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On the Doppler Broadening of Two-Photon Positron Annihilation Radiation (*) (**).

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Summary. — Doppler broadenings of positron annihilation radiation have been measured with a Li-drifted Ge-detector having a resolution of 1.76 keV FWHM for the 482 keV γ-line of ^{181}Hf. From the observed shapes of the photopeaks of the annihilation radiation in various metals, the functional form of the momentum distribution of the annihilating pair has been obtained. The least-square analysis of the experimental curves gives values for the FWHM of the unfolded pair momentum distribution curves as 2.7 keV for Cu, and 2.5 keV for Zn. These widths are consistent with angular correlation measurements on the annihilation quanta. Results obtained in the electric-field quenching in gaseous O_2 are presented. A double-subtraction method is presented and its utility is discussed in determining the Doppler broadening of the radiation originating from the positron annihilation with bound atomic electrons. A search for the one-photon ($E = (4/3)mc^2$) decay has been made. The upper limit for the relative cross-section of the one-photon to two-photon free annihilation in potassium metal is $\leqslant 7.0 \cdot 10^{-5}$. The relative merits of Doppler broadening and conventional techniques are compared.

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1. – Introduction.

One of the primary interests in the study of positron annihilation in solids has been to measure two-photon angular correlations. This type of measurement gives the momentum distribution of the center of mass of the annihilating electron-positron pair (1). Similarly, as a consequence of the center-of-mass motion, the energies of the annihilation photons, although infinitely sharp in the center-of-mass co-ordinate system, should be Doppler broadened in the laboratory system. The information obtained by the measurement of this broadening is similar, in principle, to the information obtained from the angular correlation. An early measurement of DuMond, Lind, and Watson in 1949 and of Lind and Hedgran in 1951 using a bent crystal spectrometer, showed that the effect was present but that it was difficult to observe (2).

In this paper we report a series of experiments to investigate the use of high-resolution lithium-drifted germanium (Ge-Li) solid-state detectors for the measurement of Doppler broadening and the results obtained with such techniques in the study of positron annihilation. These detectors, in addition to a good energy resolution, have a good time resolution capability which is also a potentially useful feature in the study of positron annihilation; the correlation of life-time and the momentum distribution can be measured with relative ease.

A preliminary summary of these results has been given earlier (3). Similar comparisons have also been made by other groups (4).

2. – Experimental procedure.

We have used lithium-drifted germanium (Ge-Li) solid-state detectors for this work (*). The detectors were planar drift types with typical volumes around 3 cm³.

The detector and the preamplifier were cooled to liquid-nitrogen temperatures in a cold-finger arrangement. The amplifier was a low-noise Tennelec

(1) S. De Benedetti, C. Cowan, W. Konneker and H. Primakoff: *Phys. Rev.*, **77**, 205 (1950).

(2) J. W. M. DuMond, D. A. Lind and B. B. Watson: *Phys. Rev.*, **75**, 1226 (1949); A. Hedgran and D. A. Lind: *Phys. Rev.*, **82**, 126 (1951).

(3) K. Rama Reddy, R. A. Carrigan jr., S. De Benedetti and R. B. Sutton: *Bull. Am. Phys. Soc.*, **12**, 74 (1967).

(4) H. P. Hotz, J. M. Mathiesen and J. P. Hurley: *Bull. Am. Phys. Soc.*, **12**, 74 (1967); *Phys. Rev.*, **170**, (1968); G. Murray: *Phys. Lett.*, **24** B, 26B (1967).

(*) These detectors were supplied by H. Mann of the Argonne National Laboratory counter development group to the Carnegie-Mellon University, Argonne National Laboratory joint muonic X-ray program.

Model TC-200. A Victoreen Model S-16 ACD-2 P (A) 3200 channel analog to digital converter was used to display the annihilation peak, typically, in sixty channels. A gain stabilizer was used in conjunction with radioactive sources to reduce the drifts in the pulse amplification and analysis system. With the use of gain stabilization, gamma-ray peaks were broadened by less than one half channel out of 3200 over long counting periods as compared to short counting times.

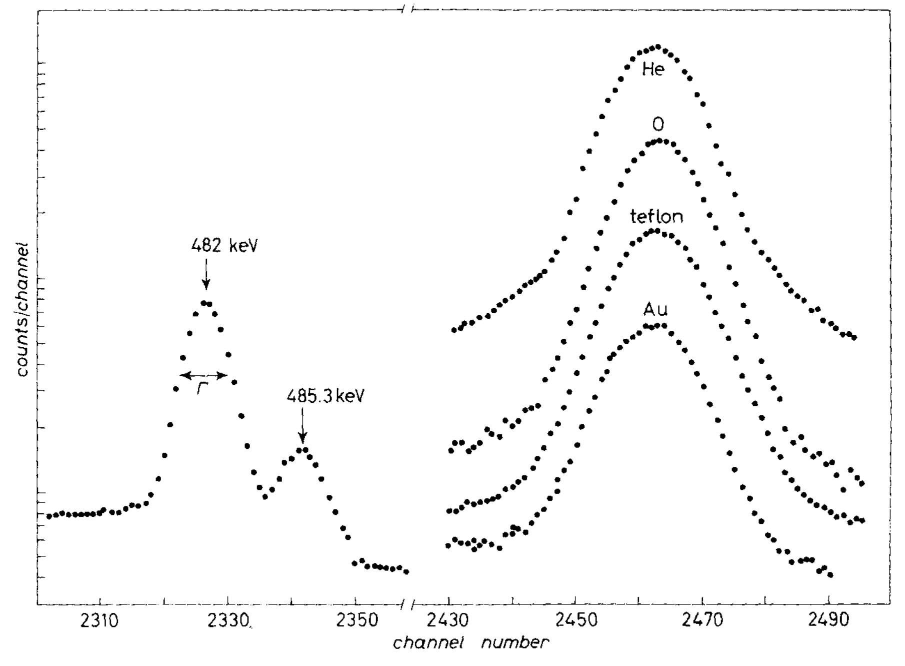

Fig. 1. – Energy spectrum of the annihilation radiation in various substances along with the 482 keV line of ^{181}Hf. $\Gamma = 1.76$ keV. 1 channel $= 0.217$ keV.

A carrier-free source of ^{22}Na of 50 microcuries was deposited on $\frac{1}{2}$ mm thick mylar and sandwiched between the two pieces of the material under study. The annihilation spectrum was measured as described previously. Typical instrumental resolutions in the experiments have been 1.76 keV FWHM for 482 keV ^{181}Hf gamma-rays and 1.84 keV FWHM for ^{137}Cs gamma-rays of 662.8 keV. In this energy region the line widths and line shapes do not change appreciably; thus the 482 keV gamma line is assumed to give the instrumental resolution of the annihilation experiments. Typical experimental spectra of the annihilation radiation in various materials and of the calibration line taken simultaneously are given in Fig. 1. Distinct changes are noticeable in the shapes of the annihilation peaks from one substance to another.

3. – Annihilation in metals.

In this Section we consider to what extent a measurement of Doppler broadening can be used in the study of the electronic structure of metals and how this compares with the angular-correlation method. The measurements of angular deviation θ from 180° between the two photons and of the fractional change in the photon energy $\Delta E/mc^2$ are related by a factor of two, *i.e.*, $\theta = 2\Delta E/mc^2$. The relation $\theta = 2\Delta E/mc^2$ is valid only when the measurements are made for the same component of the center-of-mass momentum, since the angle θ is related to the transverse center-of-mass momentum ($\theta = p_z/mc$) and the broadening ΔE is related to the longitudinal center-of-mass momentum. The center of gravity of the annihilation radiation also contains information on the average energy of the annihilating system which, in turn, is related to the effective masses involved. However, the expected shift in the center of gravity is of the order of 10 eV which is still below the limit of energy determination with Ge(Li) detectors.

TABLE I. – *Comparison of techniques.*

Feature	Conventional angular correlation technique	Doppler broadening technique
1) Basic measurement	Angular correlation	Line broadening
2) Resolution	Very good: $\frac{1}{2}$ mrad typical, 4 mrad $\simeq$ 1 keV	Not very good at present, best FWHM = 1.5 keV (equivalent to 6 mrad) but might improve
3) Electronic circuitry	High-speed coincidence circuitry	Linear pulse analysis system
4) Measured parameter	Transverse pair momentum	Longitudinal pair momentum
5) Use	*a*) Useful only for 2-photon decay *b*) Desired portion of the curve can be scanned at will	*a*) Useful for 2-photon, 3-photon and 1-photon decay *b*) Entire energy spectrum is obtained simultaneously
6) Sample constraints	Constraints on the sample are a problem: limits the angular resolution, counting rate, etc.	No problem in working with gases, plasma, high pressures, temperature, electric and magnetic fields, etc.

In Doppler broadening measurements the resolution is limited by the detector and associated electronics. The expected change in energy is $\Delta E = (v_F/2c)\,E_0 = 1.5$ keV for an electron with Fermi velocity $v_F = 1.8\cdot 10^8$ cm/s. Good resolutions are 1.5 keV corresponding to 6 mrad in angular correlation measurements. In angular correlation experiments the resolution is limited by the slit geometry and the physical dimensions of the source and sample. A typical value is $\frac{1}{2}$ mrad. Thus the resolution is more than a factor of ten better for angular correlation. However, in Doppler broadening measurements the instrumental resolution can be determined directly by observation of a gamma-ray line, while there is no such convenient calibration in angular correlation. Practical data acquisition rates appear to be more than an order of magnitude better for the Doppler broadening technique, using much weaker sources. As a result the Doppler broadening is particularly useful for studying the momentum distribution in the region of higher momenta. In this region the angular correlation measurements suffer because accidental counts become increasingly significant. In Table I the relative advantages of the two techniques are compared. In spite of its many advantages, the poor resolution of the present technique poses a serious problem for a quantitative interpretation of the experimental data. To overcome this difficulty, we have adopted the following procedure for unfolding the instrumental resolution.

The instrumental resolution line shape was assumed to be a sum of two Gaussians with the same position but differing in amplitudes and widths. The background in the region of the peak was assumed to follow a cubic polynomial. The assumed line shape is

$$(1) \qquad f(E, a_0 \ldots a_8) = \sum_{i=0}^{3} a_i E^i + a_4 \exp\left[-a_5[E-a_6]^2\right] + a_7 \exp\left[-a_8[E-a_6]^2\right] \ldots ,$$

where the a's are the fitted parameters to be determined from the experimental data. The values of these parameters are found by the method of least-squares analysis which minimize the function S^2 defined as.

$$(2) \qquad S^2 = \sum_{i=0}^{n} [F(E_i) - f(E_i, a_0 \ldots a_8)]^2\, W_i ,$$

where $F(E_i)$ is the measured quantity referring to the i-th data point with a statistical weight W_i and $\chi^2 = S^2$/number of degrees of freedom.

The true energy distribution in the Doppler broadening is assumed to be symmetric about the mean position, E_0, and to follow a functional form

$$N(E) = \sum_{i=1}^{4} [b_i + c_i(|E-E_0|)^i] ,$$

where it is assumed that contributions from each term $[b_i + c_i(|E - E_0|)^i]$ is positive definite. This form explicitly contains the truncated parabola expected on the basis of the free electron model. Using the analytic expression for the instrumental resolution a convolution function is defined by $g(E)$:

$$g(E) = \int N(E')\, f(E, E')\, \mathrm{d}E' \,.$$

The function $g(E)$ is compared with the experimental data, point by point, in the same manner as the resolution line shape.

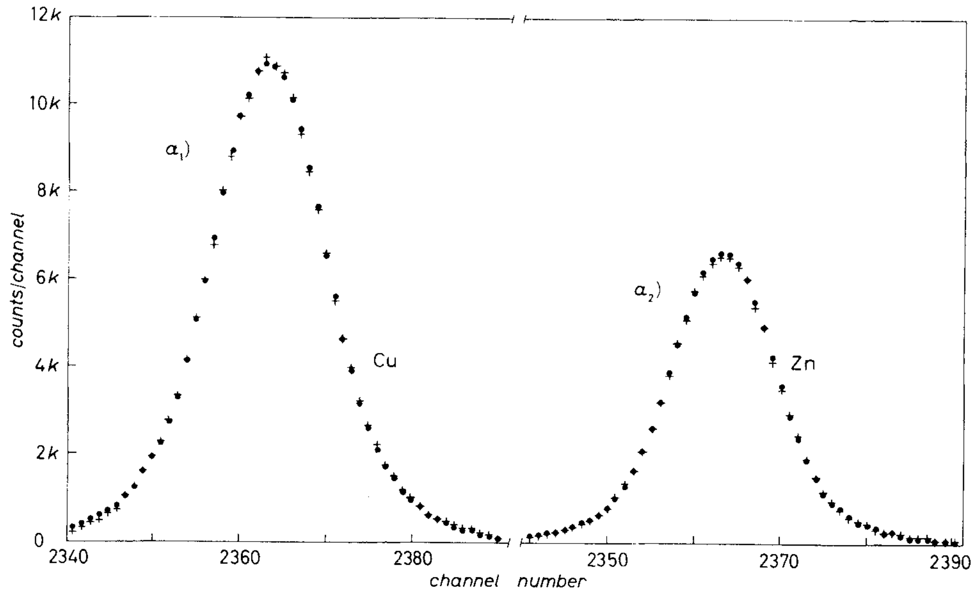

Fig. 2. – Energy distribution of the annihilation radiation, a_1) for polycrystalline copper, a_2) for polycrystalline zinc. + experimental data; • calculated « best-fit ». Calibration: 1 channel = 0.217 keV.

In Fig. 2 the continuous curves represent the calculated least-squares fit to the experimental data (denoted by circles). The abcissa is the energy and the ordinate is the counts per unit energy interval per unit time. The normalized χ^2 is greater than one, reflecting the fact that it is not a perfect fit. Similar χ^2 were obtained in fitting the calibration line, indicating that most of the difficulties in obtaining a good fit involve reproducing the calibration line. In this connection it should be noted that the lines contain ten to one hundred times as many counts as the lines employed in mesic X-ray spectroscopy and consequently the fitting problem is correspondingly more difficult.

Results obtained for momentum distributions after unfolding the instrumental resolution are given in Fig. 3. In the same Figure corresponding angular-correlation results (5) are given for comparison purposes and the agreement can be seen to be good with no systematic difference appearing as a function of angle. We have also used a Fourier-transform method (6) for unfolding the instrumental resolution. Error estimates were not made for the Fourier-transform method. However, the agreement between the two unfolding techniques is reasonable if not perfect.

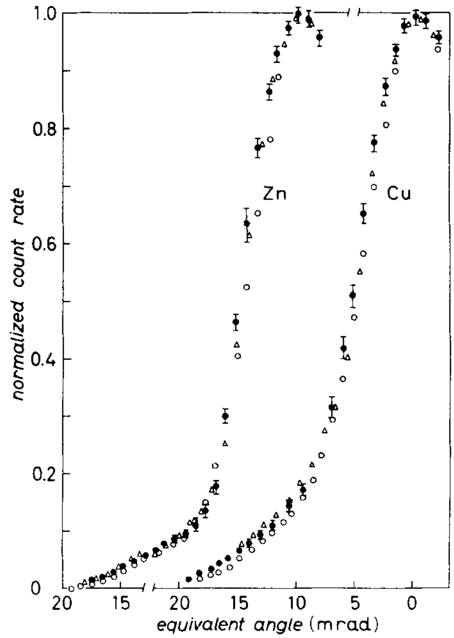

Fig. 3. – Comparison of the pair-momentum distribution spectrum obtained from the Doppler broadening technique and the angular correlation (5) technique; points (o) represent the result of the Fourier transform method (•) are from the polynominal method, and (▵), angular correlation result (5).

4. – Annihilation in gases.

There are two competing processes for positron annihilation in gases; formation of positronium which decays by two or three photons with negligible Doppler broadening (sharp component) and direct annihilation with the bound atomic electrons having a measurable Doppler broadening (broad component). A change in the relative rates of the two processes induced by external conditions will change the shape of the annihilation line and consequently the « moments of the line » will also change. Observations of such shifts offer direct evidence that the line broadening is properly interpreted.

Following Deutsch's (7) observation that an electric field can enhance the formation of positronium, there have been a number of attempts to study this phenomenon with varying degrees of success (8). In the presence of an electric

(5) A. T. Stewart: *Can. Journ. Phys.*, **35**, 168 (1957).

(6) P. M. Morse and H. Feshbach: *Methods of Theoretical Physics*, part I (New York, 1953), p. 465.

(7) M. Deutsch and S. C. Brown: *Phys. Rev.*, **85**, 1047 (1952).

(8) S. Marder, V. W. Hughes, C. S. Wu and W. Bennet: *Phys. Rev.*, **103**, 1258 (1956).

field the qualitative description of the phenomenon is that the positrons below the Ore gap gain sufficient energy to enter the Ore gap, where they have the possibility of forming positronium. Thus, the increase in the sharp component, in the presence of an electric field, should be equal to the decrease in the broad component, and the fractional transfer can be expected to be a monotonic function of the electric field. This effect has been studied by PAGE and BRIMHALL (9) in greater detail. In particular, they attempted to separate the sharp and broad components with an instrumental resolution of ~ 10 keV compared to the 1.7 keV now available.

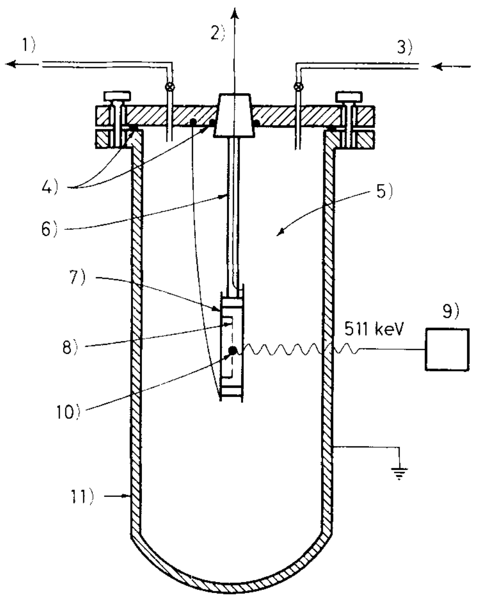

Fig. 4. – Experimental arrangement for the study of positron annihilation in gases. 1) Gas outlet; 2) high voltage; 3) gas inlet; 4) O ring; 5) gas under study; 6) plastic; 7) brass condenser plates; 8) $\frac{1}{2}$ mil Mylar; 9) detector; 10) source; 11) $\frac{1}{4}$ in. aluminium cylinder.

We have used oxygen gas in our experiments. The positron source, ^{22}Na, was deposited on a $\frac{1}{2}$ mm thick Mylar film and suspended rigidly between the polished rectangular brass plates forming a condenser. The assembly was enclosed in a pressure chamber with the potential across the condenser plates arranged so that it could be varied externally. The details of the experimental arrangement are illustrated in Fig. 4. The entire annihilation γ-ray spectrum, including the 3-photon radiation, was measured for each value of the electric field under identical conditions.

To obtain the transfer rates of the broad component to the sharp component and the Doppler broadening of the broad components, the data on oxygen was analysed by the following procedure:

a) Single subtraction method. The difference in count rate with electric field on and electric field off was obtained at each energy interval. The resulting « differences in count rate » *vs.* the channel number (energy) are given in Fig. 5 for various values of the electric field. From these curves the transfer of the broad to the sharp component as a function of electric field is clearly seen.

(9) J. E. BRIMHALL and L. A. PAGE: *Nuovo Cimento*, **43** B, 119 (1966).

The symmetry in the « difference data » reflects the stability of the electronic system used in these experiments. The area under the difference data, in the region of the annihilation peak, is almost zero at each field except at low field values (below $\simeq 3.0$ kV/cm), where there might be some uncertainty in the experimental result. This fact leads us to the conclusion that the fraction of positrons transferred from the broad component to the sharp component decays by two-photon radiation. This occurs because oxygen gas is paramagnetic, and spin-flip collisions at this pressure lead to fast conversion of triplet to singlet positronium which decays by the two-photon mode ([10]).

The percent transfer of broad component to the sharp component as a function of field is given in Fig. 6. This transfer rate at each field value was calculated by summing the absolute counts of the difference data and taking one half of the integral counts. Since the actual transfer rates are difficult to estimate from data taken with a detector having a complicated resolution function, the method used here gives a lower limit to the « fractional transfer ».

The annihilation spectrum obtained at an electric field of 15.5 kV/cm is still quite broad (FWHM = 2.32 keV as compared to the instrumental resolution of 1.76 keV), and the transfer of the total broad component to the sharp component is not complete although there is a tendency for saturation in the transfer at this field.

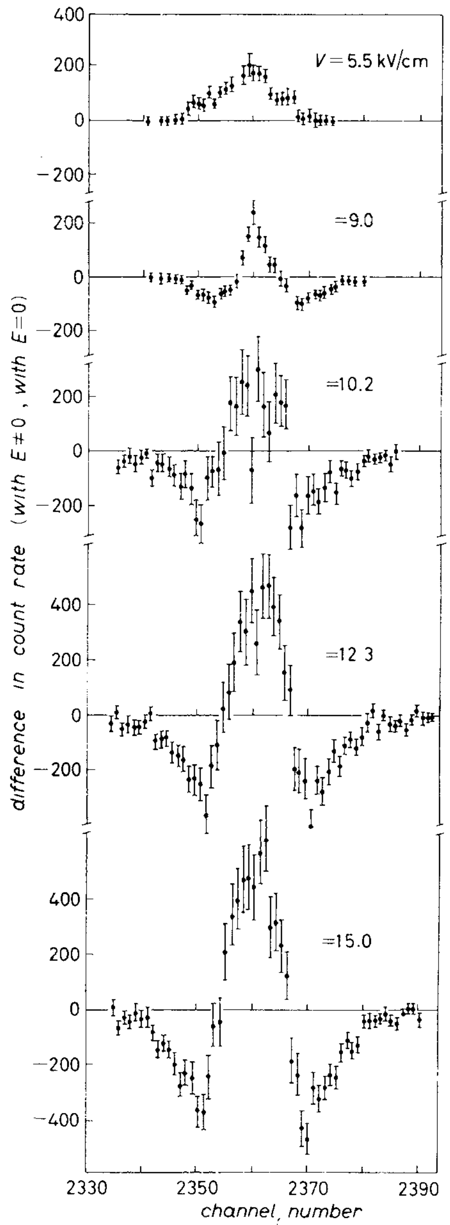

Fig. 5. – Curves illustrating the presence of the broad component in oxygen gas.

([10]) M. Deutsch: *Phys. Rev.*, **82**, 455 (1951).

b) A « double subtraction » method was used to estimate the width of the broad component. In this method the sharp component (the instrumental resolution here) was subtracted, channel by channel, from the « difference data » in such a way that the total resultant absolute counts were equal to the subtracted total counts. The resultant broad component spectrum is shown in Fig. 7. The root mean square width of Doppler broadening is estimated to be $\Gamma' \simeq 4.2$ keV. This width is twice as large (~ 17 mrad FWHM) as needed to account for the momentum distribution of the bound electron. The additional broadening indicates that the *positron may not be thermalized* in the gas before the annihilation takes place. This conclusion agrees with a similar conclusion based on the observation

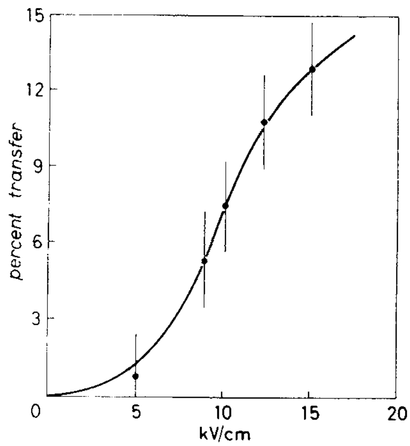

Fig. 6. – Electric field quenching in oxygen. Percent transfer (see the text) from the broad component to the sharp component as a function of d.c. electric field. $p \simeq 600$ p.s.i., $T \simeq 25$ °C.

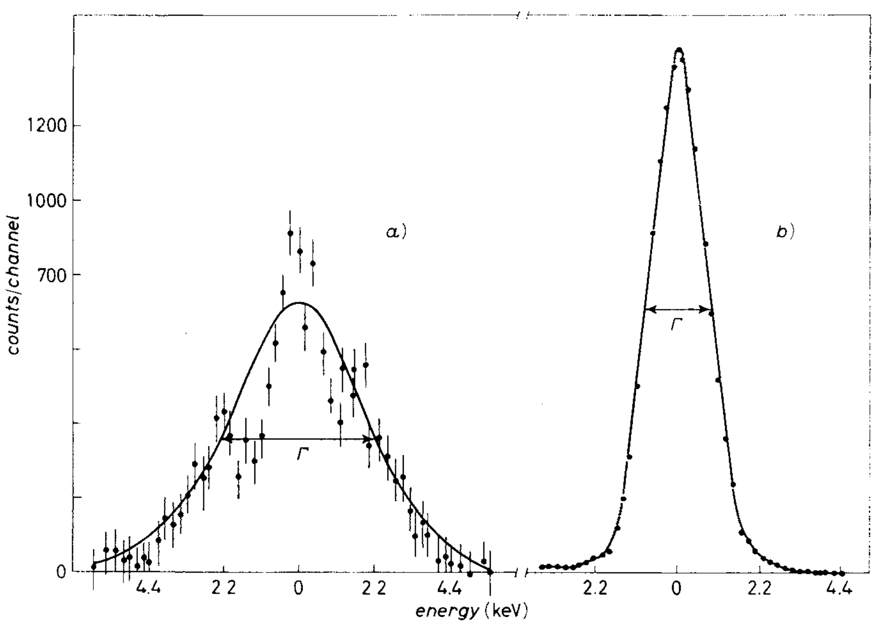

Fig. 7. – *a*) Doppler broadening in the broad components of oxygen using the « double subtraction » technique and *b*) the instrumental resolution. The continuous curve is a guide to the eye. *a*) $\Gamma \simeq 4.7$ keV; *b*) $\Gamma \simeq 1.8$ keV.

of a velocity-dependent nonexponential form for the broad component of the positron annihilation in gaseous argon [11].

5. – Search for one-photon annihilation.

The observation of one-photon annihilation would be quite significant in the study of the decay process of the positron [12]. This process can be represented as

$$e^+ + e^- + e^- \rightarrow e^- + \gamma \,.$$

From the principle of conservation of energy and conservation of momentum, it follows that the energy of the photon in the above process should be $\frac{4}{3}mc^2$ ($= 681.2$ keV). The Feynman diagrams for this process and for the two-photon decay are given in Fig. 8. It should be noticed that the one-photon annihilation amplitude involves an overlap integral containing the wave functions of the positron and the two electrons. This amplitude should give information about the electron-electron correlation in a solid. Consequently, the observa-

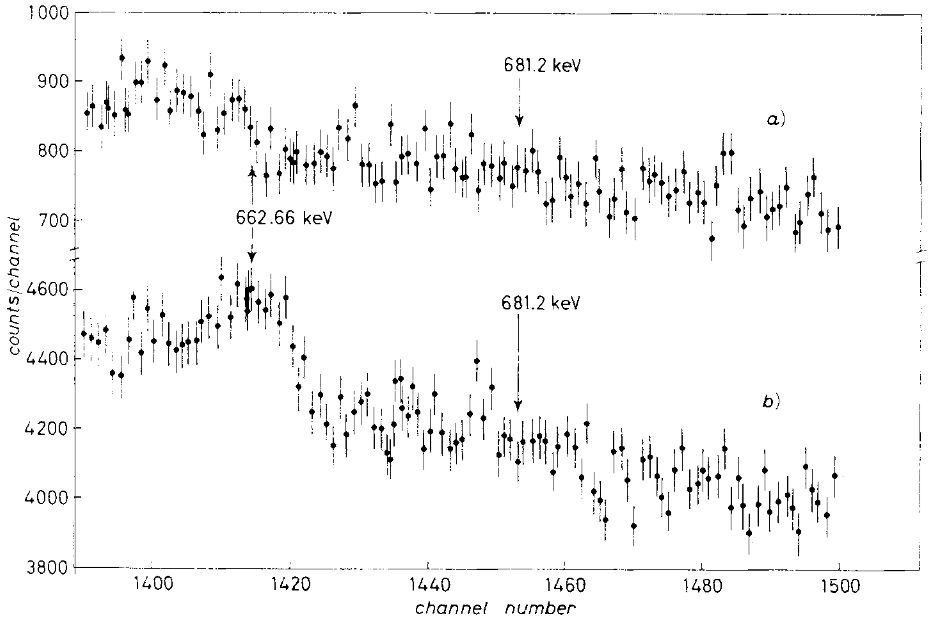

Fig. 8. – Feynman diagrams for *a*) two-photon and *b*) one-photon positron annihilations.

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[11] W. R. Falk, P. H. R. Orth and G. Jones: *Phys. Rev. Lett.*, **14**, 447 (1965).

[12] The importance of this correlation was emphasized by J. Bardeen to one of the authors (K.R.R.) in a private conversation.

tion and utilization of the one photon decay could be significant for studying the electronic structure of solids where there is no other alternative method to measure this correlation directly.

We have attempted to observe this process. In this search, the energy spectrum of the annihilation radiation, superimposed on the background spectrum from the source, was measured a Ge(Li) detector. In one search, potassium metal was used. ^{64}Cu was used as a positron source in preference to ^{22}Na; in the energy region of interest the signal-to-background ratio is better with the ^{64}Cu source. That is to say the ratio of the 510.9 keV peak to the nearby ($\sim$ 681 keV) background is significantly higher. The energy region of the photopeak, due to 681.2 keV, was searched carefully and the gamma-ray spectrum, in this energy region, is given in Fig.9 *b*). The results show, after a considerable period of counting time, that the rate for the one-photon annihilation in potassium relative to the two-photon annihilation is $\leqslant 7.0 \cdot 10^{-5}$.

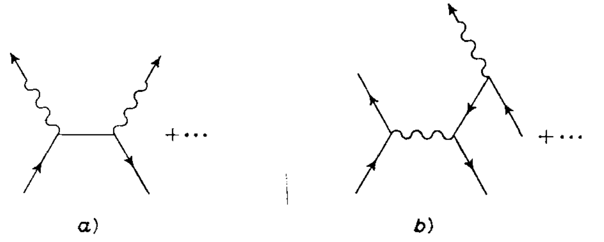

Fig. 9. – Energy spectrum in the region of 681.2 keV for the one-photon annihilation search *a*) in helium gas under pressure and *b*) in potassium.

In another search helium gas under pressure was investigated using ^{64}Cu for a positron source. Helium was selected for its high electron-electron correlations. The experimental result, in the relevant energy region, is shown in Fig. 9 *a*). From this data the ratio of one-photon to two-photon annihilation cross-section is $\leqslant 1.7 \cdot 10^{-4}$. In this estimate the statistical errors and the correction due to the positronium contribution ([13]) have been considered. Other possible errors, if any, are assumed to be small. A *similar* negative result has been reported recently by MACKENZIE and MCKEE with a statistical accuracy of $\leqslant 7 \cdot 10^{-4}$ ([14]). In this measurement the authors were interested in detecting the one-photon annihilation of the *positron polaron state* in KCl and BeO.

([13]) J. WACKERLE and R. STUMP: *Phys. Rev.*, **106**, 18 (1957).
([14]) I. K. MACKENZIE and B. J. A. MCKEE: *Bull. Am. Phys. Soc.*, **12**, 687 (1967).

To understand our experimental results, it is useful to compare with theoretical calculations. Detailed calculations are not available at present. However, an order of magnitude estimate can be made for the relative cross-section of one-photon to two-photon decay. The statistical factors, energy denominators and the annihilation amplitudes together do not differ very much in both cases. However, the amplitude for the Coulomb scattering of the positron by a «spectator» electron (see Fig. 8) into the proper momentum state and the amplitude for virtual emission of the photon in the two-photon decay differ considerably. In the one-photon annihilation process there is an extra vertex compared to the two-photon annihilation; however, the small overlap of electron-one and electron-two reduces the transition amplitude somewhat. Estimation of these amplitudes shows that the ratio of cross-sections for one-photon to two-photon processes, in a solid, is of the order of α^4 ($\alpha = e^2/\hbar c$) —which is extremely small. Thus our negative result is in agreement with theoretical expectations. It also shows that such an extremely small cross-section for one-photon annihilation, even if it is observed in the present manner, makes it an impractical research tool in solid-state physics.

6. – Conclusions.

We have undertaken these investigations to explore the use of high-resolution gamma-ray detectors for investigating Doppler broadening of annihilation radiation. Such an investigation necessarily requires a comparison to the highly successful angular correlation technique which measures essentially the same quantity. At the present stage of development the effective resolution of the Doppler broadening technique after computer analysis is about equal to many of the experimentally measured quantities obtained with angular correlation. However, the data acquisition rate is typically more than an order of magnitude faster with the Doppler broadening technique. Angular correlation is much better suited to most conventional measurements where resolution is most important and a certain portion of the momentum spectrum can be scanned at will. However, the Doppler broadening technique may have greater utility for measurements of certain effects, such as the wings of the annihilation distribution, where resolution is no longer as important and rate is increasingly so. In addition, it may be useful in situations requiring cumbersome target constraints.

We have examined several cases, such as annihilation in solids and in gases, where changes in external parameters cause a change in the annihilation line shape. In this connection a double subtraction method has been used to detect line shape changes and to determine Doppler broadening of the broad component annihilation radiation in gases. Here the high count rate and very stable intrinsic line shape of the Doppler broadening technique are useful features.

We have also searched for one-photon annihilation ($E_\gamma = (4/3)\, mc^2$). Our experimental limit for the relative cross-section of one-photon annihilation is less than α^2. Consequently, this potentially useful technique for measuring electron-electron correlation does not seem feasible using only a Ge-Li detector.

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RIASSUNTO (*)

Si è misurata la dilatazione Doppler della radiazione di annichilazione del positrone con un rivelatore al Ge drogato con Li avente risoluzione di 1.76 keV FWHM per la linea γ da 482 keV del ^{181}Hf. Dagli andamenti osservati dei fotopicchi della radiazione di annichilazione in vari metalli, si è ottenuta la forma funzionale della distribuzione degli impulsi della coppia che si annichila. L'analisi col metodo dei minimi quadrati delle curve sperimentali fornisce valori per la FWHM delle curve di distribuzione degli impulsi della coppia rivelata di 2.7 keV per Cu, e 2.5 keV per Zn. Queste ampiezze sono coerenti con le misure di correlazione angolare dei quanti di annichilazione. Si presentano i risultati ottenuti nello smorzamento del campo elettrico in O_2 gassoso. Si presenta un metodo di sottrazione doppia e si discute la sua utilità nella determinazione della dilatazione Doppler della radiazione derivata dall'annichilamento del positone con elettroni atomici legati. Si è cercato il decadimento di un fotone ($E = (4/3)mc^2$). Il limite superiore per la sezione d'urto relativa dell'annichilazione da uno a due fotoni nel potassio metallico è $\leqslant 7.0 \cdot 10^{-5}$. Si confrontano i meriti relativi della dilatazione Doppler e le tecniche convenzionali.

(*) *Traduzione a cura della Redazione.*

О допплеровском уширении излучения при двух-фотонной аннигиляции позитрона.

Резюме (*). — Были измерены допплеровские уширения при аннигиляции позитрона с помощью германиевого детектора с дрейфующим литием, который имеет разрешение 1.76 кэВ FWHM для γ линии ^{181}Hf 482 кэВ. Исходя из наблюденных форм фото-пиков излучения аннигиляции в различных металлах, была получена функциональная форма импульсного распределения аннигилирующей пары. Анализ наименьших квадратов полученных экспериментальных кривых дает значения для FWHM кривой импульсного распределения пар, как например, 2.7 кэВ для Cu и 2.5 кэВ для Zn. Эти ширины согласуются с измерениями угловой корреляции аннигиляционных квантов. Приводятся результаты, полученные при электрическом поле, подавляемом в газообразном O_2. Предлагается метод двойного вычитания и обсуждается его полезность при определении допплеровского уширения излучения, образующегося при аннигиляции позитронов со связанными атомными электронами. Были проведены поиски одно-фотонного распада ($E=(4/3)mc^2$). Верхний предел для относительного поперечного сечения одно-фотонной к двух-фотонной свободной аннигиляции в металлическом калии составляет $\leqslant 7.0\cdot 10^{-5}$. Проводится сравнение относительных достоинств допплеровского уширения и общепринятой техники.

(*) *Переведено редакцией.*