

THE CRYSTAL COUNTER

A NEW APPARATUS IN NUCLEAR PHYSICS FOR THE INVESTIGATION OF β - AND γ -RAYS. PART I

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Synopsis

Experiments show that the ionization of β - and γ -rays in *crystals* such as silver chloride is so large, that the ionization current of a single particle can be observed and can be measured with the aid of a pulse amplifier. The possibility of determining the energy spectrum of the radiation by this method is discussed. Experimental details and preliminary results are given.

Introduction. If a radioactive source emits β - or γ -radiation, it is of interest to know the intensity and the energy distribution of this radiation. One can measure the energy spectrum in a magnetic beta spectrograph fitted with a Geiger-Müller counter or a photographic plate as recording apparatus, or in a cloudchamber placed in a magnetic field. The curvature of the paths of the particles (which in the case of γ -rays means: the secondary β -rays, caused by photoelectric- or Comptoneffect) is a measure for the energy of the radiation. These methods, in particular the first one allow the accurate measurement of the energy, but make it necessary to use strong radioactive sources. If the intensity of the radiation of the substance investigated is small, which is often the case for artificially activated substances, these methods are impracticable. In that case one can only study the absorption of the radiation in matter. For β -rays, the absorption coefficient is a measure for the energy. In the case of γ -radiation, one can investigate the absorption of the radiation itself, or still better, the absorption of the secondary electrons, freed from e.g. a thin layer of aluminium. The results of this method are not very accurate, especially because, if the radiation is inhomogeneous, we cannot draw any conclusion about the energy-distribution.

The ionization as a measure for the energy. When we have to deal with α -rays, we have, in addition to the methods mentioned, another one for the measurement of the energy: the total number of ions, freed by an α -particle in a gas, is a measure for the energy of the particle. When we collect these ions on the electrodes of an ionization chamber by an electric field, and we measure the voltage pulse caused on one of the electrodes, with the aid of a pulse amplifier coupled to a cathoderay oscillograph or a galvanometer as a recording instrument, the deflection of the instrument will be a measure for the energy of the α -particle. In the case of β -radiation, this method cannot be employed. It is true that the ionization of a single β -particle in a gas is large enough to be observed, but the path of a β -ray in a gas at ordinary pressure is too large (several meters) to lie entirely within an ionization chamber of good dimensions. M a s i n g has worked out a method to measure the so-called specific ionization ¹⁾. But even if it should be possible to make this a useful method, which has not yet been accomplished, then (owing to the large error and the small solid angle that can be used) it has still the disadvantage that the specific ionization is not a good measure for the energy in a large part of the spectrum.

Principle of the new method. If it is possible to find substances, in which a β -particle is stopped over a short distance, while the ionization is still sufficiently strong to be measured, this would provide a means to analyse a β -radiation of small intensity. This leads to the question: what will be the ionization produced by β -rays in liquids, compressed gases and solids?

The ionization in liquids was studied by J a f f é ²⁾. In substances such as hexane, carbondisulfide and carbontetrachloride, the ionization turned out to be about the same as in a layer of gas of the same weight. However, recombination of ions will occur, so that the ionization current we measure, is, even in carefully purified substances, only $\frac{1}{3}$ to $\frac{1}{10}$ of the real ionization. This would perhaps still be enough, but the long collection time of the ions in liquids contributes to make the measurement rather inaccurate. In liquids of this type, therefore, the method if possible, would be very difficult and inaccurate.

Compressed gases behave, in this respect, much like these liquids. Long collection time and recombination of ions form a serious handi-

cap. These facts, together with the difficulties of the high pressure ³⁾ make this possibility not very attractive.

In *solids* in general, the ionization current is very small, owing to the small mobility of the ions ⁴⁾.

Now the idea which underlies the present investigation is that the ionization of a single β -ray may be well-observable in a special kind of solids: namely, *crystals* that show photo-electric conductivity. Inside these crystals, light is able to free electrons which, in some of these, can move over a distance of several millimeters, if we apply an electric tension to the crystal; why should β -rays not do the same? The action of β - or cathode-rays in photo-electrical crystals has been investigated by only a few authors ⁵⁾. It has been found that in diamond and in zincblende an electric current sets in indeed under the influence of cathode-rays, and that this current consists of electrons, moving through the crystal, as is also the case by the action of light. It was not known whether this effect could be made large enough. However, experiments were performed, from which one can deduce, that this is the case: The direct blackening of a photographic plate by X-rays was measured and appeared to be very large ⁶⁾. One silver atom was formed per 30 electronvolts of energy absorbed. Considering in the first place, that the interaction of X-rays and matter can only take place by the formation of secondary β -rays and, secondly, that according to the theory of G u r n e y and M o t t ⁷⁾ every silver atom in the latent image originates from an electron in the conduction band, we can conclude, that in the process of the conversion of the kinetic energy of β -rays into energy for the liberation of electrons in the silver bromide, 30 eV or less are required per electron.

Another indication is given by the measurements of the efficiency of cathode rays in producing luminescence of crystals (up to 10%) in view of the fact, that according to the theory of R i e h l and S c h ö n ⁸⁾, luminescence is closely related to the photo-electric effect.

We investigated silver chloride experimentally and found, that the ionization produced by β - and γ -rays is indeed large enough. When we put a voltage on the electrodes, applied to a crystal of silver chloride (cooled to liquid air temperature), and we connect one of the electrodes to a pulse-amplifier, the pulses produced by β - or γ -radiation from a radio-active source are visible on the recording

apparatus. In principle, therefore, the method has proved to be practicable.

Some considerations about the photo-electric effect. When we put a voltage on a crystal that shows photo-electric effect, and we irradiate it with light, an electric current starts in it: the photo-electric current. Crystals of this kind have been systematically studied by P o h l and his fellow-workers ⁹⁾. The most important results of their investigations are the following: the photo-electric effect is a quantum process. A quantum of light can free an electron from an atom in the crystal, and this electron can move freely through the crystal, until it is captured again in an impurity or an irregularity of the lattice.

Let (fig. 1) the component of its way in the direction of the field be x , and the thickness of the crystal d , then the charge Q , induced

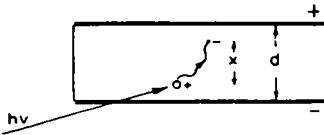


Fig. 1. Internal photo-electric effect.

on the electrodes, is $Q = (x/d)e^9$ (e being the charge of the electron). The positive charge that is left behind is also apparently able to move, ("*positiver Primärstrom*")", sometimes spontaneously by thermal activity, as in the case of zincblende, at other times only when irradiated by red light, e.g. in the case of diamond or cooled silver chloride. x has turned out to be proportional to the field strength, so that at first the photo-electric current will show a linear increase with increasing voltage, until finally all electrons reach the anode. The current has then reached a saturation value. H e c h t ¹⁰⁾ has made it clear by his experiments on silver bromide, that the electrons move in the direction of the field with a certain chance (per unit of length) to be absorbed, quite analogously to a beam of light that passes through an absorbing medium. The number of electrons, liberated in the same horizontal layer of the crystal, decreases exponentially with the distance in the direction of the field. G u d d e n and P o h l ⁹⁾ have shown that in the case of diamond and zincblende, for light of certain wavelengths every quantum of light absorbed sets free an electron, so that in this case the efficiency of the photo-electric process is equal to unity. For these crystals,

however, this was only true for certain wavelengths in the area of small absorption, the existence of which must be ascribed to impurities in the crystal.

In the area of strong absorption in the ultraviolet, originating from the atoms of the lattice itself, the efficiency turned out to be very small or zero. In contrast to these experiments, *L e h f e l d t*¹¹⁾ found, that for silver- and thallium halides the efficiency of the quantum process is also unity in the wavelength area of the absorption of the lattice. Now in view of these facts, one could reason as follows: in diamond and zinblende the photo-electric effect is caused only by the absorption of light in impurities. If we assume that the energy given off by a β -particle to an atom, produces the same action as an absorbed quantum of light, we cannot expect a large ionization to occur, when we bombard these crystals with β -rays for a β -particle will give off its energy mainly to the ions of the lattice, which do not show the effect. Meanwhile the chance of the particle hitting an impurity, from which an electron can be set free, is small. We may, however, expect good results in crystals of the type studied by *L e h f e l d t*.

Considerations of *S l a t e r*¹²⁾ make it probable, that in the far ultraviolet region a photo-electric effect of large efficiency will be found, for diamond and zinblende as well, in analogy to the phenomenon of excitation and ionization of an atom in a gas. This has not yet been found experimentally, but if we assume it to be true, we must expect an ionization by β -rays in these crystals of the same order of magnitude as in the case of silver chloride. According to theory, there is not even an essential difference between those crystals that show photo-conductivity and those in which up till now, no photo-conductivity has been found. Theory therefore predicts that all crystals will show photo-conductivity when irradiated with light-quanta of sufficient energy. The only difference lies in the distance over which the electrons can move before being absorbed again. Consequently the ionization of all isolating crystals by β -rays must be of the same order, though the ionization current may be different owing to a larger or smaller path covered by the conduction-electrons before being absorbed. In a preliminary experiment we have investigated one specimen of zinblende but could not find an effect caused by radio-active radiation, though the crystal showed a photo-electric current which could be made saturated.

Experiments with other, better samples are desirable.

A discussion of the new method. The new method we suggest, consists in the following. We apply thin electrodes to a plate of silver chloride and connect one of these electrodes to a voltage source, the other to the usual type of pulse amplifier. When we irradiate the crystal, through the thin electrode, with β -rays, a number of pulses will be recorded. The statistics of the magnitudes of the deflections of the recording instrument will then give a good picture of the energy distribution of the radiation. Now let us consider whether we may expect this to be true, that is, we have to discuss whether β -rays of the same energy will produce the same ionization current. In the first place, a β -ray will lose its energy not only by inelastic collisions, but also by radiation. This radiation will partly escape from the crystal so that it will be lost for ionization. If we make an estimate of this effect from calculations by B e t h e and H e i t l e r ¹³⁾ we find, that for energies up to 2 MeV this effect is proportionally small and will not form a serious handicap to the method. For β -rays of an energy of 2 MeV, the average loss of energy by radiation in silver chloride is about 5%. In the second place there will be fluctuations in the number of ions formed by a β -ray of fixed energy owing to the statistical character of the process. The magnitude of this effect is difficult to estimate. For α -particles it has been measured and found to be about 1% ²⁰⁾. For β -particles it will probably be larger, owing to the fact that the secondary electrons formed by a β -particle have on an average a larger energy. It will form the natural limit to the width of the spectral lines detected by this method. If we assume that β -particles of the same energy all set free the same number of electrons, that means, if we do not take into account the effects mentioned, the ionization current we measure will still not be the same for all different β -ray energies (fig. 2). The β -particle will be scattered in the crystal so that the conduction electrons of different particles will not be formed in the crystal at the same depth. The total charge we measure $Q = \sum_i ex_i/a$ is not the same if the electrons can all reach the anode (we assume that the positive charges do not move, which is the case for silver chloride at low temperature). This effect can be made small by using a crystal thick in proportion to the depth of penetration of the β -rays, so that the difference in the distance to the anode can be neglected, and also by applying a voltage below the saturation voltage, so that the majority of the

conduction electrons do not reach the anode and the free path will not depend on the place of formation. Both methods are limited by practical circumstances. The saturation voltage in the crystal increases with the square of the thickness, while working below saturation voltage has the disadvantage of reducing the voltage-pulses relatively to the level of the noise of the amplifier. The circumstances we choose will therefore always be a compromise in order to make all

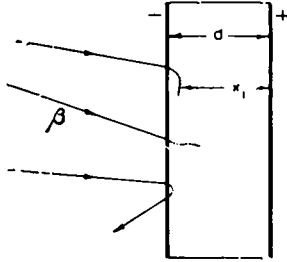


Fig. 2. Paths of β -rays in the crystal.

influences as small as possible. Some β -particles will be reflected, after having lost only part of their energy, so that these particles produce a smaller number of conduction electrons. One can avoid this by placing the radio-active source between two parallel crystal-plates of the same kind. A β -particle when reflected in one crystal, will lose the rest of its energy in the other.

A source of errors which of course is always present, is the noise of the amplifier. Owing to this ionization currents of exactly the same magnitude do not cause deflections of the same size of the recording instrument, but these deflections show a distribution around a mean value, and deflections which are too small cannot be detected at all. We shall discuss this effect which has been investigated by several authors ¹⁷⁾, in detail elsewhere.

Gamma-radiation. We shall now discuss what we may expect when we irradiate the crystal with γ -radiation. γ -radiation does not ionise the crystal directly, but only by its secondary β -rays. This secondary β -radiation is formed throughout the crystal. As we have seen in the case of saturation voltage, the charge induced on the anode depends on the distance between the place of formation of the conduction electrons and the anode. As it is necessary that β -particles of the same energy, liberating the same number of electrons, always induce the same charge of the anode, we must apply a voltage below the saturation value. With the aid of formulae, derived by H e c h t ¹⁰⁾

we can calculate that we obtain already satisfactory results, if we make the path x of the conduction electrons equal to $\frac{1}{4}d$.

The secondary β -radiation is formed by the photo-electric effect, Compton effect and pair-formation. In the first case, the γ -quantum is totally absorbed in an atom which emits an electron with the whole energy of the γ -quantum, minus the binding-energy of the electron (this binding energy, which is emitted as X -radiation may also be absorbed in the crystal and cause ionization). The photo-electrons of a certain γ -radiation must therefore, give the same ionization. The secondary β -radiation caused by the Compton effect has a continuous energy distribution of smaller energy than the photo-electrons. The pair-formation gives an electron-positron pair, of which the sum of the kinetic energies is defined by the energy of the γ -quantum. It is therefore probable that the ionization by processes of pair-formation from a certain γ -radiation will not show large differences, so that we may expect the statistics of the record of a homogeneous γ -radiation to consist of a broad continuum with two peaks. To make these peaks, — originating from the photo-electric effect and pair formation —, as high as possible, we must use a crystal containing a heavy element, in which these effects are large relatively to the Compton-effect. For this reason thallium bromide seems very suitable. Of course it would be still better to make the crystal of such dimensions, that the γ -quantum when passing through the crystal, is completely absorbed. This is not impossible, as crystals of one litre content have been made ¹⁴⁾. For measuring absolute intensities of γ -rays, the crystal counter is probably to be preferred to the Geiger-Müller-counter. The efficiency of a G.-M.-counter is difficult to determine ¹⁵⁾, because it is defined by a surface effect. For the crystal counter the chance of excitation is a volume effect, while the surface effect appears as a correction, which can be made small by choosing crystals of large size.

The experiments. The crystals used were of silver chloride, prepared from silver nitrate and hydrogen chloride. We made large crystals by the method of Kyropoulos ¹⁶⁾ (we also tried thallium bromide but did not yet succeed in making good, large size crystals of it). This method is, in short, the following: The salt is melted in a crucible of porcelain, in an electric oven (450°). Then we put a cooler (we used one of hard glass) into the liquid silver chloride. On this cooler a lump of solid AgCl grows. This lump in the shape of

a tondstone is ground on the lathe into a cylinder-shaped plate of the dimensions we need. Then the crystal is tempered^{10) 11)}, that is: for some time heated to a temperature of 400° to take away all discolouring caused by working in daylight. After that the crystal is handled only in red light (discolouring by light causes a decrease of the free path of the conduction electrons). Then we apply the elec-

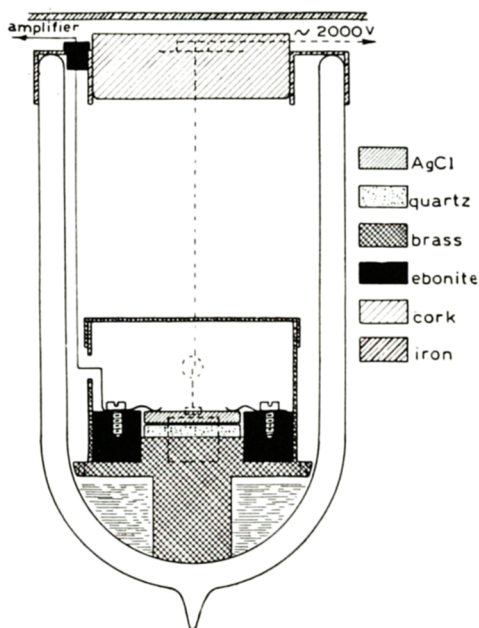


Fig. 3. Dewar vessel with brass box holding the crystal.

trodes by silvering the flat sides of the crystal plate. Now the crystal is ready to be mounted. The silver chloride crystals can only be used when they are cooled to liquid air temperature. At room temperature the crystal shows ionic conduction. Electrolytic silver is formed when we apply a voltage to it: the so-called "T u b a n d t-sche Faden", which cause sudden irregular changes in the conduction. At -186°C the crystal is a perfect isolator. Conduction along the surface does not occur either, so that no guard-ring is necessary.

Our apparatus for the cooling of the crystal was different from that of the other investigators of silver chloride. H e c h t and L e h f e l d t cooled the crystal in vacuo. We mounted it in a brass box at the bottom of a D e w a r vessel (fig. 3). The foot of the box, sur-

rounded by cotton wool (to prevent bubbling of the liquid air, which causes "valve microphonics" of the first tube) was cooled by the liquid air and so was the whole box, via the foot. The crystal was isolated from the box by a piece of quartz, which is a good conductor of heat, and fixed with silver clips (a base metal affects the crystal and soon renders it useless). The crystals used were about 4 mm thick and 4 cm in diameter. They were first tested by light, to see if the photo-electric current showed saturation for the voltage we used (up to 2500 Volt). If the crystals were right at first but showed no saturation after several weeks of use, it was sufficient to temper them again. For investigation by light we always used short flashes of light (about 10^{-3} sec) produced by a shutter; by using the pulse amplifier in that way, it was possible to measure very small effects, as then no polarization of the crystal occurred, and irradiation by red light ¹⁰⁾ ¹¹⁾ was superfluous. An estimate shows that in the case of radio-active radiation, the polarization is also negligible, if the amount of radiation is not too large. The top of the brass box was polished to prevent heat absorption from thermal radiation. The Dewar vessel was closed by a metal cover, in which ebonite supports for the connecting leads were made. In the centre was a cork which could be removed to pour in the liquid air and to introduce radioactive sources. The Dewar vessel was placed in an iron box, in which the first valve of the amplifier and the voltage source, a condenser of $2\mu\text{F}$ of good quality, were also fitted. The voltage pulses from the first valve were applied to a two-stage amplifier which was connected to a cathode ray oscillograph (Philips G.M. 3156). The spot in the screen of the oscillograph was projected by a lens on a cylinderlens of short focal length (a glass rod of 3 mm diameter) which was pressed against the photographic paper on the recording drum. The record had to be made in such a way that large statistics could be measured quickly and easily. To that end the amplifier and the oscillograph were adjusted in such a way that only the tops of the deflections were projected on the paper. (For the construction see ¹⁷⁾). In this way it was possible to collect a large number of deflections on one record, which is convenient both for recording and for calibration. This method of recording has still another advantage: when the photographic paper does not move, we obtain a spectrum of the deflections. Then the photographic density in a certain point is a measure for the number of deflections. Fig. 4 shows a record; 4A is

the background-effect, probably caused for the greater part by cosmic rays. Its intensity is very high. As the surface of the crystal is about 10 cm^2 , we find an intensity of $15 \text{ cm}^2 \text{ min.}$, which is much larger than that measured by Geiger-Müller counters¹⁸⁾. It

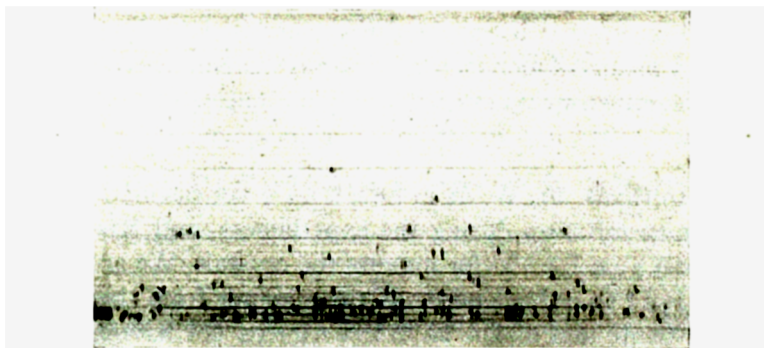


Fig. 4a. The background during one minute.

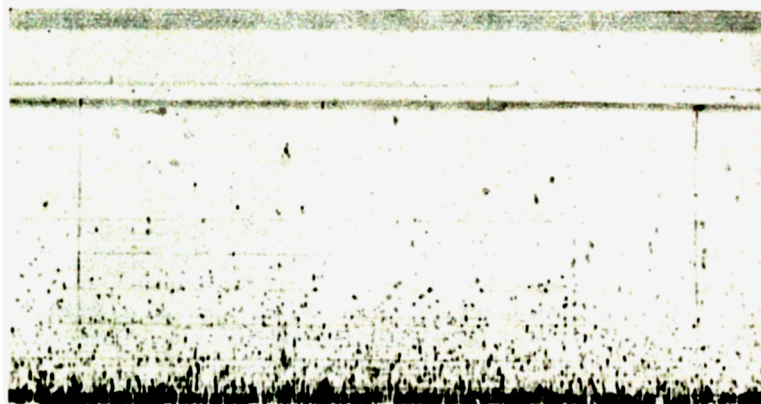


Fig. 4b. The effect of the γ -radiation of radium.

is possible that the increase can be ascribed to γ -radiation, for which the crystal counter has a larger excitation probability than a Geiger-Müller counter. On the record 4B one can also see a group of artificial deflections, obtained by applying voltage pulses¹⁸⁾ to a small well-isolating condenser (1 cm) which was permanently connected to the grid and by which the apparatus can be calibrated during the operation.

Fig. 4B is the record of the γ -radiation of radium ($B + C$). With this apparatus we have taken many records of radio-active sources of which the radiation was known. E.g. the γ -radiation of Ra($B+C$), the β -radiation of uranium and the β - and γ -radiation of thorium ($B + C$). The interpretation of the statistics obtained was, however, not yet clear, so that it appeared necessary first to investigate the crystals by means of monochromatic β -rays in a magnetic β -spectrograph. The experiments are still going on and will be published in due course.

I wish to thank Mr. W. Milt en b u r g for his kind assistance and welcome criticism during the experiments, and Mrs. E r i c a V e l t h u i j z e n - L e s k for making the crystals. We are indebted to Prof. R u t t e n for lending us the minerals from the collection at his Institute and to Dr. Th. S t r e n g e r s for having prepared for us the silver chloride.

In conclusion I wish to thank Prof. M i l a t z for the liberty he always gave me to carry out my plans, and for discussing the results with me.

Received 7-4-44.

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