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THE INTERNAL AND EXTERNAL BREMSSTRAHLUNG

ASSOCIATED WITH β -DECAY IN TRITIUM

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Administratie

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Period of work : august - november 1960

Notebooks : none

A. Introduction

By "Bremsstrahlung" is meant the continuous quantum radiation arising when electrons undergo braking in the fields of atomic nuclei. Discovered by Röntgen as long ago as 1895, this type of radiation occurs when the β -electrons from a disintegrating β -emitter are braked in the surrounding matter, being termed "external bremsstrahlung" in this case. But in such isotopes the β -electron is also accompanied by a type of radiation that is independent of the surrounding matter. This type is called "internal bremsstrahlung", because it is generated by the electron inside the radioactive atom, during the decay process. It was first demonstrated by Aston ¹⁾ in the "twenties" when, in the course of γ -ray measurements on RaE, he detected a soft, inhomogeneous form of γ -radiation.

Since this discovery, the bremsstrahlung accompanying β -decay has been investigated in other isotopes that disintegrate in this manner, preference being given to those which emit little or no contaminating γ -radiation of the homogeneous type. As a rule the investigator reports

the proportion of the kinetic energy of the β -particles that is emitted in the form of internal bremsstrahlung. Some experimental results of this kind are collected together in Table I. The effect of external bremsstrahlung can rarely be obviated and it constitutes the biggest difficulty encountered in the quantitative measurement of internal bremsstrahlung. One way of eliminating it is to deflect the β -electrons away from the measuring equipment by means of a magnetic field -- a method worked out by Langevin-Joliot ²⁾. Another is based on the linear Z-dependence of the yield of external bremsstrahlung: the magnitude of the I.B.S. effect is arrived at by experimenting with absorbers having different Z values and then extrapolating to $Z = 0$. ³⁾. No experimental data have so far been published on I.B.S. in tritium.

Since, in practice, the E.B.S. can never be eliminated, it is a sensible course to express the amount of I.B.S. as a proportion of the amount of E.B.S. generated in a given material. The latter is roughly proportional to Z, and thus the ratio between internal and external bremsstrahlung, which can be calculated, differs from material to material. Other things being equal, then, the measured activity value, which is the sum of the two kinds, provides a relative measure of the quantity of I.B.S. present.

B. Theory

It can be shown that when a tritium atom undergoes radioactive decay, a helium nucleus of mass 3 and a electron are produced. Since the electrons emitted by tritium have a continuous energy spectrum, and since nuclei can only possess discrete energy values, a third particle must be assumed to play a part in the decay process; it is assumed in fact that the energy released

is distributed between the electron and a neutrino. Considerations based on the law of conservation of angular momentum also lend colour to the neutrino hypothesis. Even from the standpoint of classical electrodynamics, it is to be expected that quantum radiation will be generated when an electron is expelled from a disintegrating nucleus. The electron and the nucleus form a dipole, and the momentum of the dipole changes suddenly as a result of the departure of the β -particle. The energy lost by the system is manifested as electromagnetic radiation -- as I.B.S. Theoretical calculations relating to the I.B.S. spectra of allowed β -transitions were carried out by Knipp and Uhlenbeek ⁴⁾ and by Bloch ⁵⁾ and the relevant publications appeared at almost the same time, in 1936. Wang Chang and Falkoff ⁶⁾ extended these calculations to forbidden transitions, and Nillson introduced corrections in respect of the Z-dependence of I.B.S. Knipp and Uhlenbeek calculated, in two different ways, the chance of a single electron producing a bremsstrahlung quantum. First they took as basis a β -electron with a given energy; by calculating the chance that the electron will emit radiation, and integrating this over the electron energy spectrum, they arrived at a spectrum of bremsstrahlung intensities. Their second method made use of Fermi's theory of β -decay, which states that a neutron in the nucleus transforms spontaneously into a proton, emitting an electron and a neutrino. Quantum-mechanical considerations led them to introduce an electromagnetic radiation field into the Fermi theory. By calculating the chances of obtaining a neutrino, an electron and a γ quantum, and summing over the allowed states of electron and neutrino, Knipp and Uhlenbeek determined the chance of generating a radiation quantum of given energy. The conclusions they

arrived at thus were the same as those of Bloch⁵⁾, as the latter duly observed in a footnote to his paper. Bloch regarded the β -decay process as taking place in two stages. In the first stage the nucleus disintegrates, giving rise to a neutrino in state G' and an electron in state s' . In the second, the electron passes from the intermediate to a final state s , emitting quantum radiation. Likewise making use of the Fermi theory, Bloch went on to work out an expression for the total I.B.S. intensity:

$$I_{\text{I.B.S.}} = \frac{1}{2} C_1 (mc^2)^6 \phi(\Delta) \quad (1)$$

where C_1 is a constant, mc^2 (which has the usual meaning) is likewise constant, and $\phi(\Delta)$ is a function of Δ alone ($\Delta = 1 + \mathcal{E}$, where \mathcal{E} is the maximum kinetic energy of the electrons, expressed in mc^2 units).

According to the Fermi theory, the average kinetic energy \bar{E} of β -electrons

$$\bar{E}_{\text{kin}} = C_1 (mc^2)^6 \left(\frac{hc}{e^2} \right) \psi(\Delta) \quad (2)$$

where C_1 is the constant occurring in formula (1), and the symbols e , h , m and c have their usual meanings. Like $\phi(\Delta)$, $\psi(\Delta)$ is a function of Δ alone. The proportion of the electron energy that is released as I.B.S. during the β -decay process can now be found by dividing (2) into (1).

$$r(\Delta) = \frac{I_{\text{I.B.S.}}}{\bar{E}} = \frac{e^2}{hc^2} \frac{\phi(\Delta)}{\psi(\Delta)} = 5,81 \times 10^{-4} \chi(\Delta) \quad (3)$$

In the case $\mathcal{E} \ll 1$ (which is applicable to tritium), this expression simplifies to

$$r(\Delta) = 5,81 \times 10^{-4} \frac{32}{33} \mathcal{E} = 5,63 \times 10^{-4} \mathcal{E} \quad (4)$$

In the case $\xi \gg 1$,

$$\chi(\Delta) = \frac{16}{3} (\ln 2\Delta - \frac{11}{5}) \quad (5)$$

However, this last formula is of little interest to us because $\ln 2\Delta$ must exceed $11/5$ although $\Delta > 5$, a requirement that is only just satisfied by Y^{90} the isotope with the highest E_{\max} in Table I. Bloch arrived at a different value for $\chi(\Delta)$ when he took as basis the reasoning of Konopinski and Uhlenbeck ⁷⁾ about β decay. If $\xi \ll 1$, $\chi(\Delta)$ has the value $(32/45)\xi$; formula (4) then becomes:

$$r_1(\Delta) = 5,81 \times 10^{-4} \frac{32}{45} \xi = 4,13 \times 10^{-4} \xi \quad (6)$$

Thus the proportion of β -energy emitted as I.B.S. is dependent only on the maximum kinetic energy of the β -radiation and, in the case $\xi \ll 1$, it is proportional to ξ .

Wang Chang and Falkoff ⁶⁾ discovered, amongst other things, that the I.B.S. effect increases with increasing degree of prohibition. They observe, however, that the ratio between the total I.B.S. intensity and the total β -intensity is independent of the degree of prohibition of the β -transitions, being expressible as

$$I_{\gamma}/I_{\beta} = \frac{4\alpha}{3\pi} (\ln 2\Delta - 2,3) \quad (7)$$

where α is the fine structure constant, which has a value of $1/137$. This formula is equivalent to (5), which is therefore valid for all types of β -transitions.

Corrections introduced by Nilsson ⁸⁾ concerned the approximations -- $Z = 0$, and the assumption of non-relativistic energies -- which had been adopted by Bloch, Knipp and Uhlenbeck for the purpose of their calculations. Nilsson considered that these approximations could give rise to appreciable errors in I.B.S. calculations. It is in fact clear that discrepancies occur between theoretical and experimental results, particularly at low values of E_{\max} ; examples may be found in Table I, where results from experimental work and from theoretical calculations are displayed side by side for various isotopes. However, Michalowicz ⁹⁾ has pointed out that Nilsson's calculations still fail to provide an adequate explanation for the discrepancies between theory and experiment.

The distance travelled by β -electrons once they have left the disintegrating atom is determined by the manner in which they yield energy to the surroundings. Electrons of 1 MeV and below yield energy for the most part by ionizing the matter that is braking them. A small proportion of the energy of comparatively slow electrons is converted into quantum radiation (E.B.S.) as a result of their undergoing braking in the nuclear fields of the atoms they encounter. According to Heitler ¹⁰⁾ the average rate of energy loss per centimetre involved by E.B.S. emission is

$$\frac{dE}{dx}_{\text{E.B.S.}} = N r_0^2 \frac{Z^2}{137} (E + mc^2) \left[4 \ln \frac{2(E + mc^2)}{mc^2} - \frac{4}{3} \right] =$$

$$3,44 \times 10^{-4} (E + mc^2) \frac{Z^2}{A} \left[\ln \frac{2(E + mc^2)}{mc^2} - \frac{4}{3} \right]$$

where N is Loschmidt's number ($N = L\rho/A$, where L is Avogadro's number, ρ is density and A is atomic weight), Z is atomic number and $r_0 = 2.8 \times 10^{-13}$ cm, the classical electron radius. For low energies, (8) transforms into

$$\begin{aligned} \left(\frac{dE}{dx}\right)_{EBS} &= 3,44 \times 10^{-4} \frac{Z^2 \rho m c^2}{A} \left[4 \ln 2 - \frac{4}{3} \right] = \\ &= \frac{Z^2 \rho}{A} \cdot 0,255 \text{ [keV/cm]} \end{aligned} \quad (9)$$

Averaged over the whole energy spectrum of the β -electrons, the energy loss per electron in consequence of the generation of E.B.S. is

$$\frac{E_{E.B.S.}}{\bar{E}_{kin}} = \frac{Z^2 \rho}{A} \cdot 0,255 \int_0^{E_{max}} \frac{1}{F(E)} dE \quad (10)$$

where $F(E) = -(dE/dx)$, a quantity that has often been determined experimentally for a given range of energies. The use of formula (10) to calculate the E.B.S. effect of the β -electrons emitted by a given isotope would involve integrating over the β -energy spectrum of that isotope, and $F(E)$ is often a complex function. For that reason attempts have been made to find another relationship which would allow the energy loss to be calculated. It has been deduced from experimental data that, in an X-ray tube, the conversion efficiency

$$\frac{E_{E.B.S.}}{E_{kin}} = \frac{\text{X-ray energy}}{\text{cathode ray energy}} = 1.1 \times 10^{-6} ZV \quad (11)$$

(V in KeV)

This formula holds reasonably well throughout the 10 - 100 KeV energy range; moreover, a very similar formula,

differing only in regard to the numerical coefficient, which is 9.2×10^{-7} instead of 1.1×10^{-6} , can be arrived at theoretically for a tube with a solid anticathode.

Over the whole of its flight, then, an electron of energy E_{kin} emits a quantity of radiation $E_{\text{E.B.S.}}$ such that $E_{\text{E.B.S.}}/E_{\text{kin}} = C_2 Z E$, C_2 having a value of $1.1 \times 10^{-6} e^{-1}$ where e is the elementary electric charge, which is unity if the electron-volt unit is employed. Hence

$$E_{\text{E.B.S.}} = C_2 Z E^2 \quad (12)$$

Accordingly, to calculate the quantity of E.B.S. produced by the β -electrons from an isotope, equation (12) must be integrated over the whole of the continuous β -energy spectrum:

$$E_{\text{E.B.S.}} = C_2 Z \int_0^{E_{\text{max}}} J(E) E^2 dE \quad (13)$$

If we follow the example of Bloch when he was dealing with I.B.S., and express this quantity of E.B.S. as a proportion of the average kinetic energy \bar{E} of the β -electrons, then

$$\frac{E_{\text{E.B.S.}}}{\bar{E}} = \frac{C_2 Z \int_0^{E_{\text{max}}} J(E) E^2 dE}{\bar{E}} \quad (14)$$

Calculations relating to the E.B.S. generated by β -electrons of medium energies (up to about 2 MeV) have been carried out by Wyard¹³). He deduced that the shape of the curve $I = f(h\nu/E_{\text{max}})$ is the same for β -electrons from isotopes whose β -energy spectra are of the same shape. The function $I = f(h\nu/E_{\text{max}})$ is obtained by integrating

$$I(h\nu) = E_0 \left\{ 4 \left(1 - \frac{h\nu}{E_0} \right) - 3 \frac{h\nu}{E_0} \log \frac{E_0}{h\nu} \right\} \quad (15)$$

over the β -energy spectrum. Equation (15) is arrived at by integrating $I(h\nu) = 4 - 3 h\nu/E$ from E_0 to $h\nu$ (E_0 being the initial energy of the E.B.S.-emitting electron; its value may accordingly be anywhere between $h\nu$ and E_{\max}).

Utilising the expressions for I.B.S. and E.B.S. provided by formulae (4) and (15), we obtain the following for the ratio between them:

$$\frac{E_{\text{E.B.S.}}}{E_{\text{I.B.S.}}} = \frac{C_2 Z \int_0^{E_{\max}} J(E) E^2 dE}{5.63 \times 10^{-4} E \bar{E}} \quad (16)$$

Here, however, e is expressed in mc^2 units whereas E is in KeV; the necessary adjustment can be made by multiplying the denominator by $1/mc^2$. Accordingly,

$$\frac{E_{\text{E.B.S.}}}{E_{\text{I.B.S.}}} = \frac{C_2 Z \int_0^{E_{\max}} J(E) E^2 dE}{5.63 \times 10^{-4} E \bar{E} / mc^2} \quad (17)$$

For a given radio-isotope and an absorber with a given Z value, then, the ratio between E.B.S. and I.B.S. is constant; also, for a given radio-isotope and a series of absorbers, the ratio is directly proportional to Z .

C. Experimental methods

Of the methods available for measuring the E.B.S./I.B.S. ratio for tritium, as detailed in the introduction, we selected that which involves the use of absorbers with different atomic numbers and we adopted this as principle for two further, mutually independent procedures. The biggest experimental difficulties arise out of the fact that tritium is a gas, so that bremsstrahlung will inevitably be generated in the walls of any containing

vessel. We were able to overcome these difficulties thanks to a method, developed by one of us ¹⁴), whereby tritium is converted into a solid tritide (fig. 1). This was taken as basis for the preparation of a metal-tritium system. By using an electrically heated tungsten coil to evaporate metal in an inert (argon) atmosphere at a pressure of about 2 cm Hg, a very active metallic powder is obtained, capable of absorbing large quantities of tritium within a few seconds. Measured with the electron microscope, the average grain size is 200 \AA (see fig. 2). Crystal planes can be observed in some of the grains.

Our first method of bremsstrahlung measurement makes use of the fact that the metallic deposit, provided its thickness is not too small compared with the range of β -electrons from tritium, can be used for two purposes, as a tritium absorber and as a medium in which E.B.S. will be generated by the bombarding β -electrons. The latter process can be studied as a function of Z by carrying out several experiments in turn with metals having different Z values. This procedure is obviously subject to the limitation that only metals capable of absorbing a reasonably large quantity of tritium can be employed; in other words, at room temperature the dissociation pressure of the metal-tritium system must be of the order of 10^{-5} mm Hg or less. A good review of metal-hydrogen systems is to be found in a book by Smith ¹⁵); if the isotope effect be disregarded, what is said there may be taken as applicable to metal-tritium systems. According to Smith, the systems most suitable for the above purpose are solid exothermic ones in which the metal is Ti, Zr or Th for example, or systems involving alkali metals or certain alkaline earth metals. A special technique has been developed to allow the bremsstrahlung emitted by tritium to be measured from outside the vacuum equipment. By dint of special pre-

cautions all the evaporated metal (30 mg) can be collected on to a thin (50 μ) nickel foil; after the metal has been tritided the radiation it emits can be detected from outside the vacuum equipment (fig. 3). A vacuum-tight seal is obtained by brazing the foil into a Covar ring, which is then fused with the glass of the vacuum equipment. The heating coil is fitted with a reflector (also shown in fig. 3) which can likewise be heated to prevent the particles of evaporated metal from settling on places other than the foil. The measuring arrangement consists of a Geiger-Müller counter tube (type no. 18507) having a 2.5 mg/cm² mica window, plus the following counting equipment:-

1. H.t. unit with pre-amplifier (PW 4022)
2. Universal electronic counter (PW 4032)
3. Ratemeter (PW 4042)
4. Pre-selector (PW 4052).

The pumping circuit (fig. 4) includes a tritium replenisher A and a calibrated reservoir B with a capacity of about 300 cc. The tritium replenisher, the design of which is due to one of us.¹⁸), consists essentially of a finely divided Ti powder deposited on a heatable surface and subsequently allowed to absorb tritium. The tritium pressure in the vacuum system can accordingly be adjusted by varying the temperature of the Ti powder. After being isolated from the rest of the circuit by the closure of cocks 1 and 2, containers A and B can be connected by opening cock 3. At a tritium pressure of about 0.1 cm Hg, approximately 1 curie of tritium is present in container B. Hence, by closing cock 3 and opening cock 2 it is possible to transfer a reproducible quantity of tritium into the foil-ended tube containing the powdered metal. The pressure in container B can be

accurately measured with the aid of a thermal conductivity gauge and a stabilized voltage source (T_{21}). The gauge provides information on pressure changes due to tritium absorption down to about 10^{-2} mm Hg; for measuring pressures down to about 10^{-6} mm Hg an ionization gauge (I_2) is employed. At this stage of the experiment -- the tritium having been completely absorbed by the layer of powdered metal -- the amount of quantum radiation passing through the foil is measured in a reproducible geometry. On account of their low energy, β -electrons from the tritium cannot penetrate the nickel foil (their maximum mass range being about 0.7 mg/cm^2). The measured electromagnetic radiation thus consists of the internal bremsstrahlung emitted by the tritium and the external bremsstrahlung generated in the deposit of powdered metal. Now, β -electrons will inevitably escape from the metallic layer and generate bremsstrahlung outside it, and to obtain a correction for this effect a separate experiment has to be carried out. A gas that is inert with respect to the metal-tritium system -- a noble gas satisfies this requirement -- is admitted into the foil-ended tube; the escaping β -electrons are braked in this gas, E.B.S. being emitted. Since the E.B.S. is approximately proportional to the atomic number of the gas, the exactness with which the correction can be found will be all the greater according as Z is high. This means that xenon ($Z = 54$) is the most suitable; however, krypton ($Z = 36$) can also be used. By filling the tube with argon, krypton and xenon in turn, for example, and studying the effect as a function of Z , it is possible to calculate the E.B.S. effect of the electrons escaping from the other side of the layer of powdered metal -- into the nickel foil ($Z = 28$) in other words. In the experimental arrangement used by us the noble gas is admitted into the vacuum equipment from reservoirs C, D or E. A mercury manometer F allows

the pressure of the gas to be accurately measured; insofar at least as xenon and krypton are concerned, the pressure can be adjusted by surrounding condensation traps G and H with liquid oxygen.

A second method of measuring the ratio $E_{E.B.S.}/E_{I.B.S.}$ has been tried out. Experimentally, it is somewhat simpler than the preceding one. It again makes use of tritium combined with a powdered metal, the only purpose of which is to anchor an adequate quantity of tritium to a given place. The method exploits the fact that the grain size of the powder (about 200 Å) is negligibly small compared with the average range in the metal of β -electrons emitted by tritium (for Ti the mass range is 1.6×10^{-4} g/cm², corresponding to a linear range of 0.3 μ). A standard quantity of powdered metal is applied as a film one grain thick to discs made of materials having different Z values (see fig. 5) so that a controlled variation in the E.B.S. is obtained; on extrapolation to $Z = 0$ this curve supplies the $E_{E.B.S.}/E_{I.B.S.}$ ratio. Certain precautions are necessary in regard to measurement of the activity of the discs: particular care must be devoted to ensuring that their geometry with respect to the G.M. counter tube remains constant (see fig. 5) and that exactly the same quantity of tritided metal powder is applied to each disc. Titanium is the metal used in powdered form. As before, it is evaporated with the aid of a tungsten heater coil in an argon atmosphere at a pressure of 2 cm Hg, but the powder is deposited on the wall of a glass balloon of diameter 6 cm; the tungsten heater is located at the centre of this balloon (fig. 1) ¹⁴). After the powder has absorbed tritium the balloon is cut off from the vacuum equipment and the contents are ultrasonically vibrated with a suitable liquid. In this way a stable suspension is obtained.

The choice of suspending medium will depend on the manner in which the suspension dries. The suspension must be applied to the disc in a quantity such that, after drying, the disc will be uniformly and completely covered by a film that is one metal particle thick. Particular attention has to be paid to the tendency of the suspended particles to coagulate around the edge of the disc. Results of our trials with various suspensions are reported on in one of the sections that follow. A certain amount of binder (E1160 nitrocellulose) is added to the suspension to prevent flocculation. Before any measurement of the activity of the discs can be undertaken, the discs must be freed from this binder by heating in vacuo to about 240°C. Up to this temperature the binder breaks down and evaporates without any appreciable dissociation of the titanium-tritium system occurring. The disc-baking arrangement may be seen in fig. 6. The activity of the discs is subsequently measured in H₂. By comparing the results with the activity of the discs in air it is possible to ascertain whether the E.B.S. effect due to the braking of β -electrons in the air between the disc and the counter tube is of any importance or not. The measuring arrangement comprises the same units as in the first series of experiments -- an end-window G.M. counter tube of type 18507 together with counting equipment.

D. Experiments: description and results

The experiments with the foil-ended tube were done first because they entailed most work. First the tube and the rest of the glassware were degassed at 450°C in vacuo. The evaporator was also degassed by passing a current of about 10 A through both the evaporator and heater coil. After argon at about 0.5 cm Hg had been admitted an attempt was made to evaporate the

Ti, but a short-circuit occurred between the evaporator terminal and the reflecting hood. In a second experiment the titanium was successfully evaporated, but it failed to take up the tritium. Probably the powder had been contaminated by gases that continued to leak out of the reflector and the heater coil during the evaporation process -- a consequence of the fact that degassing had necessarily been done at a much lower temperature, as otherwise it would have involved premature evaporation of the metal. A reasonably satisfactory solution was found to this problem by degassing in argon at 10 cm Hg. It was possible then to degass at a temperature high enough for good degassing, because the argon atmosphere of relative high density presents the evaporation of the titanium. Ti powder obtained after degassing in this manner and succeeding evaporation in a 0.5 cm Hg argon atmosphere did in fact prove to be sufficiently active. During the evaporation process the foil was cooled with a water sprinkler because, if its temperature had become excessive, the deposit of powder would have started to sinter, so losing its good absorptive properties. In the event, after 12 minutes the titanium powder had taken up practically all the tritium gas admitted to the foil-ended tube from the replenisher, in the manner described above (see Table II for the pressure readings). The evaporator coil was heated once again (with a current of about 7 A) in order to free it of tritium that had been absorbed by titanium residues. Initially, the count rate increased slightly when this was done, but afterwards it settled down to a steady value. The evacuation of the foil-ended tube brought about no change in the count rate; this steady value, corrected for the null effect, is shown in Table III. Subsequently experiments were performed with various noble gases in order to obtain corrections in respect of electrons escaping from the metallic layer; the results, again after subtraction of the null effect,

may be found in Table III. Several additional measurements of activity in the absence of noble gas were carried out in the course of these experiments, in order to ascertain whether any tritium might be leaking out of the powdered metal; it appeared that very little tritium was escaping in this way.

The titanium experiments were followed by experiments with other metals -- Be ($Z = 4$), Zr ($Z = 40$), Hf ($Z = 72$) and Th ($Z = 90$). Less favourable results were obtained in this phase of the investigations.

Beryllium (m.p. 1280°C) proved difficult to evaporate. The evaporator coil burned out before any evaporation had taken place when a current of 36 A was passed through it. Probably the reason for the failure was that the beryllium had alloyed itself with the tungsten of the coil. The experiment will have to be repeated using a Ta coil.

The coil also burned out (at about 25 A) in the zirconium (m.p. 1860°C) experiment. In a repeat experiment some zirconium evaporated, but the powder failed to absorb any tritium.

In both the hafnium (m.p. 1700°C) and the thorium (m.p. 1800°C) experiments, a little of the metal evaporated but powder formation came to an untimely end when, on account of the considerable amount of heat developed, the reflector shifted and came into contact with the evaporator coil, causing a short-circuit. A slight increase in the activity exhibited by the foil in the thorium experiment provided evidence that at least a little of the Th had evaporated. It was intended, if the thorium experiment should be successful, to correct for the activity of the thorium powder itself by carrying out a separate activity measurement before tritium was admitted to the tube.

It is clear in view of these numerous failures that a good many improvements still have to be made to the technique of this method of measuring the E.B.S./I.B.S. ratio. No further account will be given of this experimental method as our investigations aimed at improving the technique do not fall within the scope of this report.

The second method, using a titanium-tritium suspension applied to discs, was employed in experiments on eight different disc materials:

carbon (graphite) : $Z = 6$;

a silver-copper alloy with a mean atomic number of 40.7;

silver : $Z = 47$;

three silver-gold alloys with the following gold contents and atomic numbers:

25% Au : $Z = 55$;

55% Au : $Z = 63$;

75% Au : $Z = 71$;

and finally,

pure gold : $Z = 79$.

The first experiments were done with a suspension obtained in the following way. 6 mg of Ti were evaporated and 1 curie of tritium was absorbed into the resulting powdered deposit; this was then vibrated ultrasonically with 7.5 cc. of a 1:1 mixture of ethyl acetate and acetone to which 1% of E1160 nitrocellulose had been added. This produced a stable suspension. The biggest difficulty lay in obtaining a homogeneous film on the discs. One thing that soon became clear was that a rough (sand-blasted) surface was best; with a smooth surface, the powder in the suspension had a tendency to collect in

the middle of the disc. The suspension was dropped on to the discs with the aid of a small pipette with one graduation mark, fitted with a plunger. The pipette was used to transfer 30 mm³ of the above suspension on to the middle of the disc, which previously had been moistened with butyl acetate plus 1% of E1160 nitrocellulose; the whole was then allowed to dry in air, care being taken to keep the discs in a horizontal position. The result was not satisfactory, however; the discs were not evenly covered and some of the powder coagulated. Our first step was to improve the method of transferring the suspension. It was decided to increase the amount pipetted to 300 mm³ since, the greater the quantity of suspension transferred, the greater would be the accuracy with which the disc could be dosed. For this purpose, besides a new pipette of cubic capacity 300 mm³, a new suspension was required. This was prepared by evaporating 20 mg of titanium, absorbing 3 curie of tritium into it, and suspending the powder in 7.5 cc. of a 1:1 mixture of acetone and ethyl acetate plus 1% of E1160 nitrocellulose. The original suspension was diluted with 10 volumes of butyl acetate containing a 1% concentration of E1160 nitrocellulose. 300 mm³ of this dilute suspension was pipetted on to each of a small set of discs. The evenness with which the substance distributed itself was by no means ideal, but we were able at least to use these discs for tests in which the binder was baked out. In several cases the pressure rose very quickly at 200°C, a consequence of the fact that the heat was travelling upwards from the foot of the rack on which the discs had been placed. Thus the critical temperature of approximately 200°C was being attained first by the discs at the bottom of the rack and then by the higher ones. Differences in activity of $\pm 4\%$ were exhibited by individual discs, depending on the place occupied in the rack, though this would not be a serious objection in

experiments on a large number of discs. However, because an even distribution of powder over the discs had not been achieved when the original suspension had been diluted with butyl acetate, this diluent was abandoned in favour of the acetone and ethyl acetate mixture containing 1% nitrocellulose that had been employed earlier on. The distribution of the powder was now fairly even but, after baking, a good deal of carbon remained on the discs, a consequence of the relatively high concentration of nitrocellulose in the dilute suspension. This was undesirable, as carbon atoms generate E.B.S. at a constant rate and this radiation is indistinguishable from the I.B.S. emitted by tritium. A new suspension was accordingly prepared, consisting of 20 mg of Ti and 4 curies of tritium in 22.5 cc. of a 1:1 mixture of ethyl acetate with 1% of added E1160 nitrocellulose, and this was diluted with 10 volumes of pure butyl acetate; the amount of binder in the 300 mm³ pipette filling was therefore 10 times less than before. Thus 5.3 millicuries of tritium and 2.7×10^{-2} mg of titanium were now being transferred on to each disc. The results were only mediocre: a good deal of powder coagulated around the edges of the discs, at the places which dried first. When the original suspension was diluted with a pure mixture of acetone and ethyl acetate exactly the opposite occurred, the greatest quantity of powder collecting in the last place to dry, that being the middle of the disc. When a diluent was used consisting of 9 parts of acetone and ethyl acetate mixture with 4.5 parts of butyl acetate, coagulation occurred over the whole surface of the disc while it was drying. An even film was at last obtained by diluting the original suspension with pure acetone and pipetting 300 mm³ of the dilute suspension on to the discs, which were then allowed to dry; this was the procedure finally adopted. With each pipette filling, 5.3 millicuries of tritium and 2.7×10^{-2} mg of titanium were now being

transferred to the discs. A further difficulty was encountered in connection with the cleaning of the used discs. Neither brushing under butyl acetate nor ultrasonic vibration proved to be really effective. It was found that the only practicable course was to bake out the tritium in vacuo, remove the carbon by annealing in air and to get rid of the titanium by sand-blasting; after subsequent degassing the discs could be used again. This treatment was not of course given to the graphite discs, for which annealing was neither possible nor necessary. The results of a first series of experiments, in which all available discs were used, are displayed in Table IV. The temperature gradient in the rack during the baking process was responsible for a certain amount of inaccuracy, and for this reason a new holder was designed, consisting of a copper rod with cross-cut slots with which the discs were fitted. The results of a set of experiments in which the discs were baked in the new holder are displayed beside the results of the first series in Table IV. A fresh suspension was also used in the second series, made by vibrating 15 mg of evaporated titanium containing 1 curie of tritium in 11.75 cc. of a 1:1 mixture of acetone and ethyl acetate to which 1% of E1160 nitrocellulose had been added. As in the first series, this suspension was diluted with 10 volumes of acetone, and 300 mm³ of the dilute suspension was pipetted on to each of the discs. The results are displayed graphically in fig. 7. Each activity measurement comprised the following individual observations.

1. Observation of the activity of a standard preparation, to provide corrections in respect of any deviations in the apparatus; no such corrections proved to be necessary.
2. Three measurements of null effect over 10 counts.

3. Five observations of the activity of the disc, each lasting about 15 to 25 sec.
4. Same as 2.
5. Same as 1.

E. Calculations and discussion

As observed in section D, the experiments using the tube with the nickel foil brazed to its end (fig. 3) did not provide any data for deposited metals other than titanium. This means that the result in respect of titanium is of no significance for the time being. It is possible, however, to draw certain conclusions from Table III about the method itself.

1. While the noble gas is being admitted to and/or is present in the foil-ended tube, the titanium loses a little of the tritium which it has previously absorbed. This loss is probably due to absorption of impurities in the noble gas or in the apparatus by the titanium.
2. The correction for the β -electrons escaping from the powdered deposit is a necessary one. In xenon ($Z = 54$) at 5 cm Hg the effect of the E.B.S. generated therein is $(2770 - 2000)/2000$ or about 49%; on this basis, the effect for $Z = 22$ (titanium) is $(22/54)40$, or about 16% of the radiation generated in the absence of noble gas. Knowing the effect of the bremsstrahlung generated in the noble gas also puts one in a position to correct for the bremsstrahlung generated in the Ni foil.

3. As the pressure of noble gas increases, the count rate rises in the first place, passes through a maximum and then falls off. A possible explanation is as follows. A plane can be imagined lying parallel to the foil at a distance equal to the average range of the electrons in the noble gas; this plane may be considered as the source of the radiation. Accordingly, the distance between plane and foil is inversely proportional to the density and hence to the pressure of the noble gas. However, the efficiency with which the radiation from the noble gas is measured improves with increasing magnitude of the solid angle subtended to the counter tube by the imaginary radiating plane, which of course is bounded by the wall of the foil-ended tube. This solid angle increases with increasing pressure because at higher pressures the imaginary plane is closer to the foil and hence to the counter tube. Thus the count rate increases as the pressure rises. However, at high pressures a given change in pressure implies a smaller displacement of the plane than an equal change at low pressures; the change in the count rate is also smaller at high pressures. The fall-off in count rate at "high" pressures of up to 1 atm. is probably due to tritium still present in the evaporator. The second heating referred to in section D does not drive off the tritium completely, possibly because it has distributed itself throughout the titanium residue. Bremsstrahlung from the tritium in the evaporator undergoes more absorption in the higher gas densities prevailing at higher pressures. If this explanation is correct, the fact that the effect of the tritium in the evaporator is difficult to correct for must deprive the experiments with the foil-ended tube of much of their value.

4. Differences in the magnitude of the effect for the different noble gases used are naturally due to the fact that the E.B.S. yield in a given gas is dependent on its atomic number.

The results of the coated disc experiments, as tabulated in Table IV and graphically displayed in fig. 7, conform fairly closely to the function

$$CR_Z = A + BZ - CZ^3 .$$

CR_Z is the average count rate found with a number of discs made of a material of atomic number Z . A , B and C are positive constants. A is the I.B.S. associated with tritium decay, BZ is the E.B.S. generated in the disc, and CZ^3 is the amount of this E.B.S. that is lost on account of absorption by the material of the disc itself. This interpretation of CZ^3 can be justified in the following way. The β -electrons penetrating the disc and emitting E.B.S. have a mean range that in approximation is inversely proportional to the atomic number Z of the material. Half of the bremsstrahlung generated is emitted in the direction of the surface through which the electrons have entered, but some of it is absorbed before reaching that surface. The loss on account of absorption is proportional to Z^4 . As a result, the measured E.B.S. will exhibit an absorption effect that is dependent on Z^3 . Graphical analysis has revealed that the departures from the straight line $CR_Z = A + BZ$ are a function of Z^3 . The ratio between the E.B.S. generated by β -electrons in titanium and the I.B.S. emitted by tritium can now be arrived at by multiplying the count rate at $Z = 22$ by the absorption effect $C(22)^3$ and dividing by the count rate at $Z = 0$. It is practically impossible to

draw a curve approximately satisfying $ZR_Z = A + BZ - CZ^3$ through the observed values plotted on the graph, and for this reason we cast about for another approach. The method of smallest squares naturally gives the best approximation but in the event another method was adopted which is equally satisfactory. This involves determining, first the absorption factor C, and then the count rate CR_{corr} corrected in respect of the absorption effect, both as function of Z. Because this corrected count rate is a linear function of Z ($CR_{\text{corr}} = CR_Z + CZ^3 = A + BZ$) a straight line can be drawn on the graph, lying as near as possible to the plots of the corrected values. The necessary calculations are shown stage by stage in Table V; the first two columns of figures are the same as those appearing in Table IV. It may be said that in first approximation the observed count rates for C and Ni were not influenced by E.B.S. absorption within the disc (C and Ni have fairly low Z values and absorption is proportional to Z^3). On this basis it is possible to find provisional values of A and B which, in their turn, can be used to calculate the absorption factor C for each material (columns 3 and 4 of Table V). By subsequently correcting the measured count rate in respect of the average absorption effect CZ^3 (Z_6) one obtains CR_{corr} as a function of Z. In fig. 8 a straight line $CR_{\text{corr}} = f(Z)$ has been drawn for both series of experiments. However, before the overall ratio between external and internal bremsstrahlung can be determined it is necessary to investigate the influence of the electrons emitted by the tritium in the direction of the counter tube. Less radiation will be generated in the air between the disc and the window of the counter tube because the thickness of this air layer is only about one-tenth of the mean range of electrons in air. This

has been confirmed by the finding that the count rate for a given disc was slightly lower in air than when the disc and the counter tube were placed in an H_2 atmosphere. The difference was due to the lower rate of absorption in H_2 of the bremsstrahlung coming from the disc. However, the β -electrons issuing from the disc are completely braked in the mica window of the G.M. counter tube, in which they generate E.B.S., the reason being that the maximum range of β -electrons emitted by tritium (the mass range is about 0.6 mg/cm^2) is smaller than the mass thickness of the mica window (2.5 to 3.5 mg/cm^2). Like the I.B.S. and the E.B.S. from the disc, only half of the E.B.S. effect in the mica window undergoes observation, and the quantity measured would therefore be equal to that from the disc if this were also made of mica, both effects being due to half the total number of disintegration electrons. Once $CR_{Z=0}$ and $CR_{Z=22}$ have been found from fig. 8 the required ratio, that between the E.B.S. generated in titanium and the I.B.S. generated by β -electrons from tritium, can be worked out from

$$\alpha = \frac{E_{EBS}}{E_{IBS}} = \frac{2(CR_{Z=22} - CR_{Z=0})}{CR_{Z=0} - \frac{\bar{Z}_{mica}}{22}(CR_{Z=22} - CR_{Z=0})} \quad (18)$$

\bar{Z}_{mica} is the mean atomic number of mica, and has a value of 9.44. Fig. 8 gives $CR_{Z=0}$ and $CR_{Z=22}$ values of 5780 and 4580 c/min respectively for experimental series I, and 1170 and 935 c/m respectively for experimental series II. Insertion of these values in formula (18) yields $\alpha_{exp} = 0.593$ for series I and $\alpha_{exp} = 0.564$ for series II.

Alternatively, a theoretical value can be found for the E.B.S./I.B.S. ratio from formula (17):

$$\alpha_{th} = \frac{E_{EBS}}{E_{IBS}} = \frac{C_2 Z \int_0^{E_{max}} J(E) E^2 dE}{5.63 \times 10^{-4} \epsilon E \frac{1}{mc^2}} \quad (17)$$

where $C_2 = 1.1 \times 10^{-6}$

$Z = 22$ for titanium, the standard absorber material chosen.

$\int_0^{E_{\max}} I(E) E^2 dE$: Graphical integration of the curve in fig. 9 yields a value of 46.0 for radiant intensity over the whole energy spectrum of the β -electrons emitted by tritium.

$\mathcal{E} = 18.0$ KeV for tritium

$\bar{E} = 5.5$ KeV for tritium

mc^2 is equivalent to 511 KeV.

On insertion of the above values, formula (17) yields a theoretical ratio of

$$\alpha_{th} = \frac{1.1 \times 10^{-6} \times 22 \times 46.0}{5.63 \times 10^{-4} \times 18.0 \times 5.5 \cdot 1/511} = 10.2,$$

which is 17.5 times greater than the ratio as determined experimentally.

In the method adopted by us, unwanted effects (such as the generation of E.B.S. in binder residues, titanium, impurities etc.) are responsible for a fixed contribution to the E.B.S. -- fixed inasmuch as it does not vary with Z -- which is observed and measured as I.B.S. because it cannot be distinguished from the I.B.S. The consequence is that α_{exp} will always be smaller than α_{th} . Now, the α_{exp} found by us is certainly smaller than α_{th} , but what is not clear is why the results of two wholly independent experiments should agree so closely if bad experimental conditions really make such a big difference.

On carrying out similar calculations for P-32 and Tl-204 (for which E_{\max} is 1701 and 765 KeV respectively) we find that the discrepancy between theory and experiment becomes wider with decreasing energy of the emitted electrons (Table VI). The greatest discrepancy is for tritium, whose β -electrons are of low energy. Now formula (14), from which the E.B.S. has been obtained, is actually the most suitable one for low energies, so the cause of the systematic errors must be sought in formula (4), from which the I.B.S. has been obtained; it is fairly accurate for high-energy β -transitions but not, perhaps, for β -emitters such as tritium.

If, on the assumption that this conclusion is correct, $E_{\text{E.B.S.}}$ in titanium is taken to be $2.0 \times 10^{-4} \bar{E}$, the experimental results yield in a value of 1.2×10^{-4} for the I.B.S. effect.

Possible explanations for differences between experimental and the theoretically predicted I.B.S. values were reviewed by Langevin-Joliot²⁾. In his paper certain explanations (e.g. the effect of the magnetic moment of the neutrino) were rejected out of hand. He did however think that auto-ionization might be playing a part: there is a certain chance that an atom resulting from disintegration may be left in an excited or ionized rather than the ground state. According to Levinger¹⁶⁾ the chance of a K-shell electron being thus involved is $p_k = 0.90/Z^2$. For helium atoms, then, $p_k = 0.23$; accordingly, the effect may be on a considerable scale. However, the radiation generated when a helium atom returns to the ground state has a maximum energy of about 25 eV (the K-absorption edge for He occurs at 504 Å)¹⁷⁾ and because of the low energy of the X-radiation that is generated, the auto-ionization effect will not be detected by the method of measurement employed by us. A further possible source of error lies in the approxi-

mation on which formula (4) is based. In order to arrive at this formula for I.B.S., Bloch⁵) assumed that the Coulomb field exercised a negligible influence on the generation of radiation in the decaying atom. The assumption that $Z=0$ may in some cases have to be corrected for, but there is no need to do so in the case of tritium because here the assumption is reasonably close to the reality. As already concluded, the discrepancy between theory and experiment must be due to formula (4) being invalid for β -decay in tritium.

The results of our "coated disc" experiments to determine I.B.S. in tritium still have to be checked by another, independent experimental method -- possibly by the method using the foil-ended tube, as described above. Even so, it may already be assumed with reasonable certainty that tritium, on account of the low energy of the β -radiation it emits, does not behave in exact conformity with the theory.

Eindhoven, 4th August 1962

Philips Research Laboratories
N.V. Philips' Gloeilampenfabrieken

FMS

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O. Mifflin
Gratig

TABLE I
Values of I_{γ}/I_{β} in mc^2

Isotope	Z	E _{max}	Exptl. results	Theoret. results	
Y90	39	2180	2.2×10^{-3}	2.1×10^{-3}	Langevin-Joliot a
P32	15	1701	2.32×10^{-3}	2.38×10^{-3}	
			1.7×10^{-3}	1.55×10^{-3}	
			5.3×10^{-3}	2.3×10^{-3}	
Y91	39	1537	1.9×10^{-3}	1.97×10^{-3}	Langevin-Joliot a
Bi ²¹⁰	83	1170	0.84×10^{-3}	1.11×10^{-3}	
(RaE)			0.4×10^{-3}	0.42×10^{-3}	
Pr143	59	930	1.3×10^{-3}	2.3×10^{-3}	
Tl204	81	765	3.7×10^{-4}		Ricci b
Pm147	61	225	8.6×10^{-5}	2.9×10^{-5}	Most of the results shown here originally appeared in tabular form in an article by Michalowicz, Ann.Phys. <u>2</u> , 139 (1957). Other results have been drawn from (a) Langevin-Joliot, Ann. Physique <u>2</u> , 16 (1957) and (b) Ricci, Il Niova Cimente <u>8</u> , 1, 1-16 (1958).
S ³⁵	16	167	2.23×10^{-5}	1.7×10^{-5}	
			3.2×10^{-5}	1.7×10^{-5}	
H3	1	18	1.2×10^{-4}	1.98×10^{-5}	

TABLE II

Pressure changes in foil-ended tube after admission of
about 1 c of tritium

$t(\text{min})$	$U^{\text{th}}_{\text{SKt.}}$	$P(\text{mm Hg})$	P mm Hg
0	49.5	-	6.3×10^{-1}
0	68	-	2.7×10^{-1}
6	87.5	-	3.4×10^{-2}
9	88.5	3×10^{-2}	
12	89.5	1.5×10^{-2}	

TABLE III

Count rate in counts per minute

		1cmHg	2cmHg	5cmHg	10 cm Hg	20 cm Hg	60 cm Hg	
T_2 admitted	2080							Tube contained 30 mg Ti powder
Tube evacuated	2079							
X admitted		2461	2665	2770	2728	2672	2520	
Tube evacuated	2005							
Kr admitted			2384	(4.6) 2569	(9.4) 2469	(22) 2562	(57.2) 2444	
Tube evacuated	1986							
18 hrs. later	1965							
A admitted		1982	1949	1945	1970	1951	1914	
Tube evacuated	1935							

TABLE IV

Material	Count rates in:							
	Experimental series I				Experimental series II			
	Before baking	After baking	Error %	No. of discs	Before baking	After baking	Error %	No. of discs
C (graphite)	5434	4859	4.3	5	963	995	2.0	6
Ni	6462	6199	4.6	4	1168	1237	2.1	4
Ag-Cu. alloy	7064	6925	2.6	3	1249	1322	3.5	2
Ag	7335	7176	3.8	3	1353	1438	1.8	3
Ag-Au alloy 75:25 w/w	7307	6463	9	3	1331	1391	1.8	3
Ag-Au alloy 50:50 w/w	7183	6892	6.8	3	1361	1348	1.6	3
Ag-Au alloy 25:75 w/w	7178	6901	1.6	3	1397	1432	2.5	3
Au	6866	6696	4.3	3	1340	1405	1.7	3

Activity values, in counts per minute, of titanium-tritium suspension on discs with different atomic numbers, before and after baking.

In series I the dose of suspension on each disc contained:

Tritium Ti
5.3 mc 2.7×10^{-2} mg

In series II -do-

2.6 mc 3.8×10^{-2} mg

TABLE V

Steps in calculations undertaken to determine a linear
function $A + BZ = CR_Z$
(Data from disc experiments)

Experimental series I

Material	CR_Z	$-CZ^3$	Z^3	$-C \cdot 10^3$	$-\bar{C}Z^3$	CR_{corr}	Z
C	4859	0	216	0	1	4860	6
Ni	6199	0	10.640	0	48	6247	28
Ag-Cu	6925	49	67.100	0.73	302	7227	40.7
Ag	7176	178	104.000	1.71	467	7643	47
Ag-Au _{25%}	6463	1381	166.000	8.33	746	7179	55
Ag-Au _{50%}	6892	1442	262.000	5.51	1178	8070	63
Ag-Au _{75%}	6901	1918	358.000	5.36	1609	8510	71
Au	6696	2608	493.000	5.30	2215	8911	79
				----- $-\bar{C}=4.49 \times 10^{-3}$			

Experimental series II

Material	CR_Z	$-CZ^3$	Z^3	$-C \cdot 10^4$	$-\bar{C}Z^3$	CR_{corr}	Z
C	995	0	216	0	1	996	6
Ni	1237	0	10.640	0	8	1245	28
Ag-Cu	1322	55	67.100	8.20	52	1374	40.7
Ag	1438	7	104.000	0.672			47
Ag-Au _{25%}	1391	143	166.000	8.62	127	1518	55
Ag-Au _{50%}	1348	274	262.000	10.45			63
Ag-Au _{75%}	1432	278	358.000	7.77	275	1707	71
Au	1405	393	493.000	7.97	378	1783	79
				----- $-\bar{C}=7.68 \times 10^{-4}$			

TABLE VI

Isotope	E.B.S. in Ti mc^2/\bar{E}	I.B.S.	α_{th}	α_{exp}
P 32	1.22×10^{-2}	1.87×10^{-3}	6.5	6.6 - 6.8 ¹⁾
Tl 204	0.56×10^{-2}	0.84×10^{-3}	6.7	2.8 ²⁾
T 3	2.0×10^{-4}	1.98×10^{-5}	10.2	0.58

¹⁾ α_{exp} for P-32 has been worked out from data in H.G. Mahl, Verwendung der Bremsst. zur Untersuchung der Verteilung reiner β -Strahler im menschlichen Organismus, Strahleninstitut der Freien Universität Berlin, and in K. Siegbahn, β and γ ray spectroscopy, p. 649 et seq.

²⁾ α_{exp} for Tl-204 is given by Ricci, Il Nuovo Cimento 8, 1, 1-16 (1958).

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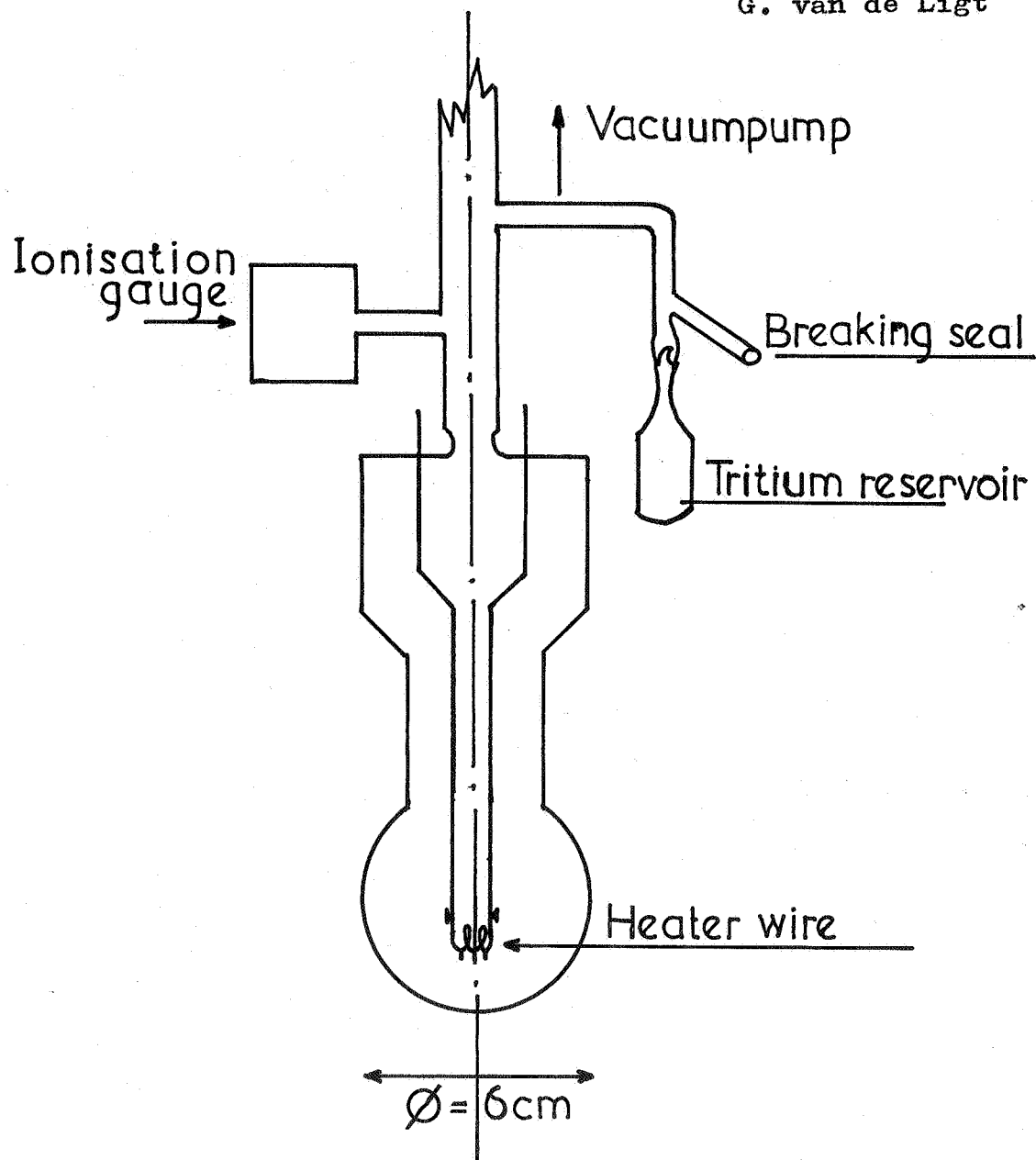
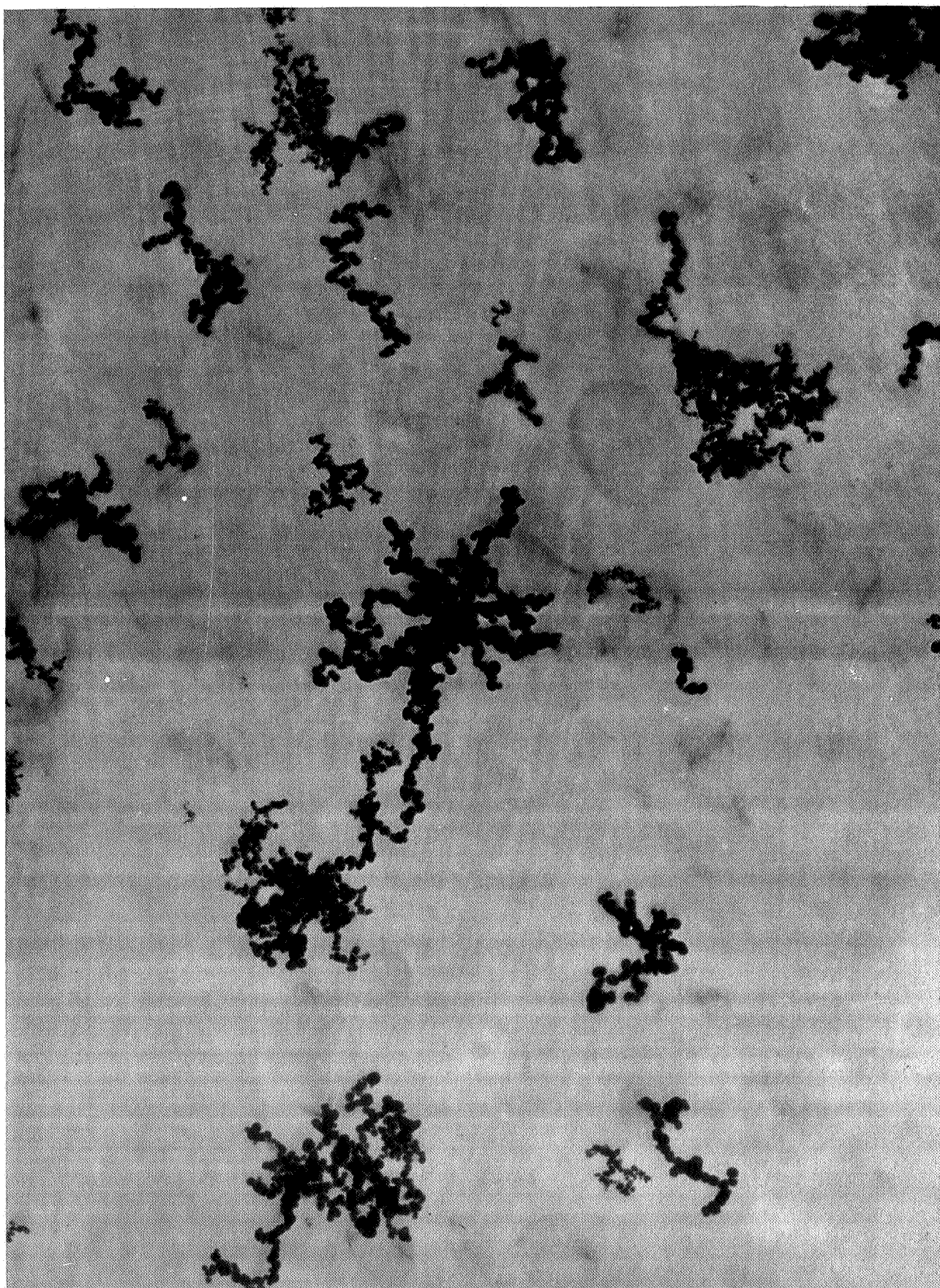


Fig. 1

Experimental arrangement for the preparation
of titanium powder

Fig. 2 Electron-micrograph showing titanium powder prepared by evaporation in argon. Argon pressure 2.5 cm Hg, magnification 50000 x. (0.2μ)



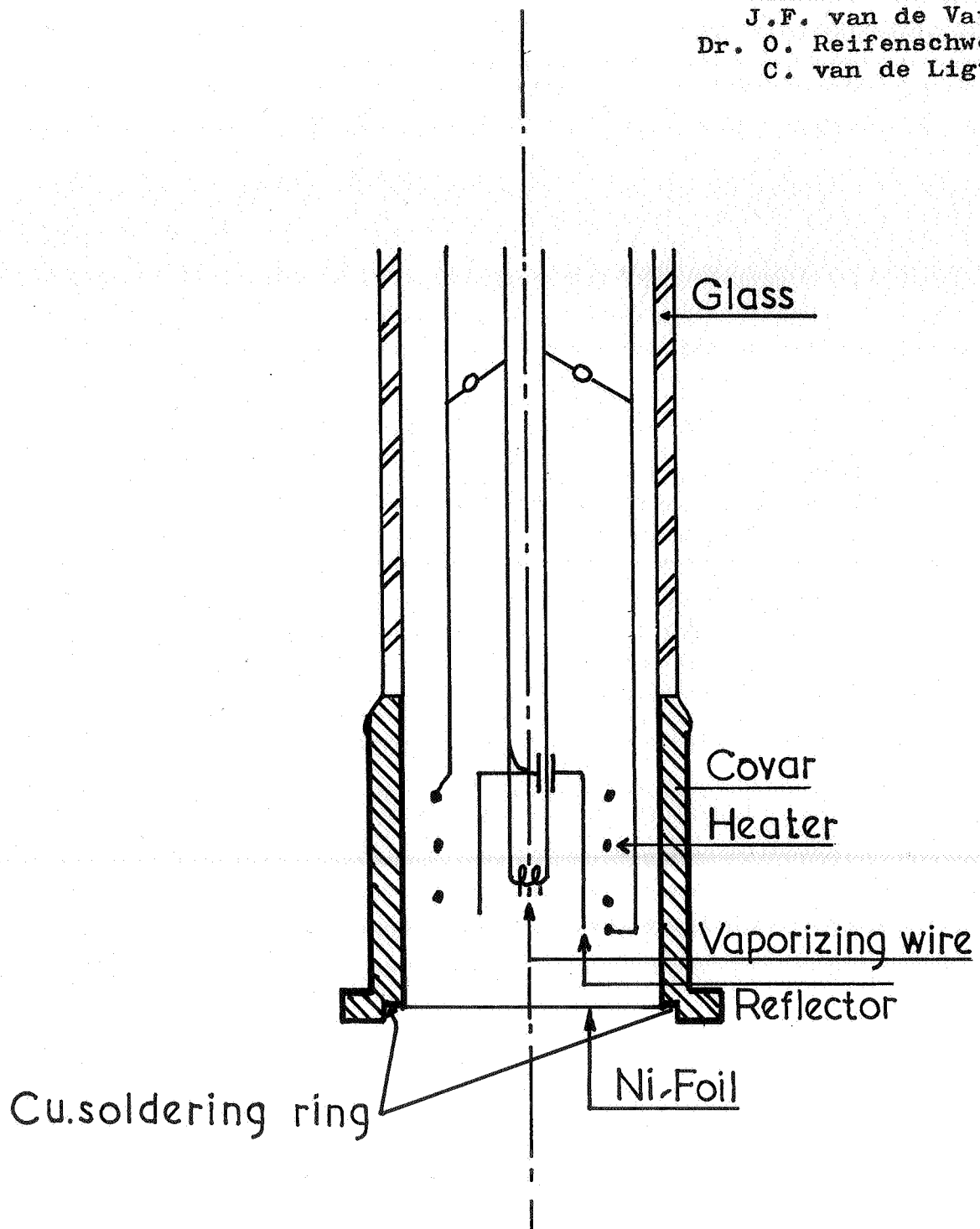


Fig. 3

Foil-ended tube for measuring the intensity
of bremsstrahlung

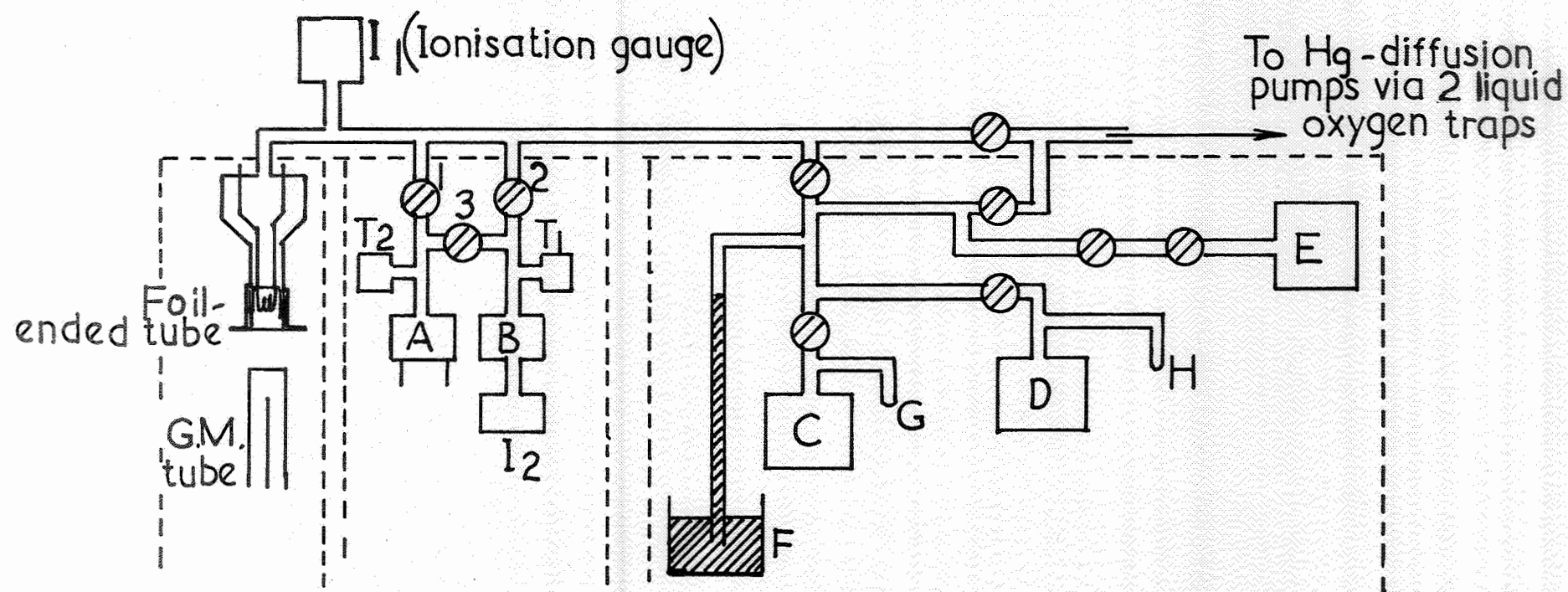


Fig. 4

Vacuum system

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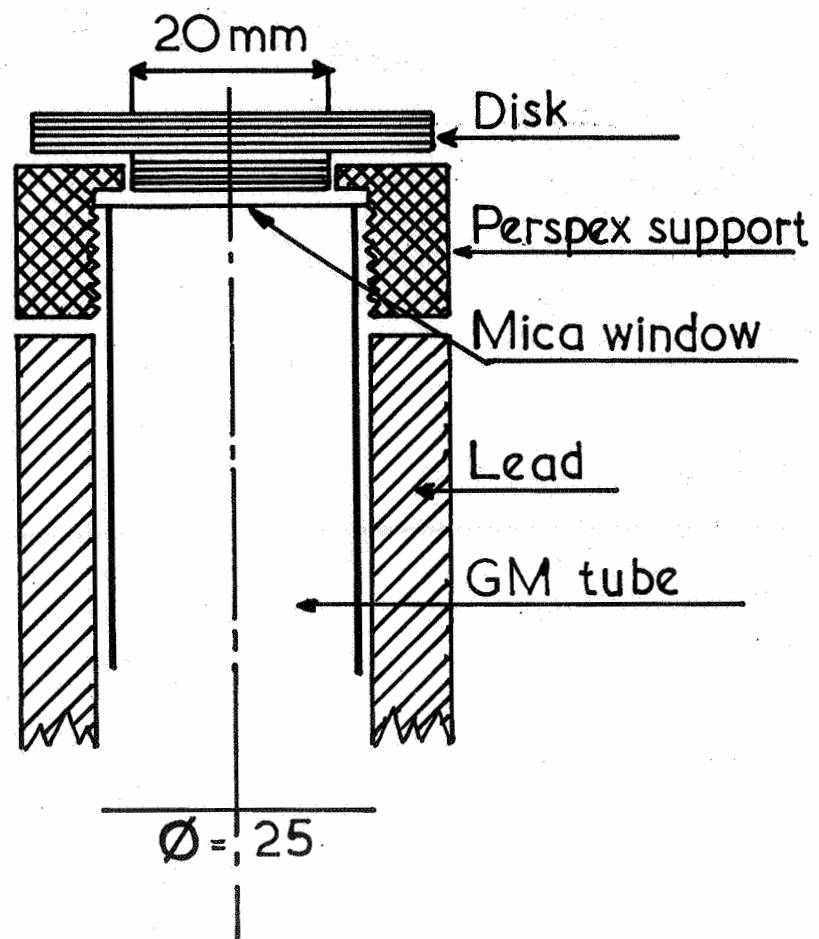


Fig. 5

Experimental arrangement for measuring
the intensity of bremsstrahlung from the disks

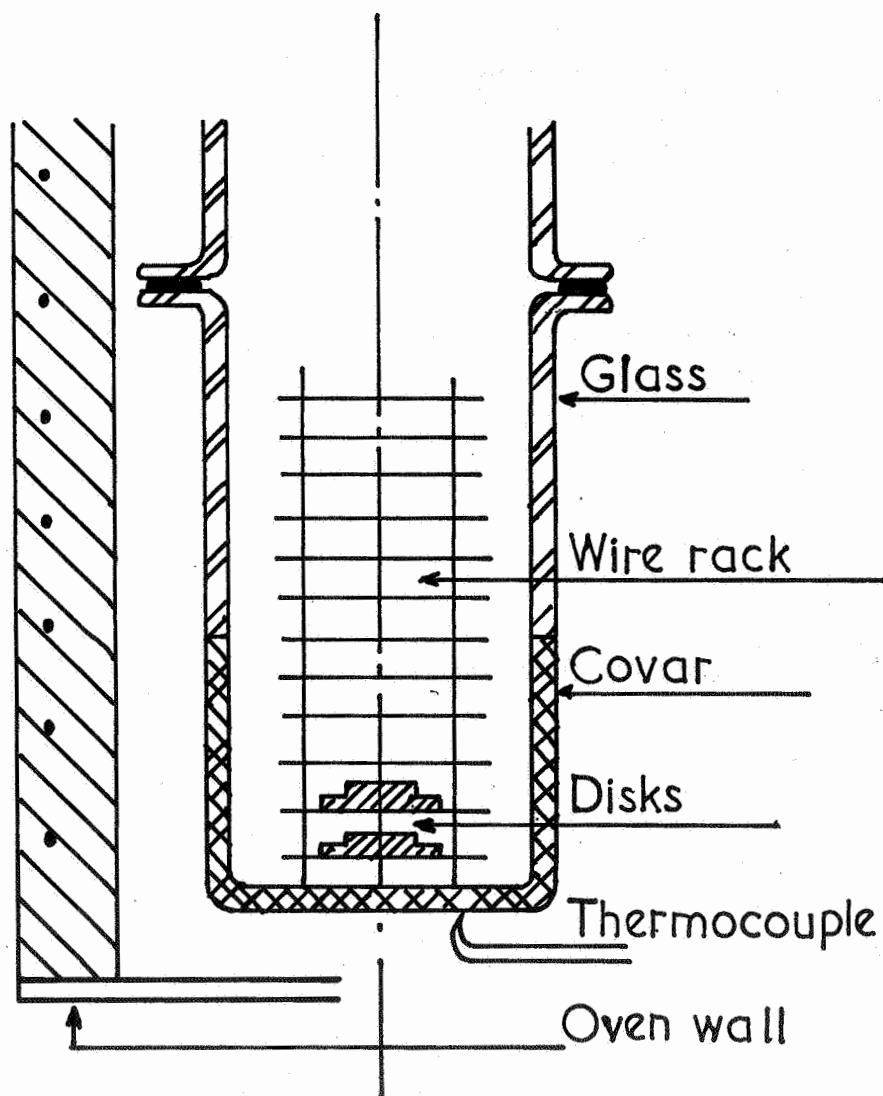


Fig. 6

Experimental arrangement for baking
the disks

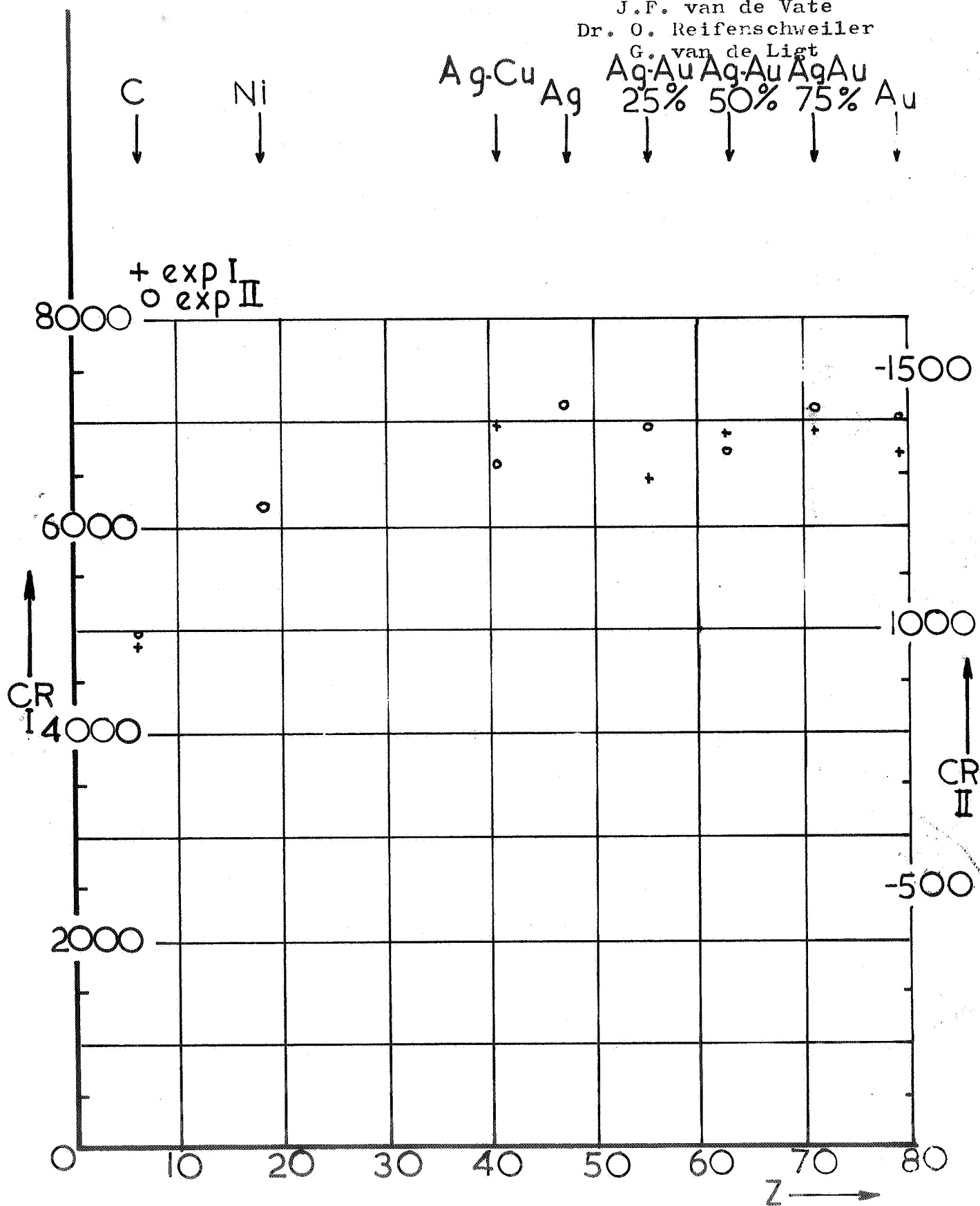


Fig. 7

Count rate as a function of the atomic number of the absorbing element

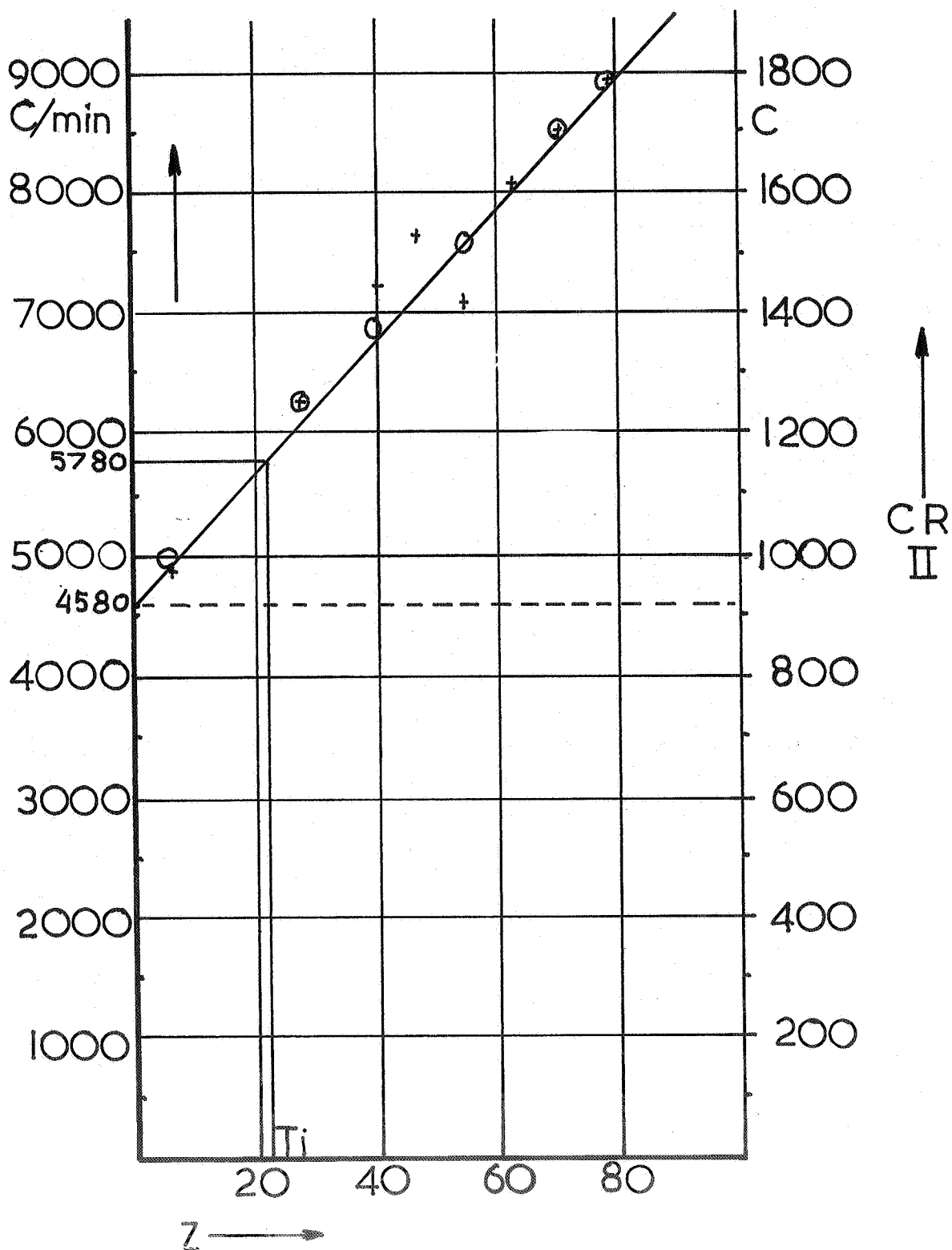


Fig. 8

The same function as displayed in fig.7 (count rate V. atomic number) after correction in respect of bremsstrahlung absorbtion in the disks

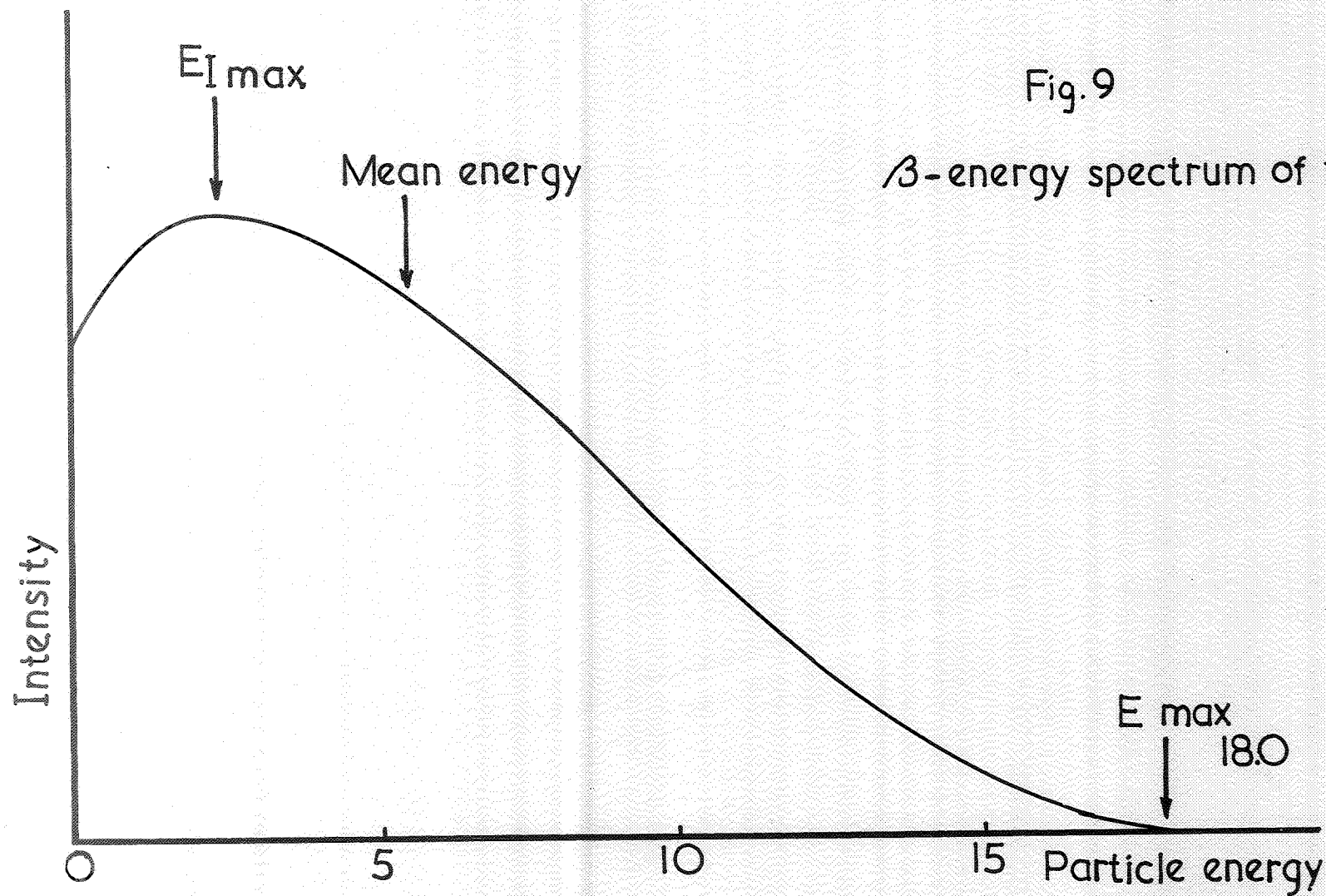


Fig. 9
 β -energy spectrum of tritium

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