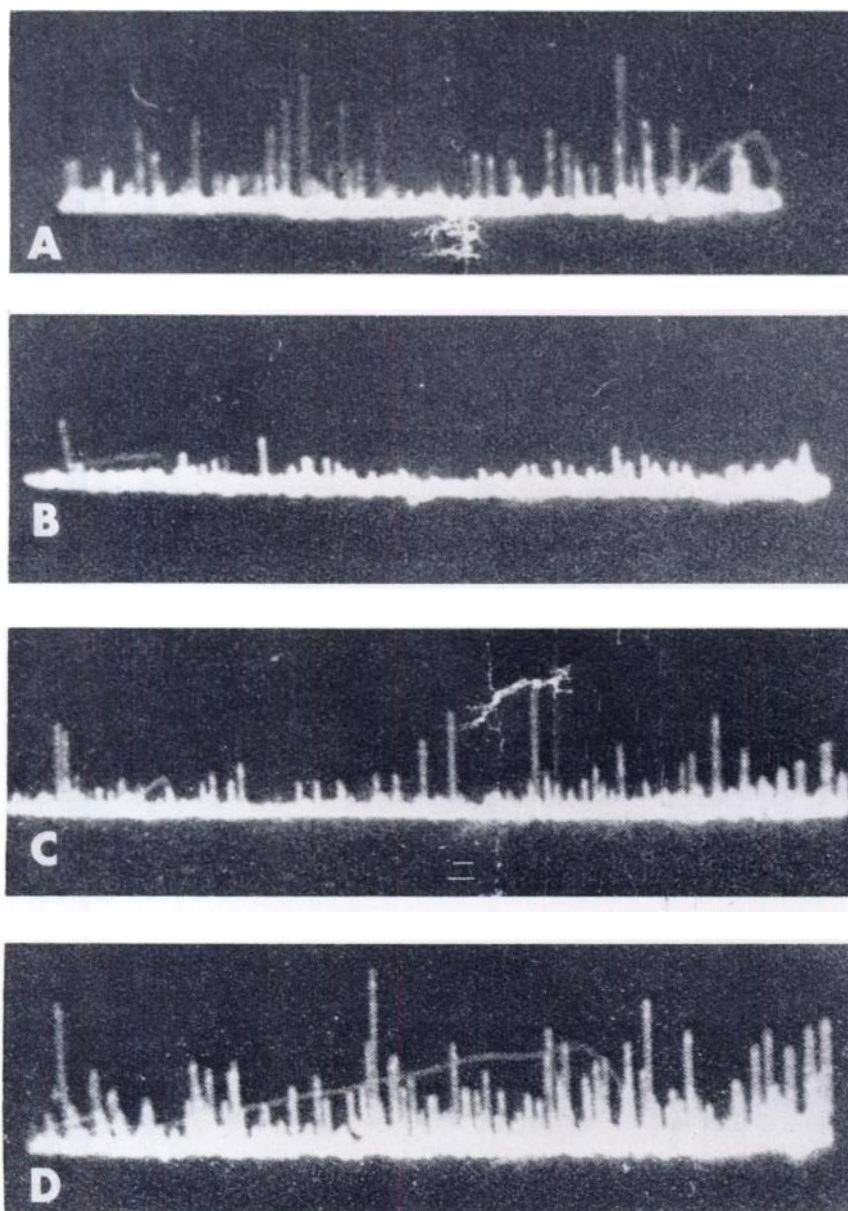


FIG. 1. Scintillation pulses as observed on oscilloscope. (A) Calcium tungstate excited by alpha particles. (B) Background pulses of naphthalene at -20°C . (C) Naphthalene excited by beta particles. (D) Naphthalene excited by gamma rays. (Courtesy of H. Kallmann)

behaviour of alpha and beta particles, which permits them to be distinguished without difficulty."

Kallmann went on to point out the importance of having many double bonds in organic scintillators, and of the necessity for the scintillator to transmit its own fluorescent light with negligible attenuation. In this respect he had found naphthalene much better than zinc sulfide. He also recognized the value of naphthalene's fast scintillations ($\ll 1 \mu\text{sec}$), and that the weakness of the flash could be lived with if the dark noise of the phototube were reduced by cooling it to around -40°C .

Finally, an abstract by Deutsch appeared in the May 15, 1948, issue of the *Physical Review*. It included a reference to Kallmann's work as reported in *Natur und Technik* the previous summer, and drew worldwide attention to the new method for detecting and analyzing nuclear radiations. Immediately Kallmann was swamped with letters and visitors from the United States, England, and France, and even from Russia he received greetings attached to a carton of cigarettes. Kallmann and "the World" had discovered the scintillation detector.

Kallmann came to New York University in 1949 as professor of physics and remained there until his return to Germany after retirement in 1968. In 1950 he reported another highly significant discovery (*Physical Review*, June 1, 1950): the possibility of liquid scintillation counting. During studies of the fluorescence of liquid solutions under irradiation, he had found that "special solutions" were efficient enough to serve for scintillation detection. "Solutions of toluene and xylene, in which were dissolved fluorene, carbozole, phenanthrene, or anthracene, proved to be nearly equally useful." Characteristically, he had at once recognized a very favorable combination of scintillator and solvent.

ROBERT HOFSTADTER AND THE THALLIUM-ACTIVATED SODIUM IODIDE SCINTILLATOR

As we all know, our well counters and gamma cameras do not employ naphthalene or liquid scintillators, since their scintillations are weak, their densities are low, and their low proton numbers give them poor photoelectric cross sections, all of which limits their usefulness in gamma spectrometry. Robert Hofstadter pointed this out in 1948 (*Physical Review*, July 1, 1948), and proceeded to describe his studies of alkali halide fluors:

"In tests made by placing crystals of NaI, KI, and naphthalene on photographic plates much greater light output was observed from NaI than from naphthalene samples of comparable size...." The inherent radioactivity of potassium iodide and its poor light output discouraged its use, and Hofstadter

turned his attention to powdered sodium iodide to which he added "a pinch of thallium halide." The consequences to the field of nuclear medicine were enormous.

At the Scintillation and Semiconductor Counter Symposium in December 1974, Hofstadter gave an address on "Twenty-five Years of Scintillation Counting" (1). He reviewed the early developments that led him to the discovery of the (thallium) sodium iodide (crystal) detector.

"Since 1938 I had developed an interest in luminescence, which is the ability of a material to light up when it absorbs visible or ultraviolet light. I was especially stimulated in 1947 when I heard rumors that Hartmut Kallmann had used naphthalene (moth ball material) to detect gamma rays by their scintillations in this material. I reasoned that if moth balls worked as scintillation detectors, other materials might give even better performance.

"At the General Electric Laboratories I had worked with Mr. Frank Quinlan, a technician, who was carrying on some research on thallium-activated potassium iodide. Thallium activation means that a small amount of thallium salt is added to a melt of the host crystal, in this case, KI. I was attracted by the clarity and beauty of Quinlan's crystals. Nine years later, *because of some unexplained urge*, I asked Mr. Quinlan for some of his crystals, which he kindly gave me. Deutsch's reporting of Kallmann's work reminded me that the KI (Tl) might be a material to try as a scintillation counter because of its known luminescent behavior. The first try showed that KI (Tl) scintillated just like naphthalene."

A little later, some "rather obscure German papers" suggested to Hofstadter that sodium iodide might well fluoresce, and also be transparent to its own fluorescence. He obtained some reagent-grade sodium iodide powder, melted it, doped it with a bit of thallium halide, and cooled the mix to a glaze. Tests demonstrated the material's superior luminescence, but did not prove that it would scintillate. So Hofstadter prepared a polycrystalline sample of the NaI (Tl) and sealed it in a 1/2-in. quartz test tube to protect it from atmospheric moisture. His description continues: "I strapped the sample of NaI (Tl) to a multiplier phototube with an aluminum reflector and connected the phototube to high voltage and an amplifier (Fig. 2). On the first trial I obtained very large pulses on the oscilloscope! I was amazed to see that when I tried naphthalene under comparable conditions, I could barely see the naphthalene pulses compared with NaI (Tl) pulses. Now I knew I really had something very good."

At the time of his first experiments with sodium iodide, Hofstadter was an assistant professor of physics at Princeton. In 1950 he moved to Stanford

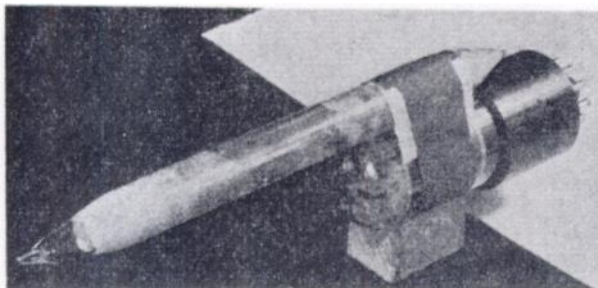


FIG. 2. Mounting arrangement of the first sodium iodide scintillation counter. (Courtesy of R. Hofstadter)

University. In recognition of his work in high-energy physics, he shared the 1961 Nobel Prize in physics. He still takes an active interest in the development of nuclear medicine and attends some of the meetings of the Society of Nuclear Medicine, but few of the members recognize him.

Thus, the beginning of photoelectric scintillation detection can be traced back three decades to Kallmann and to Hofstadter, who initiated it. Since then, many investigators have participated in improving not only the scintillator but the associated multiplier phototubes. The latter went through many stages of basic improvements, of which the introduction of the "tea cup" dynodes became the latest refinement in 1976. The circuitry developed for G-M counting was initially adopted for scintillation counting, but slowly there evolved stable and much faster circuits that were especially suitable for scintillation counting. They incorporated a pulse-height analyzer as the most important improvement.

Kallmann and Hofstadter each discovered his scintillator by design and not by chance, and during nearly 30 years, no superior scintillator has evolved. Kallmann's combination of a scintillator and electron-multiplier phototube has become an integral unit that functions reliably for long periods of time.

Hofstadter's thallium-activated sodium iodide crystal is not an ideal scintillator chiefly because it is too slow to match modern electronic circuitry. More-

over, its energy resolution for a standard monoenergetic gamma emitter is only about 10%, compared with better than 1% for semiconductor detectors such as lithium-drifted germanium. But the latter requires liquid-nitrogen cooling, and its efficiency for gamma detection is much lower than that of NaI (Tl). Both of these failings impose serious limitations for the application of semiconductor detectors in nuclear medicine. Because NaI(Tl) is hygroscopic, the scintillation crystals must be "canned" hermetically to exclude even small amounts of moisture. Also, sodium iodide has a high optical index of refraction, and special precautions are required to provide effective light collection with minimal loss of light by internal trapping. Despite these limitations, sodium iodide not only is the gamma-ray detector of choice for nearly all applications in nuclear medicine, but currently there is nothing else in sight that is likely to match it with respect to size and detection efficiency.

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FOOTNOTE

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REFERENCES

1. DEUTSCH M: Naphthalene counters for beta- and gamma-rays. *Nucleonics* 2: No 3, 58-59, 1948
2. BLOCH W: Kann man Elektronen sehen? *Natur und Technik* 1: No 13, 15-17, 1947
3. DEUTSCH M: High-efficiency high speed scintillation counters for beta- and gamma-rays. *Phys Rev* 73: 1240, 1948
4. KALLMANN H: Scintillation counting with solutions. *Phys Rev* 78: 621, 1950
5. HOFSTADTER R: Alkali halide scintillation counters. *Phys Rev* 74: 100, 1948
6. HOFSTADTER R: Twenty-five years of scintillation counting. *IEEE Trans Nucl Sci* 22: 13-35, 1975