Light Pulses Excited by α Particles in Argon. A Gaseous Scintillation Detector.

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Summary. — The light pulses produced by the passage of individual α-particles through gases have been detected and analyzed by means of a suitable experimental technique. In Part I, the time shape of pulses obtained with argon of various purities is studied with respect to the mechanism of photon production. In Part II, we deal with the possibilities of the particle detectors based on the gas scintillation. A scintillation chamber filled with argon was found to be suitable for the spectral analysis of α-particles, yielding a 13% resolution at ~ 5 MeV, for instance. The resolving time of this instrument is a very short one.

Introduction.

The luminescence caused by the passage of α-particles through gases has been studied in many works with the method of measuring the integral intensity of the light produced by a beam of particles ([1]).

With the progress of the photomultiplier technique the accuracy of such measurements was improved ([2]).

An interesting development in these works, made possible by the high sensitivity of photo-multipliers, consists in the method of revealing the luminescence produced by individual α-particles in the gas under consideration. This was done by MUEHLHAUSE in 1953 ([3]) who utilized both the visible and the UV component of the luminescence, by covering the phototube surface with a proper wavelength shifter (stilbene or NaI).

([1]) R. AUDUBERT and S. LORMEAU: *Compt. Rend.*, **228**, 318 (1949).

([2]) A. GRÜN and E. SCHOPPER: *Zeits. f. Naturfor.*, **6a**, 698 (1951); A. WARD: *Proc. Phys. Soc.*, **67a**, 841 (1954).

([3]) C. MUEHLHAUSE: *Phys. Rev.*, **91**, 495 (1953).

The aim of the present work is to describe and discuss the measurements made by us in the case of argon of various purities.

The first part concerns the study of the α-pulse shapes with respect to the mechanism of photon production, and the results are considered in connection with those obtained in previous researches on photons produced in argon excited by Townsend avalanches.

In the second part some possible applications of this method in the realization of α-particle spectrometers are described and the possibilities of such instruments in comparison with those of conventional types are discussed.

Study of Light Emission.

1. – The Scintillation Chamber.

These measurements have been made with a cylindrical scintillation chamber 4 cm high and of 5 cm diameter (Fig. 1.). The walls are stainless steel and are lined internally with copper sheeting covered with white enamel that serves as light diffuser.

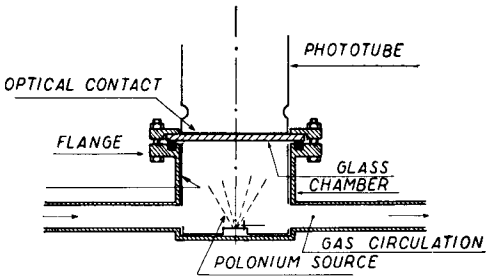

Fig. 1. – Scheme of the chamber and geometrical arrangement used for the measurements of the α scintillations in gases.

The chamber is closed from above with glass. Vacuum and pressure are assured by means of a flange with a polyethylene gasket.

On the external face of the glass disc is placed the head of the photomultiplier.

On a support at the bottom of the chamber, a thin layer of a few sq. mm which emits a 1000 α-particles per minute, is deposited. This source can be screened with a small disc, driven from the outside by a magnet.

2. – Vacuum and Filling Apparatus.

The chamber is connected with an apparatus which serves to evacuate the chamber itself, to purify the argon and to prepare gas mixtures.

This apparatus is shown in Fig. 2. Its containers and conductors are made of metal: so are the valves, joints and gaskets. The use of grease and mastic is avoided.

A silicone oil diffusion pump, connected in series with a rotating pump permits a vacuum of 10^{-5} mm_{Hg}, which is measured by a Penning ionization gauge. The pressure of the gases introduced in the apparatus is measured with a Bourdon manometer.

The scintillation chamber is inserted in a circuit of conductors in which the

gas is made to circulate. This circuit includes a furnace containing turnings of calcium and magnesium alloy (90% Ca - 10% Mg) which, if heated to 400-500 °C, allows the circulation and the purification of argon.

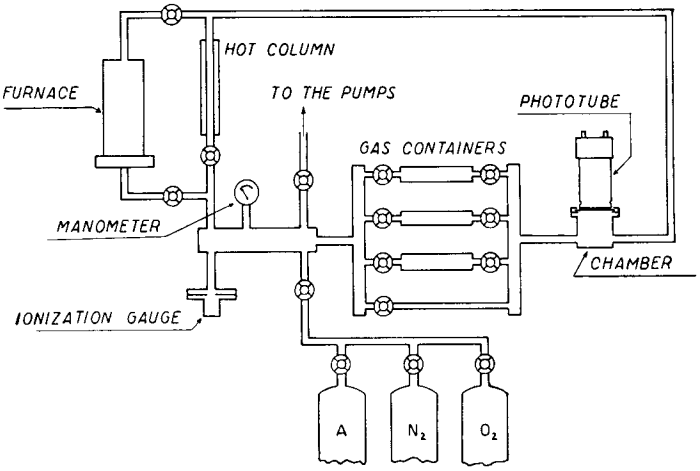

Fig. 2. – Filling and mixing apparatus, with a purification circuit.

It also includes a series of containers in which it is possible to enclose other gases that are to be mixed with argon.

When it is needed to use a mixture containing gas that can be adsorbed by the furnace, the latter is eliminated from the circuit and in such case the gas circulation is maintained by an electrically-heated vertical column inserted parallel to the furnace. Thus a rapid mixing of the introduced gases is obtained.

3. – Conversion of UV Photons.

When excited with α-particles the gases studied by us emit photons that are mostly ultraviolet. To convert these photons into light detectable by the photomultiplier, sodium salicilate has been used, as a fluorescent substance. This salt, excited by UV light, emits fluorescent light in the blue region of the spectrum ([4]), to which the photomultiplier used by us has a good relative sensitivity. The fluorescence quantum yield is practically constant for an exciting wave length of 850 to 2500 Å ([5]). Sodium salicilate has been applied as a methyl alcohol solution on the enamelled surface of the light diffuser.

By evaporating the solvent, a layer, almost opaque, was formed, that stuck well to the surface. The internal surface of the glass that closes the chamber

([4]) J. De Mont: *Fluorochemistry* (New York, 1945), p. 201.
([5]) E. Inn: *Spectrochim. Acta*, **2**, 2 (1955).

was coated with a similar technique by a very thin and transparent layer of sodium salicilate.

The layers described remain unchanged over a long time even in vacuum, due to the negligible vapour pressure of sodium salicilate.

4. – Electrical Devices and Measuring Methods.

The light emission produced by individual α-particles has been registered with a photomultiplier E.M.I. 6262. The electric pulses at the output of the photomultiplier have been studied with respect to amplitude and duration. For measuring the amplitude, the pulses were sent to a conventional amplification chain through a time constant $RC \cong 200$ μs which is much greater than the duration of a single pulse, that never exceeded a few μs.

The corresponding voltage pulses at the output of the amplifier were measured in Volt by means of an oscillograph Cossor, model 1035.

The time development of single pulses was studied by using a faster amplification chain, consisting in a cathode follower and an amplifier model 600. The rise time of this chain was $\sim 5 \cdot 10^{-8}$ s and the time constant at the input could be variated from $1 \cdot 10^{-7}$ to 10^{-6} s in order to obtain a suitable pulse resolution.

The pulses at the output of the amplifiers have been observed, depending on their duration, by means of an oscillograph with synchroscope Dumont mod. 248, using sweep durations of 5 and 25 μs, or by means of a faster oscillograph, using sweep durations of 0.2, 0.6 and 2 μs.

A few pulses of each type were registered photographically.

5. – Experimental Results with Pure Argon.

For these measurements the apparatus was filled with argon of a purity exceeding 99.9%.

Throughout a series of measurements the argon was made to circulate through the furnace described. It is known (6) that by this method the argon is purified of most of foreign gases and is kept from contamination with organic vapours, so that it reaches a point where the fraction of molecular impurities is some 10^{-4} or less.

The pressures used were of 80-100 cm_{Hg}.

Under these conditions, pulses of α-particles were observed and measured that were much higher than the dark pulses of the photomultiplier, with good reproducibility.

By comparing the pulses of individual α-particles with dark pulses due to single electrons from the photocathode, the former were estimated as due to about 25 photoelectrons each.

The rise time of pulses observed with the oscillograph was found to be about $1.5 \cdot 10^{-7}$ s at the indicated pressure.

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(6) L. Colli and U. Facchini: *Phys. Rev.*, **88**, 987 (1952).

The study of the pulse duration was carried out with various pressures between 50 and 100 cm_{Hg}. It was found that for this range the decay time of pulses was constant within experimental errors. The value thus obtained was $2.5 \div 3$ μs (Fig. 3).

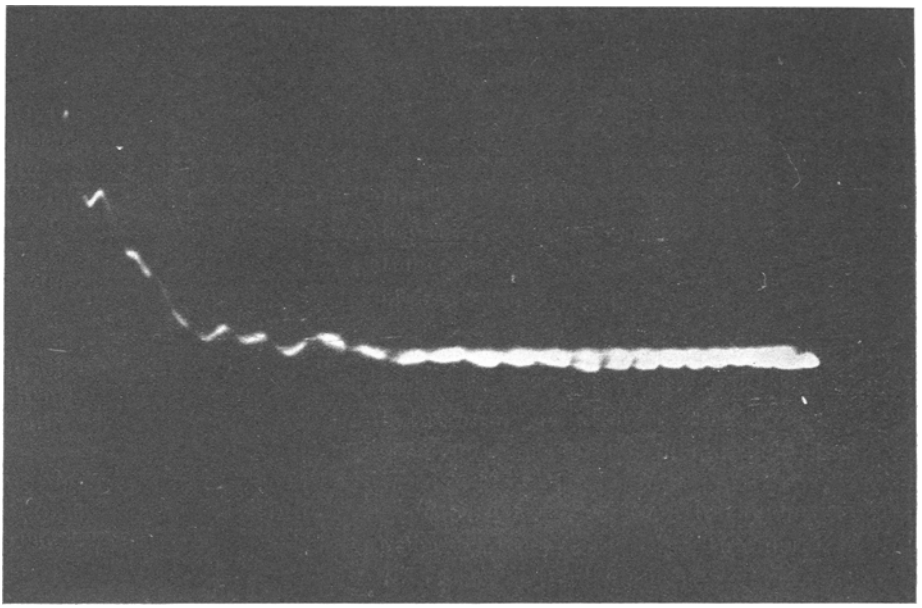

Fig. 3. – The decay of an α pulse in pure argon, 80 cm_{Hg}, showing a decay time of ~ 3 μs (time markers: 1 μs each).

6. – Discussion of Results.

The order of magnitude found for the pulse duration suggests the hypothesis that the photon emission implies the decay of a metastable level, by the same type of process which accounts for the emission of UV photons in pure argon excited with electron avalanche. This phenomenon was studied extensively by L. COLLI (7), who suggest the following mechanism to explain the emission: the metastable level $^3P_2$ produces by three-body collisions the formation of an excited argon molecule A_2^* according to

$$A(^3P_2) + A(^1S_0) + A(^1S_0) \rightarrow A_2^* + A(^1S_0) .$$

A three-body collision accounts for the conservation of momentum and energy.

An UV photon is emitted in the transition from the excited molecular

(7) L. COLLI: *Phys. Rev.*, **95**, 892 (1954).

state of argon to the fundamental state:

$$A_2^* \to A(^1S_0) + A(^1S_0) + h\nu \,.$$

The emission intensity I versus time was experimentally found well represented by the formula

$$I = C\left(\exp\left[-\frac{t}{\tau_A}\right] - \exp\left[-\frac{t}{\tau_M}\right]\right),$$

were, under the considered scheme, τ_A is the time constant for the formation of the molecules A_2^* and τ_M is the lifetime of the molecular state.

The experimental observations of this process determined that:

$$\tau_A \simeq \frac{1}{10p^2}\,\mathrm{s}\,, \qquad (p\text{: pressure in } \mathrm{mm_{Hg}}),$$

and

$$\tau_M \simeq 3.5\ \mu\mathrm{s}\,.$$

We note that in our case, too, the decay time of photon emission is independent of the pressure, and that our results are in good quantitative agreement with the measurements reported above.

This enhances the hypothesis that in pure argon excited with high energy α-particles, the photon emission is, at least for the most part, due to a process of the same nature as that observed in the case of excitation produced by electron avalanche.

7. – Measurements with Mixtures of Argon and N_2, O_2, CO_2.

It was expected that small amounts of molecules of foreign gases in argon, would produce quenching collisions on the excited argon atoms or molecules, which would result in a reduction of the intensity and of the duration of the light emitted by argon.

The energies of excited argon atoms and molecules may in some cases be stored by the foreign molecules during the quenching collisions, and be partly emitted as photons of different energies.

These hypotheses account, qualitatively, for the results obtained in the following measurements with mixtures of pure argon with N_2, O_2, CO_2:

The concentrations of N_2, O_2, CO_2 were determined by pressure measurements. Due to the little accuracy of this method, the percentages shown in Table I, left column, are to be considered as informative. The total pressure of the gas was 80-90 $\mathrm{cm_{Hg}}$.

In Table I, left side, are given the pulse amplitudes and their relative values with respect to the amplitudes obtained with pure argon. At the right side are indicated the pulse rise times and decay times.

With percentages from 0.5 to 2.5% of N_2, a progressive reduction both in the amplitudes and in the rise and decay times was observed. For instance, by introducing 0.5% of N_2 in pure A, the pulse amplitudes diminished by about 50% and the decay time became ~ 0.5 μs (Fig. 4). The addition of a few ten thousandths of O_2 and CO_2 produced effects of the same type.

TABLE I. – *Amplitudes and durations of light pulses measured with the sodium salicilate UV converter.*

	PULSE AMPLITUDES		PULSE DURATIONS		
	Phototube EMI 6262 Supply voltage 1070 Volt Amplification gain ~ 470 Time constant $RC \sim 200\,\mu s$		Rise time of electric circuit $\sim 5 \cdot 10^{-8}$ s		
Gas composition	Pulse amplitude Volt	Relative amplitude	Rise time $\tau_r \cdot 10^{-6}$ s	Decay time $\tau_d \cdot 10^{-6}$ s	Time constant $RC \cdot 10^{-6}$ s
A (99.99%)	50	100	0.15	2.5 ÷ 3	1; 0.2
A + O_2 [N_2] %					
0.5	25	50	0.15	0.5	1; 0.2
1	17	35	0.1	0.25	0.2; 0.1
2	15	30	0.07	0.15	0.1
A + O_2 [O_2] %					
0.05	17	35	0.05	0.1	0.1
0.15	12	25	0.05	0.1	0.1
0.4	7	15	0.05	0.1	0.1
A + CO_2 [CO_2] %					
0.03	15	30	0.05	0.1	0.1
0.07	10	20	0.05	0.1	0.1
0.15	7	15	—	—	—
A + N_2 + O_2 [N_2]% [O_2]%					
2 0.25	10	20	0.05	0.1	0.1
2 0.75	5	10	0.05	0.1	0.1
A + N_2 + O_2 [N_2]% [O_2]%					
2 0.05	10	20	0.05	0.1	0.1
2 0.15	8	15	0.05	0.1	0.1

A more accurate study of this effect was not possible because in most cases the rise and decay of the pulses under observation seemed limited by the rise time and time constant of the electronic circuit, i.e. $5 \cdot 10^{-8}$ and 10^{-7} s respectively.

Mixtures of three components were studied: the addition of a few thousandths of O_2 or a few ten thousandths of CO_2 to a mixture of $A_2 + 2\%\ N_2$ reduced critically the emission intensity.

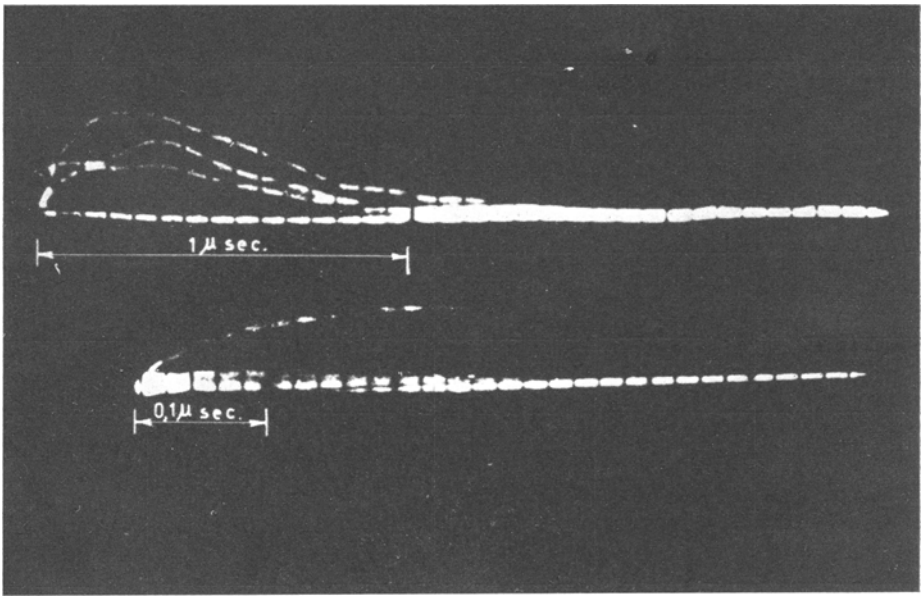

Fig. 4. – Shapes of α pulses in (A + 0.5% N_2), 80 cm_{Hg}. Rise time ~ 0.15 μs; decay time ~ 0.5 μs (differentiating time constant 0.2 μs).

8. – Components of Light Pulses in Visible Spectrum.

Successive measurements were carried out to study the emission of the photons in the visible spectrum with various mixtures of the same gases. For this purpose the sodium salicilate converter was removed.

The results of the measurements are the following: no visible light was detected with purified A, with A containing O_2 from 0.5 ‰ to 2.5 ‰ or CO_2 from 0.02 ‰ to 0.7 ‰.

On the other hand a visible light component was detected in mixtures of A containing N_2 from 0.35% to 2%. The pulse amplitudes obtained in these cases are shown in the following table:

TABLE II. – *Amplitudes of light pulses in the visible spectrum.*

Phototube EMI 6262 - Voltage supply 1070 V-Amplification gain ~ 470 - $RC \simeq 200$ μs.

A + N_2 mixtures [N_2] %	Visible light pulse amplitude Volt
0.35	~ 5
0.7	~ 7
1.2	~ 8
2	~ 4

Scintillation Chamber as α Spectrometer.

In this part the construction and the adjustment of a scintillation chamber, suitable for use as α spectrometer, is described.

The main characteristics of this instrument, namely, the amplitudes and durations of single α-pulses and the statistical distribution of the amplitudes of pulses obtained with monoenergetic α-particles, were studied.

An instrument of this kind has recently been built by G. BOICOURT and J. BROLLEY ([8]). To convert UV scintillation in noble gases they used a polystyrene film containing 5% of tetraphenyl-butadiene applied directly on the photomultiplier, following a method suggested by EGGLER and HUDDLESTON ([9]). Some results obtained by them with spectroscopical grade krypton are similar to those described here: a 20% resolution on the ^{234}U α peak, for instance. With argon, however, they obtained much lower pulse amplitudes.

In our experiments we have used tank argon of 99.9% purity without further purification, for the purpose of investigating the possibilities of the instrument in more simple operations.

9. – Experimental Apparatus.

Fig. 5 shows the system used to evacuate and outgas the scintillation chamber.

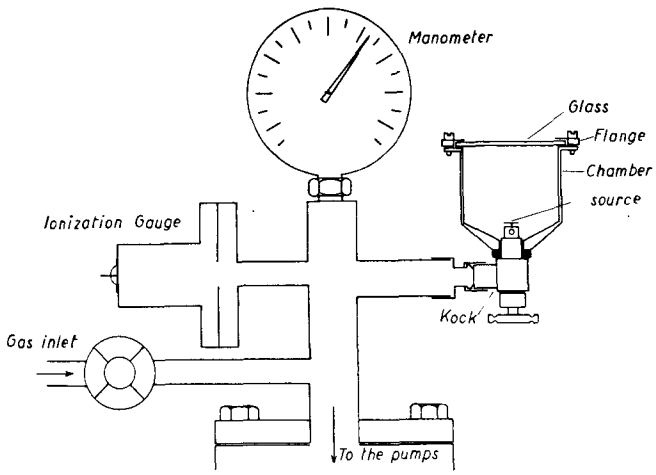

Fig. 5. – Vacuum system for the operation of the scintillation chamber.

([8]) G. BOICOURT and J. BROLLEY jr.: *Rev. Scient. Inst.*, **25**, 1218 (1954); see also UCRL-4536.

([9]) C. EGGLER and C. HUDDLESTON: *Phys. Rev.*, **96**, 100 (1954).

This chamber (Fig. 6) is cylindrical, 5 cm wide and 3.5 cm, terminating in a cone.

It is closed by a glass disc by means of a flange with a polyethylene gasket. Its walls are of silvered brass.

The chamber is provided with a cock at the bottom, so that it may be closed off and removed from the rest of the system.

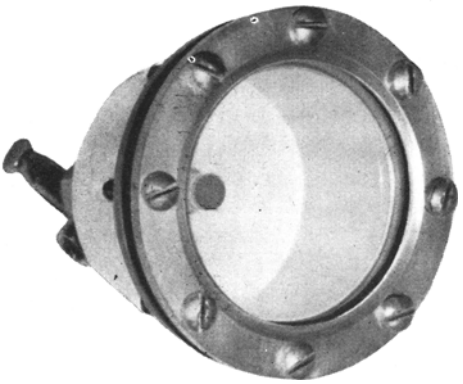

Fig. 6. – A view of the scintillation chamber: the sodium salicilate coating and the α source at the bottom can be seen.

The α-particle source is placed on a stainless steel disc of 5 mm diameter, attached to one end of the cock that is inserted in the lower part of the chamber.

The α source used is polonium.

The internal surface and geometry of the chamber were studied with view to uniformity and better collection of the light. For this purpose a different coating technique from that described in Sect. **3** was usefully employed.

An opaque and very white coating was obtained by spraying a saturated solution of sodium salicilate in methyl alcohol on the chamber walls that had been heated to over 100 °C. This sodium salicilate layer was used both as an UV converter and a light diffuser.

The light pulses produced in the chamber by α-particles were analyzed by a photomultiplier, type Dumont 6292, the photocathode of which was placed in optical contact with the chamber window.

The amplitude and duration of the pulses at the output of the photomultiplier were measured by means of the electric apparatus and methods described in Sect. **1**. Spectra of the amplitudes of the pulses, that were suitably shaped by a reflexion line, were obtained by means of a 99-channel pulse analyzer.

10. – Measurement Results.

Preliminary results indicated that the spread of the α-pulse amplitudes depended on the pressure of the filling argon. It should be noted that such spreading is partly due to a geometrical effect on the collection of light by the UV converter, that happens because the α-tracks, along which the UV photons are emitted, are not collimated. This effect can be reduced by confining the α-particle paths within a little space around the source, using a high gas pressure. On the other hand it was observed that, in our conditions, the increase in the pressure caused an appreciable reduction in the mean amplitude of the pulses. This effect can probably be attributed to the increased quenching of the excited argon molecules by impurities.

This results in a reduction of the mean number of the photoelectrons emitted

by the photocathode per each α-particle, i.e. in a worse statistical distribution of the pulse amplitudes of the output of the photomultiplier.

In our conditions, the convenient pressures were found to be 150-200 cm_{Hg}.

A spectrum of pulse amplitudes obtained in the above described manner is shown in Fig. 7. The distribution has a symmetrical shape, and its full width at half maximum is $\sim 13\%$.

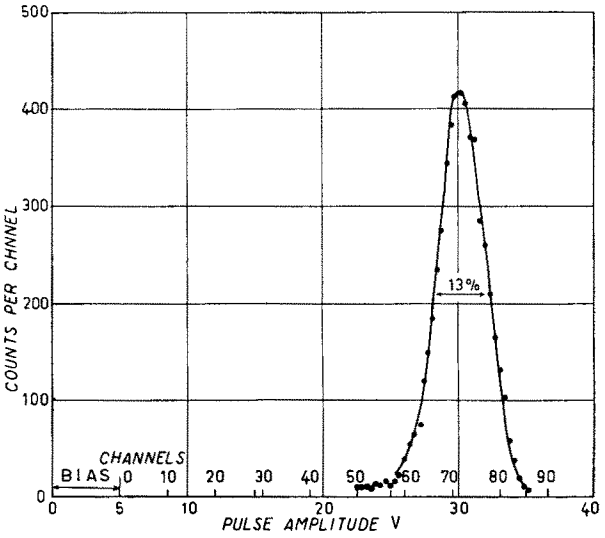

Fig. 7. – Pulse spectrum of α-particles from a thin Polonium source (pulse cut off at 2 μs).

In this case an estimate was made of the number of photoelectrons liberated at the cathode of the phototube during each scintillation: under the assumption that the statistical values of this number follow a Poisson distribution, the relative standard deviation of the pulses at the end of the multiplier, measured by us, implies that the average value is more than ~ 300.

A number of the same order has been found by comparing the α-pulses with the dark pulses produced by single electrons from the cathode.

This permits the assumption that above neglected causes for spread, such as geometrical effects on the light collection, are less effective than the purely statistical deviation.

In this case too, it was interesting to examine the shape of the individual pulses. The rise of the pulses appeared limited by the rise time of the electronic chain of $\sim 5 \cdot 10^{-8}$ s.

The duration of pulses were of some 10^{-7} s, much shorter than those of some μs observed with highly purified argon, as described in Sect. **4**.

11. – Testing Other UV Converters.

We have experimented with the same material used by BOICOURT and coll. as UV converter: a transparent film of polystyrene with 5% of tetraphenylbutadiene was applied to the silvered walls of our scintillation chamber.

Measurements with this converter gave results that are not very different from those obtained with the sodium salicilate converter, though the pulse amplitude obtained was about half.

It was observed that soon after the chamber had been filled, the pulses decreased appreciably.

This may indicate that such converters contaminate the argon ([8]).

12. – Possibilities of the Scintillation Chamber.

12·1. *Constancy of operation.* – The measurements obtained with the scintillation chamber having a converter of sodium salicilate were not always exactly reproducible in various fillings, probably because of uncontrollable quantities of impurities contained in the filling argon. However, once the chamber was filled its responsive characteristics remained constant for many days.

In particular, pulse spectra obtained over a period of a few days were in excellent agreement.

12·2. *Counting rate.* – The principle on which the gaseous scintillation counter is based and the sufficiently high statistics in the generation of each pulse make it permissible to suppose that this instrument furnishes a counting rate approaching 100% ([9]), at least at not-too-low energies.

12·3. *Energy and time resolution.* – In order to compare this instrument with those of current use, it may be observed that the scintillation chamber permits an energy resolution qualitatively comparable with that of crystal spectrometers ([10]), while it cannot compete with the ionization chamber when a refined analysis is required.

On the other hand the resolving time of the scintillation chamber is expected to be considerably shorter with respect to the ionization chamber and to inorganic scintillators [ZnS, NaI (Tl) etc.] owing to the fact that the pulses of the former instrument have a rise time not more than some 10^{-8} s, versus some 10^{-6} s of the ionization chamber and ZnS, and some 10^{-7} s of the NaI(Tl).

We can conclude that the scintillation chamber may be used, with simple and easy technique, as a counter and as a spectrometer for α-particles (or, in a like manner, for other heavy particles). The characteristics of its response may offer advantages in some types of work, particularly where a good time resolution is required.

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We are very indebted to prof. G. BOLLA for his interest and to prof. U. FACCHINI for his useful suggestions and stimulations.

Thanks are due to Dr. B. DE MICHELIS for his help in some measurements.

([10]) A definitely better resolution can be obtained with some kind of crystals cooled at low temperature (liquid nitrogen) though this requires careful technique and less simple operation: see B. HAHN: *Helv. Phys. Acta*, **26**, 271 (1953).

RIASSUNTO

Si descrivono metodi per rivelare ed analizzare gli impulsi di luce prodotti dal passaggio di singole particelle α nei gas. Nella Parte I si studia l'andamento temporale di impulsi ottenuti con argon di varie purezze ed i risultati si mettono in relazione con il meccanismo di produzione dei fotoni. Nella Parte II, si tratta dell'utilizzazione di questi impulsi per la realizzazione di strumenti rivelatori di particelle α. Si descrive una camera a scintillazione funzionante con argon, che consente l'analisi spettrale di particelle α con una risoluzione che è dell'ordine del 13% a ~ 5 MeV, ed offre un tempo risolutivo particolarmente breve.