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ABSTRACT

A gas scintillation counter is described which operates to 40 atm pressure. An attempt was made to maximize the scintillation light output in helium. Helium-scintillation properties studied with this counter include the effect of various wall-coating materials, gas-purity effects, the linearity of pulse height with energy, and the variation of pulse height with pressure. With purified helium and for a given amount of energy deposited in the gas, the scintillation pulse height was found to increase with pressure P approximately according to the function $kP/(1+KP)$, where k and K are proportionality constants. A semi-quantitative argument is presented, explaining this pulse-height increase with pressure in terms of electron-ion recombination times in helium.

I. INTRODUCTION

In conjunction with an experiment to measure muon-capture rates in helium,[1] we have studied some properties of scintillations in helium gas. This article reports our observations, and attempts to explain an observed increase in pulse height with gas pressure. A recent article by Murray summarizes a large fraction of the work done on noble-gas scintillation.[2] Because the mechanism of scintillation is complicated, most information about gas scintillation is empirical. By coating the walls of the gas container with a "wave shifter" backed by a reflecting material, previous investigators obtained the most light output for a given amount of deposited energy. (A wave shifter is a phosphor that absorbs the ultraviolet light of the primary scintillation and radiates it in a spectral region more nearly matching the spectral sensitivity of the photocathode.) Using the previous investigations as a guide, we performed several tests in an effort to increase the light collected by the phototube. The final configuration of our gas counter is similar to that described by Shamu.[3]

II. COUNTER CONFIGURATION

Figure 1 is a sketch of our apparatus. Helium gas at liquid-nitrogen temperature ($\approx 80°K$) was held in a cylindrical stainless steel vessel with Lucite windows. Pressures ranged from 0 to 37 atm (absolute).

A leaktight seal of Lucite to stainless steel at liquid-nitrogen temperatures and pressures up to 40 atm was difficult to obtain. The successful design, achieved after many tests, is shown roughly in Fig. 1.[4] The seal is basically a shaped Kel-F O-ring bearing against specially shaped surfaces. All the contacting surfaces were painstakingly polished, since the

most minute scratch constituted a large leak. A leaktight container was necessary both to keep the expensive He^3 gas (used in the muon-capture experiment[1]) from escaping and to prevent impurities from entering the gas cell while it was under vacuum. The final leak rate through the window seal was about 2 cc per month at 30 atm, and the loss through plumbing joints was perhaps a factor of 100 greater than this. Thus, the amount of gas lost through leaks was negligible.

The gas was surrounded by a cup-shaped plastic scintillation counter. This counter had an inner diameter of 4 in. and an inner length of 5 in. Counters B2, 3, and 5 (the cup counter) were used in the muon-capture experiment, and were not important for the scintillation tests. The inner wall of the cup counter was coated with a highly reflective, opaque layer of aluminum (0.3-μ thick) which prevented the scintillation light from the gas from mixing with scintillation light from the cup.

Light from the gas scintillator was channeled out of the front Lucite window through a hollow aluminized brass elbow into an RCA 7046 photomultiplier (counter 4). An attempt was made to take additional scintillation light out through the edge of the Lucite window; however, tests showed that not enough light came out this way to make the effort worthwhile.

Pulses from the phototube were sent through the electronics shown in Fig. 2. The positive signal from the last dynode of the phototube was used to gate the negative anode signal. After amplification the anode signal was delayed so that it fell within the 150-nsec pulse from the gate generator. Finally, the amplitude of the integrated anode pulse was measured on a pulse-height analyzer (PHA). Because the phototube and all the electronic components were linear, the amplitude of the pulse measured in the PHA was

proportional to the scintillation light occurring within the 150-nsec gate. The scintillation light in turn should be proportional to the energy left in the gas.

An Am^{241} α source deposited on the end of a curved steel wire could be inserted into the helium gas through a gas-lock system. The source could be moved around to different parts of the gas cell in order to check the uniformity of response inside the gas volume. It was also used for frequent energy calibrations of the gas scintillator. Other calibrations were obtained from the reactions $n + He^3 \rightarrow p + H^3$ ($Q = 0.77$ MeV) and $\mu^- + He^3 \rightarrow H^3 + \nu$ (triton recoil energy = 1.896 MeV).

III. WALL COATINGS

Three different wall coatings were tried: (a) sodium salicylate on aluminum, (b) p-p' diphenylstilbene (DPS) on aluminum, and (c) DPS on MgO backed by aluminum. The front Lucite window had a transparent coating of about 30 $\mu g/cm^2$ of DPS in all the tests as well as in the final experimental arrangement. The DPS and aluminum coatings were in all cases obtained by vacuum evaporation. The MgO was smoked onto the cup counter by burning a magnesium ribbon in air.

The sodium salicylate, dissolved in a methyl alcohol solution, was sprayed onto the aluminum surface to an average thickness of ~1 mg/cm^2. Under optimum conditions this coating gave a resolution (FWHM) of 27% of the pulse height for the 5.4-MeV Am^{241} α source. This variation in pulse height was due to the random nature of photon collection and indicated that the photomultiplier collected ~60 photoelectrons for each α particle pulse. There was also a 30% variation in pulse height as the α source moved over the volume of the helium gas. In the actual experiment, light came from muon-capture events all over the gas volume, and therefore this variation adds to the resolution coming from photoelectron statistics.

In the test with about 100 $\mu g/cm^2$ of DPS coated directly on the aluminum surface, the pulse-height variation over the gas volume was extremely bad and in fact varied as the solid angle of the phototube, showing that only light heading toward the phototube reached it. That is, there was very little reflection of light from the DPS and aluminum surface. This is in accord with the view that a large fraction of the light generated in the DPS undergoes total internal reflection. In an attempt to get the internally reflected light out, several samples were made with the Al and DPS coatings on a rough (sandblasted) surface. This increased the light output substantially; however there was still much less light from this combination than with the DPS deposited on a white surface, such as paper.

The final configuration tested consisted of a ~50 $\mu g/cm^2$ thick layer of DPS backed by ~1 mg/cm^2 thick layer of MgO deposited over the aluminum coat that covered the cup counter. Magnesium oxide is a white, diffusely reflecting substance and has a high reflectivity for both visible and ultraviolet light. Because of its granular surface, it does not allow much light to be internally reflected in the DPS coating. With the MgO and DPS combination, the α source had a 16% resolution under the best conditions and the pulse height varied only 5% over the volume of the gas. Thus this combination is about three times more efficient than the sodium salicylate wave shifter. Since it did not appear that we could do much better than this, the MgO and DPS combination was chosen for the muon-capture experiment.

IV. GAS-PURITY EFFECTS

Many researchers have found that the purer the gas, the more scintillation light output.[5] Our results again confirm this fact. At room temperature we found that the pulse height decreases to about half in 2 hours.

This presumably is due to contamination of the gas by the wave-shifter coating. When the gas container was cooled to dry-ice temperature (195°K) this effect disappeared and the pulse height remained stable in time; in fact, a contaminated gas recovered its original properties. Additional cooling (to liquid-nitrogen temperatures) did not further increase the pulse height.

By passing reactor-grade helium through powdered charcoal maintained at liquid-nitrogen temperature, we doubled the scintillation pulse height. Passing the helium through a liquid-nitrogen trap improved the light output only slightly, thus showing that the absorbing properties of charcoal were essential in the purification. All indications were that a further purification of the gas could lead to an additional increase in pulse height.

V. PRESSURE EFFECTS

Early in the scintillation tests we noticed that the pulse height increased with increasing pressure. This effect occurred with both the DPS and sodium salicylate wave shifters. Figure 3 shows the pulse-height variation with pressure of the α source's 5.4-MeV peak. The range of the alpha was entirely within the gas for all points above 2 atm. Gas impurities changed the shape of this curve by causing lower saturation levels.

Shamu reports a similar pulse-height increase with gas pressure,[3] but gives no explanation for this increase. Rubbia and Toller report there is no variation in pulse-height for helium pressures between 20 and 100 atm.[6] Figure 3 seems to confirm this. The small decrease in pulse height above 30 atm is probably due to absorption of the scintillation light by the source holder, since at these pressures the range of the α particle is of the same order of magnitude as the source dimensions. If the data of Avivi and Cohen[7] for scintillations in Xe and Kr below 1 atm (room temperature) is combined

with that of Sayres and Wu[8] above 20 atm, one can see that a shape like Fig. 3 is a general property of noble gas scintillation. The overall pressure variation shown in Fig. 3 has not been reported previously,[9] because for one reason or another observers did not make measurements between 1 and 10 atm. It also appears that the pulse height in the heavier noble gases increases more rapidly with pressure and therefore saturates at lower pressures.

The increase in pulse height up to pressures of 20 atm is associated with the lifetime of light emission from helium. If the helium scintillation pulses are observed on an oscilloscope, one notices a variation with pressure in the timing of the scintillation light. At low pressures (~1 to 2 atm) some pulses associated with the α particle occur as much as 20 μsec after the initial pulse. As one increases the gas pressure these late pulses occur sooner, and by the time the pressure has reached 20 atm one can no longer observe any slow component of the light. At all pressures it appears that if one could integrate all the output light over time, there would be no variation with pressure. In our electronics system the pulse-height analyzer measures only helium scintillation light that occurs in the first 100 nsec after the start of scintillation. Thus the variation of the pulse height with pressure is due to a variation in the timing of the output light. Avivi and Cohen also observe this pressure variation of the light-emission rate in Xe and Kr and obtain light-emission lifetimes of ~1 μsec at 1/2 atm. Qualitatively one would expect that the electron and ion densities associated with ionization by an α particle would increase proportionally with the pressure (the α range is inversely proportional to the pressure). This would make it easier at the higher pressures for the electrons and ions to recombine and to produce the scintillation light. Avivi and Cohen attribute

their long decay times to the production of metastable states that yield light when collisions occur with other atoms. However, the semi-quantitative calculation given below shows that these long decay times are adequately explained by the recombination lifetime of electrons and ions.

VI. ELECTRON-ION RECOMBINATION TIMES IN HELIUM

If n_- and n_+ are electron and ion densities respectively, then the rate of recombination is[10]

$$\frac{\partial n_+}{\partial t} = \frac{\partial n_-}{\partial t} = -\zeta\, n_+ n_- , \tag{1}$$

where ζ is the recombination constant and equals 1.7×10^{-8} cc/ion-sec in helium at ~1/30 atm.[11] The recombination constant should be independent of pressure, but no measurements of ζ have been made in our pressure range. The first differential equation gives $n \equiv n_+ = n_- + c$ where $c = 0$ for an electrically neutral gas. Substituting n in Eq. (1) and solving the differential equation, one obtains

$$n = \frac{n_0}{1+n_0\zeta t}, \tag{2}$$

where n_0 is the initial ion density. The density of recombinations occurring between $t = 0$ and t_1 is just the difference between the initial ion density, n_0, and the density at time t_1, i.e.,

$$n_0 - n = \frac{n_0^{\,2}\,\zeta t_1}{1+n_1\zeta t_1}. \tag{3}$$

The light output is proportional to the number of recombinations, N, occurring between $t = 0$ and $t_1 \approx 100$ nsec, and one must multiply Eq. (3) by the volume of the ionization, V, to get a number rather than a density. Thus

one can write

$$N = (n_0 V)\left[\frac{KP}{1+KP}\right], \tag{4}$$

where $K \equiv n_0 \zeta t_1 / P$ is approximately independent of the pressure, P. Since V is roughly inversely proportional to P and n_0 is proportional to P, the product $n_0 V$ should be independent of the pressure. Thus the light output can be written

$$L = \frac{kP}{1+KP}, \tag{5}$$

where k is a proportionality constant independent of the pressure. The solid line in Fig. 3 is a curve of this shape with K = 0.17/atm, and it fits the data points below 20 atm reasonably well. The discrepancy with Eq. (5) of the data above 20 atm could be real, but more likely is due to a nonadiabatic change in the gas pressure near 20 atm. Pumping the gas into the target required a rapid and uncontrolled increase in the pressure from 20 atm to the maximum pressure near 37 atm. All points in this region were obtained while decreasing the gas pressure. The rapid increase in pressure during pumping could have caused a decrease in the pulse height and could explain the deviation of the data points from the solid curve.

In any event, one can calculate a value for K from the expected α-ionization density to see if it checks with the value of K obtained from curve fitting. If one uses the ratio of 30 eV per ion pair produced by the α, one obtains for the expected α-ionization density

$$n_0 = \frac{1}{V}\,\frac{5.4\ \text{MeV}}{30\ \text{eV}}. \tag{6}$$

At a pressure of 7 atm the α range is 1 cm, and V will be a cylinder of 1-cm length and radius R. The value of R is not easy to calculate, but it is probably close to a mean free path in the gas, which is approximately 1000 Å at 7 atm pressure. Using this value for R and the definition of K, one obtains

$$K \equiv \frac{n_0 \zeta t_1}{P} = \frac{5.4\times10^6}{30} \frac{\zeta\, t_1}{\pi R^2 (1\text{ cm}) P} = 0.14\ \text{atm}^{-1}, \qquad (7)$$

which is in excellent agreement with the value of K obtained from curve-fitting Eq. (5) to the data. Because the approximations are rough and the value of ζ, the recombination coefficient, was extrapolated from 1/30 atm to $\approx$30 atm, one must consider the above calculation to be good only to a factor of $\approx$10. However, the excellent agreement between the two values of K supports a recombination-time explanation for the pulse-height variation of Fig. 3.

VII. SCINTILLATOR PERFORMANCE

Figure 4 shows the performance of the helium-gas scintillator in measuring three energies under experimental conditions. The light output was found to be linear with energy loss (see Fig. 5) and to vary only 5% over the volume of the counter. The final energy resolution of 38% for the 1.9-MeV triton recoil corresponds to an average of 30 effective photoelectrons. If one considers the 15% quantum efficiency of the phototube, one would expect 10^4 photons from 1.9-MeV deposited in the gas. Thus it appears that the detection of helium scintillation light is still a very inefficient process.

FOOTNOTES AND REFERENCES

* This work was done under the auspices of the U. S. Atomic Energy Commission.

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FIGURE LEGENDS

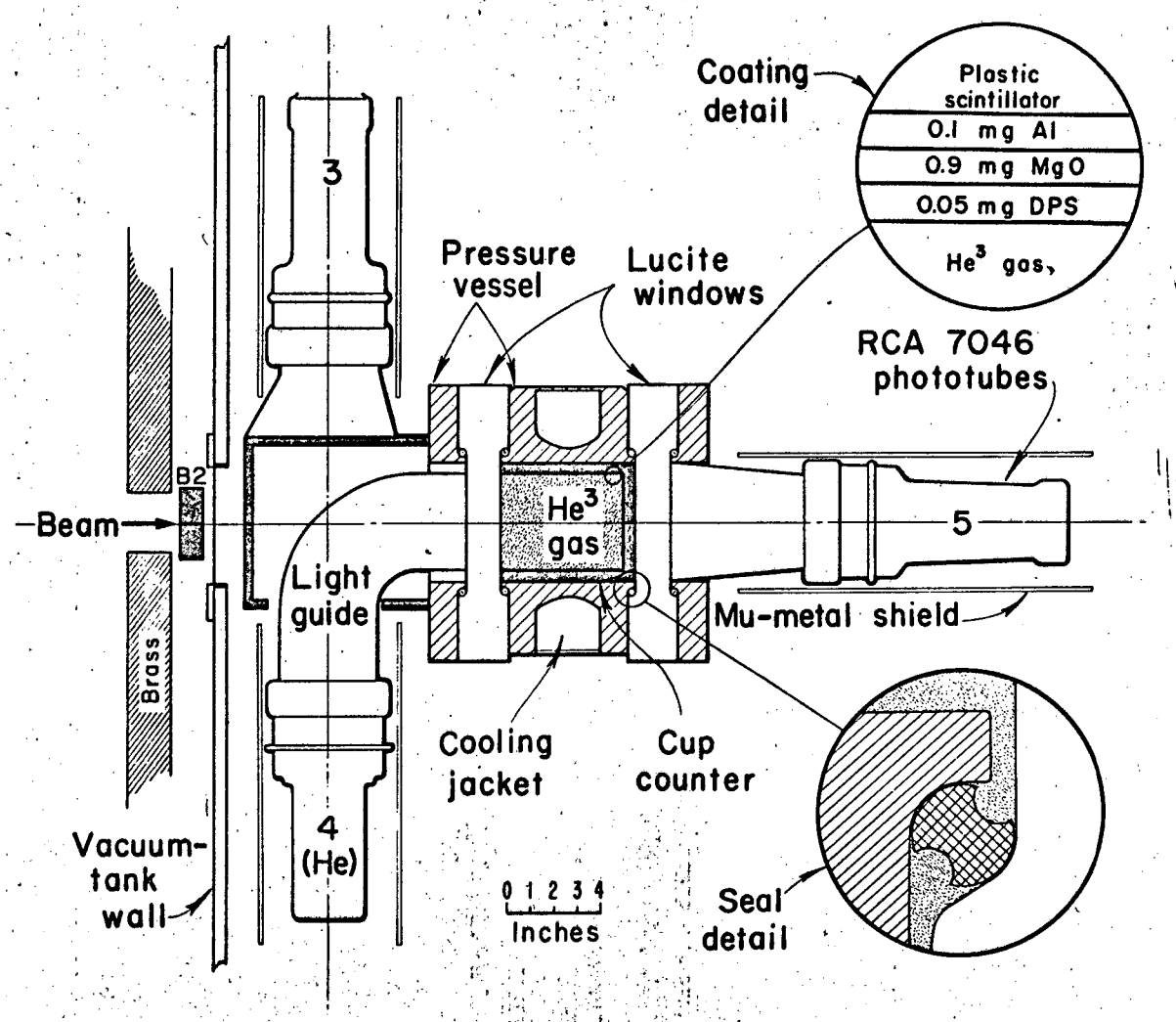
Fig. 1. Top view of the apparatus with details of the pressure seal and the cup-counter coating.

Fig. 2. Block diagram of the electronics used to analyze the amplitude of scintillation pulses.

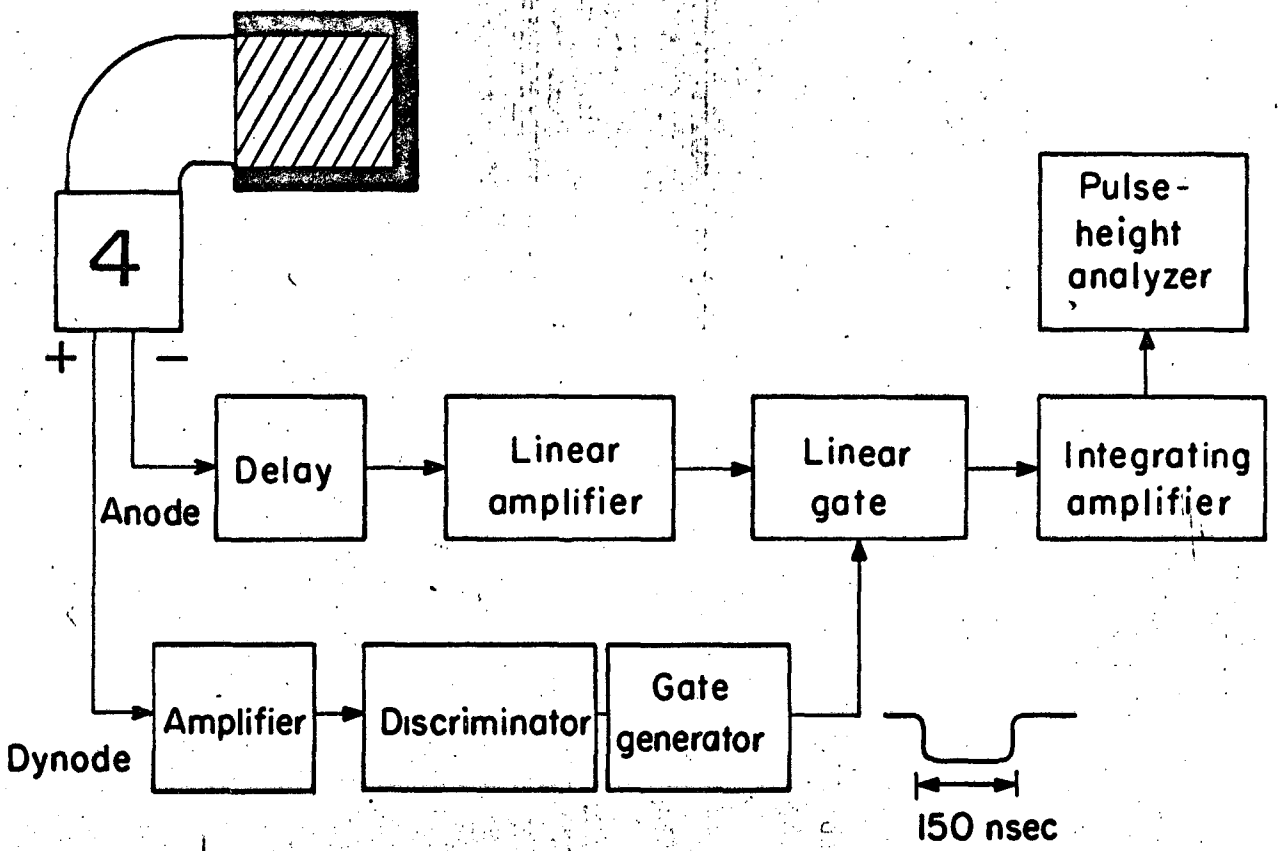
Fig. 3. Variation of a 5.4-MeV alpha-scintillation pulse height vs helium-gas pressure. Both He^3 and He^4 follow the same curve, and its shape is independent of the wave shifter used. The solid curve is a least-squares fit of the data points below 20 atm to a function of the form $kP/(1+KP)$ with $K = 0.17$, and the dotted curve is the function $1 - \exp(-KP)$ with $K = 0.19$, where P is the gas pressure.

Fig. 4. Calibration energy peaks showing the resolution of the helium scintillator. The neutron peak was obtained from the capture of thermal neutrons in He^3 (i.e., $n + He^3 \rightarrow H^3 + p$), in which 0.77 MeV is released; the triton peak is from the final data on muon capture in He^3 (i.e., 1.9-MeV triton recoils); the alpha peak is from an Am^{241} alpha source (5.4 MeV).

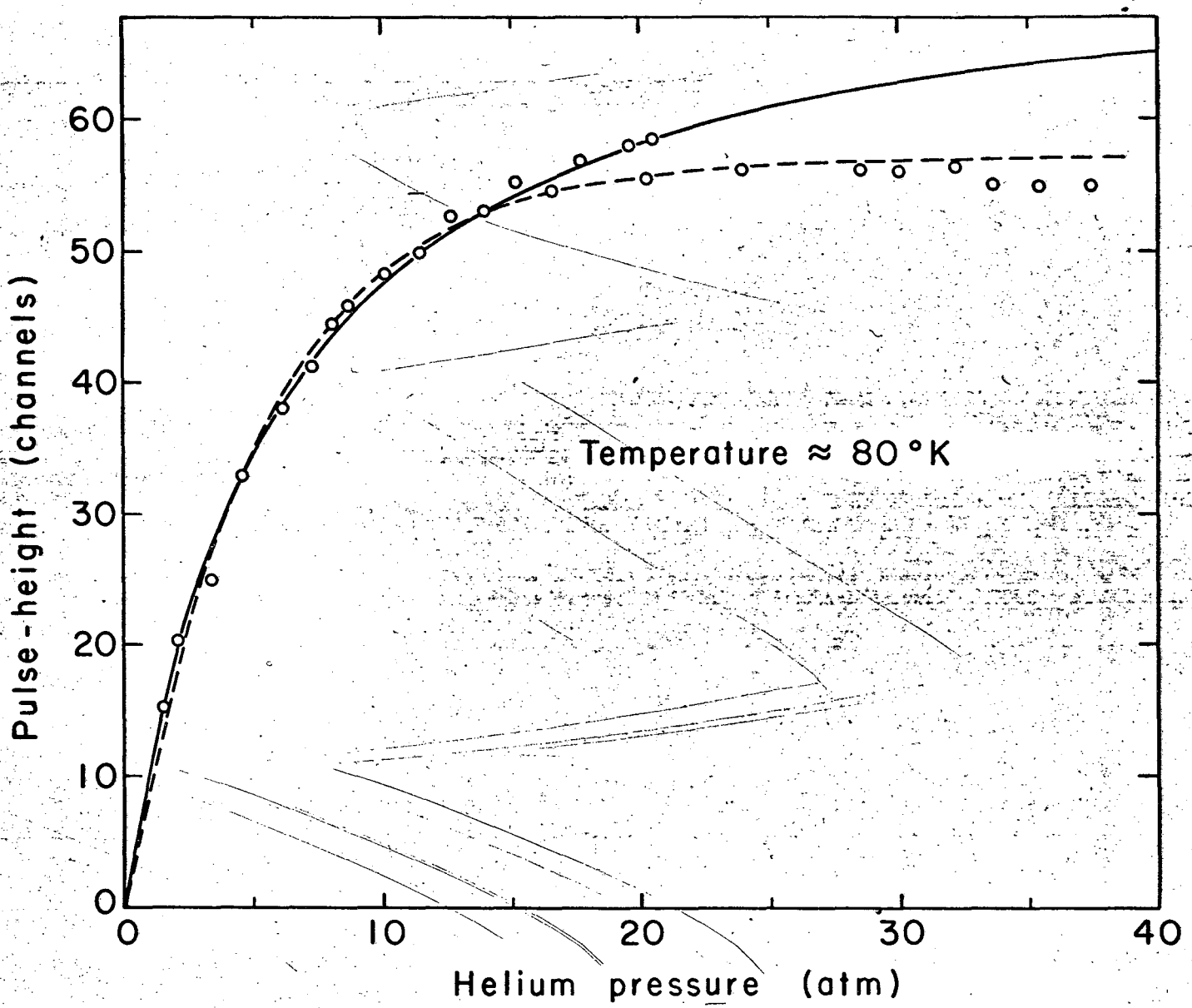
Fig. 5. Energy calibration of the pulse-height analyzer at two gas pressures: (A) neutron captures, (B) triton recoils, (C) Am^{241} alpha particles. The pulse height of the α scintillation at the higher gas pressure has decreased because some light is absorbed by the source holder at this pressure.


Coating detail
Plastic scintillator
0.1 mg Al
0.9 mg MgO
0.05 mg DPS
He³ gas,
3
Pressure vessel
Lucite windows
RCA 7046 phototubes
B2
Beam
He³ gas
5
Light guide
Mu-metal shield
Brass
Cooling jacket
Cup counter
4 (He)
Vacuum-tank wall
0 1 2 3 4
Inches
Seal detail

MUB-1673-B


4
+
−
Anode
Dynode
Delay
Linear amplifier
Linear gate
Integrating amplifier
Pulse-height analyzer
Amplifier
Discriminator
Gate generator
150 nsec

MUB-4335


Pulse-height (channels)
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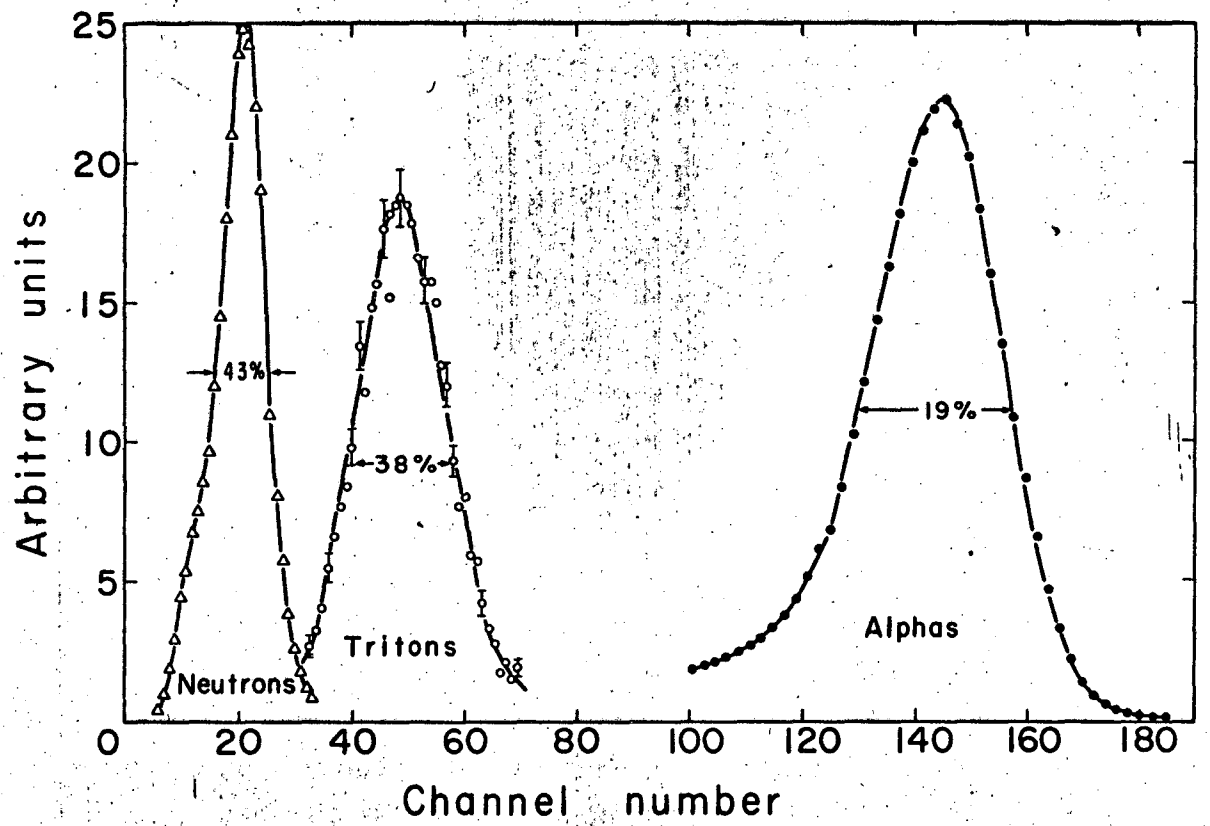
20
30
40
50
60
Temperature ≈ 80 °K
0
10
20
30
40
Helium pressure (atm)

MU-34003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20
15
10
5
0
Arbitrary units
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38%
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Neutrons
Tritons
Alphas
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40
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80
100
120
140
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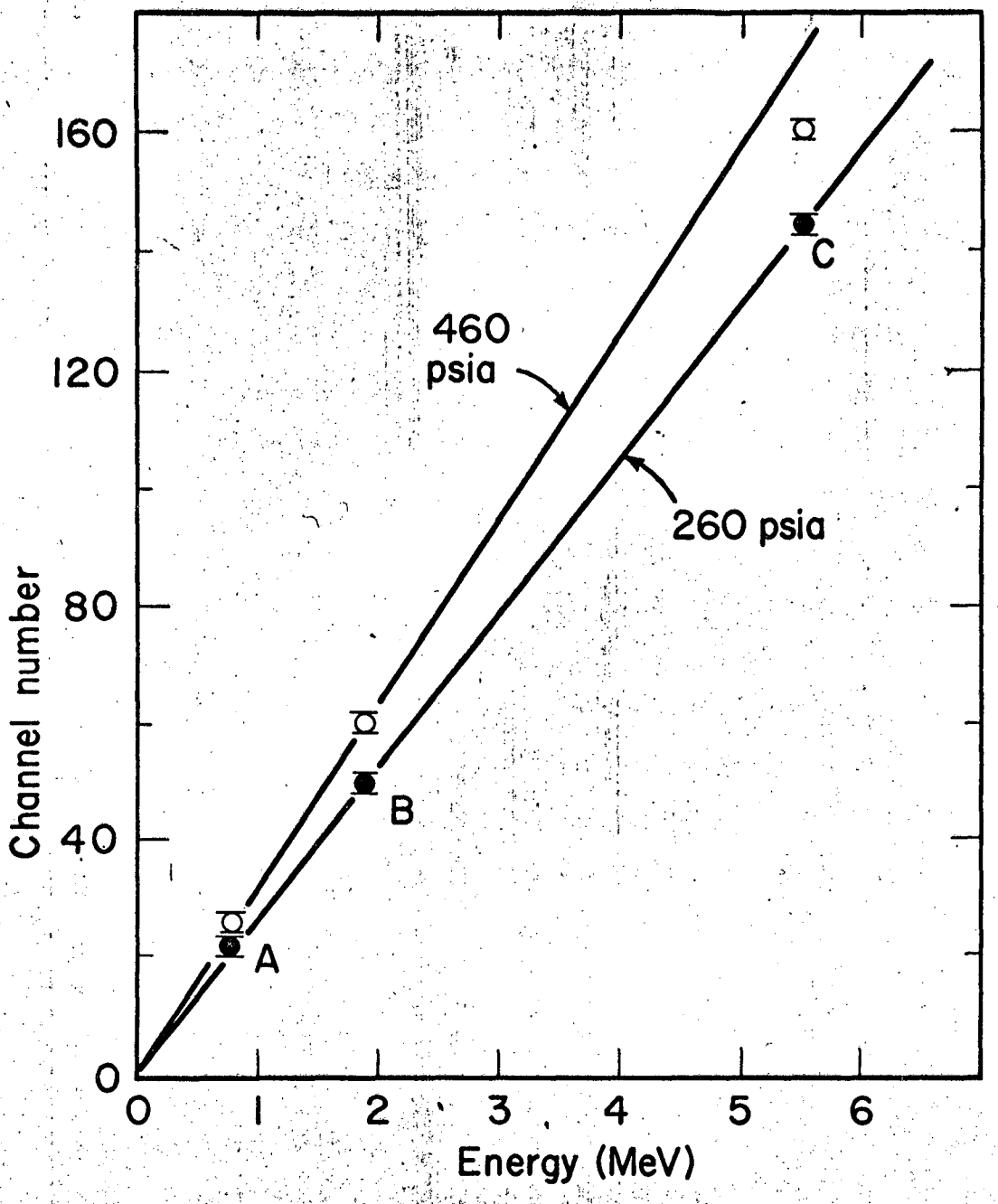
Channel number

MUB-1671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Channel number
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1
2
3
4
5
6
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psia
260 psia
A
B
C

MU-30090