Non-linearity of recoil pulse height events in He-3 tubes

M. Manolopoulou a,*, M. Fragopoulou a, S. Stoulos a, W. Westmeier b, A. Lagogiannis c, M. Zamani a

a School of Physics, Aristotle University of Thessaloniki, Thessaloniki 54124, Hellas (Greece)
b Philips-Universität, 35032 Marburg, Germany
c N.C.S.R. Demokritos, Ag. Paraskevi Athens 153 10, Hellas (Greece)

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1. Introduction

$^3$He gas has been widely applied to slow and fast neutron detection and spectrometry due to its high neutron absorption cross-section, chemical inertness and good behaviour in proportional counters with high pressure [1]. When neutrons interact with $^3$He, three competing reactions have to be considered: $^3$He(n,p)$^4$He, $^3$He(n,d)$^3$H and elastic scattering, each one of which is predominant in a different energy region. The cross-section of the $^3$He(n,p)$^4$He reaction is decreasing from 5330 barns for thermal neutrons, down to 2.6 barns for 56 keV neutrons, where it becomes equal to the elastic scattering cross-section. For high neutron energies, the elastic scattering cross-section becomes larger than that of the (n,p) reaction, and the difference becomes more pronounced with increasing neutron energy. For neutron energies above 4.3 MeV, the (n,d) reaction is also contributing, although the cross-section is an order of magnitude lower than that of the elastic scattering one [2].

For spectrometry applications, a linear energy response of the spectrometry system is expedient. In proportional counters, energy linearity is affected by the general and/or self-induced space-charge effect, the impact of which depends on the gas amplification, count rate, particle energy at high gas gains, pressure and other characteristics of the counters [3–7]. Recombination effects have also been proposed as an explanation for various effects, such as a count rate-dependent peak shift, an alteration of spectral response or peak shifts observed in Xe-based spectrometers at low neutron energies, and they were attributed to recombination effects [12]. In a comparative study of He-3 counters using mixtures with Ar or Kr, a distinct difference was found between the experimentally determined recoil peak position in the spectrum and the expected one for neutron energy 2.4 MeV in the counter containing Kr. The deviation became larger as the average Linear Energy Transfer (LET) of the recoiling nucleus was increased above 350 eV/μm [13].

Although recoil peak displacement has been observed sporadically for He-3 tubes, there is a lack of systematic study that could eventually lead to an energy calibration for recoil events. Such a calibration would be advantageous in neutron spectrometry applications because recoil events of high energies could be recorded in the same spectrum area as the events of $^3$He(n,p)$^4$He
reactions of lower energies. In this work, the pulse height produced by the recoiling $^3$He nuclei is studied as a function of neutron energy and for different operating voltages in a commercially available He-3 counter. Possible reasons for the recoil peak displacement are discussed. The stopping power function of the gas mixture for $^3$He nuclei, which is calculated via a Monte Carlo method, is used to derive the energy calibration equation for recoil events.

2. Experimental details

The characteristics of the He-3 tube used in this study\(^1\) are: an effective length of 15 cm, a diameter of 5 cm, a cathode material of stainless steel type SS304 and an anode of a tungsten wire with a 25 µm diameter. The filling gas apart from $^3$He (64.7%) contains Kr (33.3%) as a high stopping-power medium and CO$_2$ (2%) as quench gas. The overall pressure of the gas mixture is 6 atm (607.95 kPa).

The spectrometry system consists of a high-voltage power supply, a charge-sensitive pre-amplifier (Canberra 2006), a Gaussian pulse shape amplifier (Tennelec TC243) and a multichannel analyser (Canberra Multiprot II). In addition, a precision pulse generator (Ortec 419) was used for the adjustment of the system.

Irradiations were performed with mono-energetic neutron beams in the energy range from 463 keV to 6.7 MeV provided by a Tandem, Van de Graaff accelerator facility at the Institute of Nuclear Physics, NCSR Demokritos, Athens, Hellas. Neutrons with energies in the range of 463 keV to 3.3 MeV were obtained via the $^7$Li(p,n)$^7$Be reaction. When the projectile energy exceeds 2.2 MeV, the neutrons from the $^7$Li(p,n)$^7$Be* reaction is clearly distinguishable from the $^7$Be at the ground state, a second group of neutrons are also emitted from the $^7$Li(p,n)$^7$Be reaction (first excited state of $^7$Be, 0.429 MeV)\(^1,s,14\). At projectile energies above 3.68 MeV, neutrons from the $^7$Li(p, $^3$He)$^4$He reaction start contributing to the overall neutron yield, while at energies above 7.06 MeV, neutrons from the $^7$Li(p,$n_2$)$^7$Be* reaction (second excited state of $^7$Be) can also contribute to the spectrum. Among these, the $n_1$ neutrons have the highest yield, and at the projectile energies below 5 MeV used in this study, the zero-degree yield of these low-energy neutrons is less than 10% of the $n_0$ neutrons\(^1\). The peak due to neutrons from the $^7$Li(p,$n_1$)$^7$Be reaction is clearly distinguishable from the peak of the main neutron group ($n_0$ neutrons), due to the energy difference between the neutrons emitted from these two reactions\(^1,16,17\). Neutron energies in the range of 463 keV to 6.7 MeV were produced by the $^3$He(d,n)$^3$He reaction. When the projectile energy exceeds 2.2 MeV, neutrons from the $^3$He(n,p)$^3$T reaction; in Section 3.3, the energy calibration for recoil peaks; and finally, in Section 3.4, the equation for energy calibration for recoil events.

3. Results and discussion

Presentations include the following sections: in Section 3.1, the general features of the counter; in Section 3.2, the energy calibration of the peak arising from the $^3$He(n,p)$^3$T reaction; in Section 3.3, the energy calibration for recoil peaks; and finally, in Section 3.4, the equation for energy calibration for recoil events.

3.1. General features of the He-3 counter

In cylindrical proportional counters, the electric field strength per unit pressure (reduced electric field, $E_{Fr}$) along the radial distance $r$ from the counter axis is as listed below:

$$E_{Fr} = \frac{V}{\ln(r_b/r_a)} F_r,$$

(1)

where $r_a$ and $r_b$ are the cathode and anode radii, respectively, $V$ is the applied voltage and $p$ is the gas pressure. The high field-strength region in which amplification mainly occurs extends typically to a few wire radii, according to $r^{-1}$ dependence. In most of the counter volume, the ions are created by the primary interactions of the charged particle, the electric field just drifts the ions towards the corresponding electrodes. When the electrons reach the multiplication region ($r \leq r_c$), the development of the Townsend avalanches starts, resulting in a mean multiplication factor or gas gain $M$ given by the following:

$$\ln(M) = \int_{r_a}^{r_c} \alpha(r) \cdot dr,$$

(2)

where $\alpha(r)$ is the specific ionisation or first Townsend coefficient\(^1,s,14\). Several relations have been derived for the gas gain according to the function used for the Townsend coefficient $\alpha$, which is gas dependent\(^6,19,20\). Usually, several assumptions are made for simplification, like that the only multiplication process is through electron collision and that recombination, electron attachment to impurities and space charge effects are negligible\(^1,6,20\). One of the most widely used expressions for $M$ is the Diethorn equation, which was derived assuming the linearity of the specific ionisation with electric field strength and is shown below:

$$\ln(M) = \frac{V}{\ln(r_b/r_a)} \frac{\ln(2)}{\Delta V} \left( \ln \left( \frac{V}{p r_a \ln(r_b/r_a)} \right) - \ln K \right).$$

(3)

The constant $\Delta V$ corresponds to the potential difference through which an electron moves between successive ionising collisions, while $K$ is the minimum reduced electric field strength required for ionisation\(^1\). The two parameters can be experimentally determined as constants of the linear relationship fit of $[\ln(M) \cdot \ln(r_b/r_a)]/V$ vs. $[\ln(V/p \cdot r_a \cdot \ln(r_b/r_a))]$.

The pre-amplifier pulse amplitude $V_{pr}$ was measured for thermal neutrons from a paraffin-covered Am–Be source, as a

\(^1\) Manufactured by LND INC., New York, USA.
Table 1
Average energy required to produce an ion pair [23], W, and stopping power, S, of a 764 keV proton for pure gases [24].

<table>
<thead>
<tr>
<th></th>
<th>W [eV/ion pair]</th>
<th>S [MeV cm² g⁻¹]</th>
</tr>
</thead>
<tbody>
<tr>
<td>He</td>
<td>42.3</td>
<td>345.7</td>
</tr>
<tr>
<td>Kr</td>
<td>24.05</td>
<td>120.4</td>
</tr>
<tr>
<td>CO₂</td>
<td>32.8</td>
<td>268.2</td>
</tr>
</tbody>
</table>

function of the high voltage applied. Amplitude is related to gas gain through the equation: \( V_{Ap} = N Ne/M/C \), where \( N \) is the number of primary ion pairs formed by the charged particle, \( C \) is the capacitance of the counter and \( e \) is the elementary charge. The number of primary ion pairs created by a particle of energy \( E \) is calculated with \( N = E/W_m \), where \( W_m \) is the average energy spent to produce one ion pair in the gas mixture. The \( W_m \) value is calculated to be 37.9 eV according to the expression introduced in ICRU Rep.31, neglecting the quenching gas contribution [21,22]. Values for stopping power (S) and average energy required to produce an ion pair (W) for each of the constituents of the filling gases are presented in Table 1. Gas amplification values measured are in the typical range for \(^3\)He neutron-counting applications, varying from 6 to 44 in the proportionality region of the counter.

Gas gain as a function of the applied high voltage is presented in Fig. 1. The application of the Diethorn equation for gas gain results in the following parameter estimates: \( V_{Ap} = 35.65 \pm 0.69 \) V and \( K = 1.471 \times 10^4 \pm 2.3 \times 10^2 \) V/(cm atm). The latter indicates that the multiplication region is limited to about two anode radii.

When \(^3\)He counters are irradiated with mono-energetic neutron beams, the resulting pulse height distributions are composed of

i. The “full energy” peak. This peak is formed when the total kinetic energy of the \(^3\)He(n,p)\(^3\)H reaction products is deposited in the effective gas volume. The reaction is exothermic, with \( Q = 764 \) keV. Proton and triton share the energy of the reaction, and in the case of thermal neutrons, they are moving in opposite directions, receiving 573 and 191 keV, respectively. Thus, thermal neutrons as well as neutrons with energy less than the Full Width at Half Maximum at 764 keV are recorded in this peak, usually called the thermal–epithermal peak. At high energies, the neutron kinetic energy is added to the Q value of the reaction, and the total kinetic energy is distributed among the two reaction products according to their emission angles in the lab frame. For neutrons with energies above several MeV, the particle ranges become larger than the dimensions of the effective volume of the counter. As a result, a fraction of the energy is deposited to the wall of the counter, the so-called wall effect. Therefore, the maximum neutron energy that can form a full energy peak depends on the geometrical characteristics of the counter and the stopping power of the filling gas. Proton and triton ranges for the counter used in this study are presented in Fig. 2, as calculated with SRIM/TRIM code [18]. According to these data, the maximum neutron energy that can form a full energy peak is around 7 MeV, as above this energy, the proton range becomes larger than the effective length of the counter.

ii. The recoil distribution. The energy transferred to a nucleus via elastic scattering of a neutron is defined as the following:

\[
E_R = \frac{4A}{(1 + A)^2} \cos^2 \theta E_n, \tag{4}
\]

where \( E_n \) and \( E_R \) are the neutron and recoiling nucleus energies, respectively, \( A \) is the ratio of the target nucleus mass to the neutron mass and \( \theta \) is the scattering angle of the recoiling nucleus in the lab coordinate system [1]. The cross-section of the \(^3\)He(n,elastic) reaction is larger than that of \(^3\)He(n,p)\(^3\)H for neutron energies above 56 keV, e.g., it is 2.4 times larger for a 1 MeV neutron energy [2]. As all scattering angles are allowed, in principle, the result in the spectrum is a continuum between a minimum of zero energy and a maximum of whenever the recoiling nucleus is emitted at 0° with respect to the neutron direction. This maximum energy is calculated via equation (4) to be 75% of the neutron energy for the \(^3\)He nucleus and to be 29%, 22% and 5% for the C, O and Kr nuclei, respectively. It should be pointed out that although the cross-section of elastic scattering for fast neutrons is larger than the \(^3\)He(n,p)\(^3\)H reaction, the result in the pulse height distribution is not so prominent as compared with the full energy peak, due to the distribution of the recoil events over a wider energy region of the spectrum (Fig. 3).

iii. Gamma-ray distribution. \( \gamma \)-rays interact mainly with the counter walls and Kr atoms producing electrons. Due to their long range, electrons can deposit a limited amount of energy (usually less than about 200–300 keV) in the counter,
producing pulses with small amplitudes and long rise times. These pulses are recorded in the low-energy region of the pulse height distributions, presenting an exponentially decreasing function of which the exponent is a function of the $\gamma$-ray field intensity [1,17,25].

For neutron energies above 4.3 MeV, the $^3$He(n,d)D reaction contributes to the pulse height distributions, with $Q = -3.27$ MeV. The cross-section of this reaction is from 1 mb at 4.38 MeV to 57.5 mb at 7 MeV (Fig. 3).

### 3.2. Full energy peak calibration

Pulse height distributions were obtained for 14 neutron energies in the range from 463 keV to 6.7 MeV using five operating voltages between 1500 and 1870 V. With increasing applied voltage, the gas amplification rises exponentially, resulting in higher pulse amplitudes. To compensate the results of varying high voltage, the amplification factor of the main amplifier had to be changed accordingly so that pulses corresponding to the full energy peak had final amplitudes less than the maximum voltage that can be processed by the Analog-to-Digital Converter (ADC, maximum pulse height 10 V). The comparison of the pulse height distributions is accomplished by normalising the spectra to the same amplifier gain, using measured amplification factors. This procedure was considered more appropriate than normalisation of spectra to the centroid of thermal peaks, as the energy distribution of scattered neutrons from the surrounding area was unknown. It should be noted here that as the counter was covered with 1 mm Cd foil, the “thermal” peak registered neutrons with energies above about 1 eV up to several tenths of keV due to system resolution. Some of the reduced pulse height distributions obtained are presented in Fig. 3. Using the above procedure for spectra normalisation, the thermal peak centroids of the reduced spectra did not show any significant variation in each applied voltage (Table 2).

While thermal peaks exhibit a Gaussian shape, full energy peaks show an asymmetry that increases with energy at their left/low-energy part, indicative of incomplete charge collection. A similar effect was observed by Loughlin et al. in a He-3 ionisation counter [26] and by Dietz et al. in a He-3 proportional counter [27]. The physical reason for this asymmetry was attributed either to pre-amplifier pulses with long rise times, when protons and tritons are emitted perpendicularly to the anode, or to a space-charge effect. Asymmetry was also observed in the thermal peak during a previous study of the same type of counter as that used in the present work when the operating voltage was increased [17]. In this case, the asymmetry was connected to longer rise time pulses, thus to particles with trajectories almost perpendicular to the anode, and it was attributed to a kind of self-induced space charge effect: when the electron swarm of one of the two particles reaches the multiplication region, it prevents proportional avalanche growth of the second particle.

Full energy peaks were fitted by adding two Gaussian-shaped peaks of different Full Width at Half Maximum (FWHM) to take into account the observed asymmetry. System energy calibrations for the five used operating voltages used are presented in Fig. 4. A linear relationship of deposited energy in the counter with pulse height is observed for a neutron energy up to 6.7 MeV. However, the least squares fitted lines have an offset, a small positive value in channel zero, ranging from 40 to 150 keV with uncertainties from 14% to 46%. As the electronic system was accurately calibrated for zero, this positive offset is indicative of increasing incomplete charge collection with rising deposited energy in the counter. However, due to large uncertainties, no systematic behaviour can be observed with voltage change.

### 3.3. Recoil peak calibration

Pulse height distributions expected for the recoil energy continua have the same shape as the differential scattering cross-section in the centre-of-mass system, as a result of reaction kinematics [1]. The finite resolution of the system introduces a distortion of this shape, mainly in the area of maximum recoil energy [28]. To estimate the resolution influence on the recoil peak shape, dispersal according to a Gaussian distribution is assumed, and the differential scattering cross-sections are folded with the experimentally determined resolution function using Monte Carlo calculation [2,17]. Some of the calculated results are presented in Fig. 5, illustrating that the recoil peak maximum is expected in the spectrum at a somewhat lower energy than the utmost recoil energy. The difference is decreasing almost linearly from 4.5% to 1.5% for neutrons in the energy region of 1–7 MeV, as the resolution is improved with increasing energy (Table 3).

The recoil peak in the experimental spectrum was fitted using the right part of the peak, including the area around the peak centroid, with a suitable background function: this is linear for $E_n > 1.1$ MeV and exponentially decreasing for the $\gamma$-ray contribution in the lower neutron energies. The relation of the recoil peak

![Fig. 3. Pulse height distributions obtained with He-3 counter operating at 1700 V for several neutron energies, normalized to the same amplifier gain.](image)

**Table 2**
Variation of thermal peak centroid in the reduced spectra for each operating voltage.

<table>
<thead>
<tr>
<th>High Voltage [V]</th>
<th>Number of spectra</th>
<th>Centroid range: min–max (ch)</th>
<th>Centroid average ± 1 σ (ch)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1500</td>
<td>8</td>
<td>60.30–62.64</td>
<td>61.49 ± 0.90</td>
</tr>
<tr>
<td>1600</td>
<td>9</td>
<td>89.94–94.72</td>
<td>91.69 ± 1.54</td>
</tr>
<tr>
<td>1700</td>
<td>14</td>
<td>144.62–152.00</td>
<td>148.35 ± 2.73</td>
</tr>
<tr>
<td>1800</td>
<td>9</td>
<td>232.78–241.73</td>
<td>235.35 ± 3.32</td>
</tr>
<tr>
<td>1870</td>
<td>8</td>
<td>301.37–307.95</td>
<td>305.11 ± 2.92</td>
</tr>
</tbody>
</table>
centroid as a function of the reduced energy of recoiling \(^3\)He nuclei by the amount calculated as a result of resolution influence (Table 3) is presented in Fig. 6. Non-linear behaviour is observed, at least for lower energies. Similar results were presented for recoil helium peaks in other gas proportional and scintillation counters [12,29].

A comparison of the two energy calibrations of the system using full energy or recoil peak shows two distinctly separate curves. For the same deposited energy in the counter, there is a clear deviation between the two pulse heights, with the recoil always smaller than the full energy peak. Pulse height differences measured for different high voltages can be compared by converting them to energy using the corresponding full energy peak calibration of each operating voltage. That difference, denoted as \(\Delta p\), demonstrates the energy equivalent of the pulse-height loss between the expected and measured recoil peak centroid position in the spectrum. In Fig. 7, \(\Delta p\) is plotted against reduced recoil peak energy due to resolution influence.

Physical reasons that are known causes of pulse-height loss are space charge and recombination effects. In proportional counters, significant space charge effects have been observed at high count rates, as an integrated influence on the gas gain of the positive ions of many discharges as they drift across the counter (the general space charge effect). Also, a space charge effect has been observed when the counter operates in the limited proportionality region \((10^2-10^6\) electrons in an avalanche\) as a self-induced phenomenon that is caused by the large total charge density in the avalanche [3–7,30]. In the latter case, the space charge produced in an avalanche by the preceding electrons in a primary electron cloud is so large that the electric field strength near the anode is weakened, thus reducing the amplification of the following electrons. In the proportionality region and with low

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**Table 3**
Difference between maximum \(^3\)He recoiled nucleus energy and expected recoil peak energy [MeV], due to resolution influence.

<table>
<thead>
<tr>
<th>Maximum recoil (^3)He energy (MeV)</th>
<th>Expected recoil peak energy (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.51</td>
<td>0.49</td>
</tr>
<tr>
<td>0.67</td>
<td>0.64</td>
</tr>
<tr>
<td>0.83</td>
<td>0.79</td>
</tr>
<tr>
<td>1.36</td>
<td>1.31</td>
</tr>
<tr>
<td>1.51</td>
<td>1.46</td>
</tr>
<tr>
<td>1.89</td>
<td>1.82</td>
</tr>
<tr>
<td>2.50</td>
<td>2.42</td>
</tr>
<tr>
<td>2.82</td>
<td>2.73</td>
</tr>
<tr>
<td>3.29</td>
<td>3.20</td>
</tr>
<tr>
<td>3.78</td>
<td>3.69</td>
</tr>
<tr>
<td>4.21</td>
<td>4.12</td>
</tr>
<tr>
<td>4.65</td>
<td>4.57</td>
</tr>
<tr>
<td>5.02</td>
<td>4.95</td>
</tr>
</tbody>
</table>
significant difference was found in
Moreover, as the relative peak shift due to the space charge (Fig. 7), the space charge influence on
general shift of the recoil peak to lower energies, an extra

distances should undergo less influence from recombination
in the studies of the limited proportionality region [7,30], due to
large proton, triton and 3He ranges in the counter (several m, see
Fig. 2) instead of several hundreds of μm for photoelectrons.
Moreover, as the relative peak shift due to the space charge
should be proportional to the gas gain [3] and, as no statistically
significant difference was found in Δp with high voltage variation
(Fig. 7), the space charge influence on Δp should not be
considered as the primary cause.

Recombination, the other known cause of pulse-height reduction,
is described by two main processes: the initial recombination or

geminate named after Onsager [31] and the volume recombina-
tion. In initial recombination, a fraction of electrons produced by
the primary charged particle are thermalised in the vicinity of
their parent ions and, in the absence of an electrical field, have
a 100% probability to recombine with them, while even in the
presence of a strong electrical field, the initial recombination
probability does not drop to zero. Initial recombination dominates
at low count rates, at gas densities of several mg/cm3 and at
electrical field strengths above 1 kV/m [32]. In the volume
recombination process, electrons thermalised away from their
parent ions can recombine with ions of another particle (thus
depending on the count rate) or with their parent ions being
under random thermal motion. In the case of low count rates,
even a small electrical field can remove the electron cloud from
the area of their parent ions, reducing the volume recombination
probability or overcoming it entirely. An extensive discussion of
these processes can be found in the works of Bolotnikov and
Ramsey [33] and Jaffe et al. [34].

The volume recombination process can be excluded as a reason
of the reduced pulse height of the 3He recoiling nucleus as
compared to pulse heights from proton-to-triton nuclei having
the same total kinetic energy, as its influence on both pulse
heights should be the same due to the random nature of this
process. Moreover, considering the operating conditions of the
counter, such as a low count rate and a high electric field above
10 kV/m, volume recombination can be excluded.

In contrast, initial recombination is a process that mainly
depends on the local ion density and the time that electrons
spend in the area of primary ionisation, which in turn, is based on
the electric field strength, density and electron mobility in the gas
mixture. At a given operating voltage, gas pressure and composi-
tion in a counter, the initial recombination will therefore depend
on the local ion density and the electric field strength in the area
of primary ionisation of each particle. In cylindrical counters,
the reduced electric field strength is inversely proportional to the
radial distance (Eq. (1)); thus, it is expected that pulses from
identical particles with the same kinetic energy will undergo
different degrees of reduction depending on the radial distance of
their electron cloud as well as the orientation of their particle
trajectory in respect to the anode. For pulses belonging to the
recoil peak, 3He nuclei travelling parallel to the anode at closer
distances should undergo less influence from recombination
effects than those at larger distances. This results in, besides a
general shift of the recoil peak to lower energies, an extra

broadening of the measured distribution, in addition to the
broadening caused by other statistical factors that affect the
resolution of the counter. Such behaviour has also been reported
recently for high pressure proportional counters used for photon
detection [10].

The function describing initial recombination is expected to
vary slowly with electric field strength [33]. With increasing high
voltages, less influence should be expected on the final pulse
height, thereby reversing the self-induced space charge effect
influence. According to the data presented in Fig. 7, no systematic
trend was observed concerning differences between the measured
and expected recoil peak positions when the operating voltage
was increased from 1500 to 1870 V, partially because of the
uncertainties involved. The mean values of Δp for different
operating voltages and recoil nucleus energies > 1.9 MeV have
standard deviations of 3–5%. Therefore, the two competing effects
driven by electric field strength cannot account for more than
about 5% of the Δp differences observed.

The other factor that affects initial recombination probability is
the local ion density, which depends on the electronic
stopping power of the gas mixture for each type of particle. The
two mechanisms by which protons and alpha particles are
slowed down in a medium are electronic and nuclear interac-
tions. Nuclear stopping power is important only for low
energies, e.g., around 1% for 20 keV protons or 150 keV alphas
in water [35]. The stopping power, S = dE/dx, of the counter gas
mixture for protons, tritons and 3He nuclei was calculated using
SRIM Monte Carlo code [18]. Computation was performed for
several energies ranging from 0.5 to 10 MeV, with a standard
trajectory bin of 20 μm. The calculated stopping power S(r) as a
function of 3He nuclei path length, r, was fitted with the
following function:

\[ S(r) = c_1 \ln \left[ c_2 \left( \frac{d_1}{r} + \frac{d_2}{r} \right) + c_3 \exp \left( \frac{d_1}{r} + \frac{d_2}{r} \right) \right] \]

where \( c_1, \ldots, c_6, d_1, d_2 \) and \( d_3 \) are the fitting constants. Eq. (5)
was derived empirically for the description of the Bragg-curve
shapes. Details regarding the fitting quality are presented in
Appendix A.

According to the Bragg curve, charged particles lose energy at
higher rates near the end of their trajectories. As the stopping
power is increased, the number of ion pairs produced along a
particle’s path per unit length is increased as well, thereby
enhancing the initial recombination probability as a result of an
increasing ion pair density. Therefore, the recombination influ-
ence on pulse height should depend on the fraction of energy
deposited at a higher stopping power. If we consider an arbitrary
value of stopping power, \( S_0 \), the proportion of energy loss at a
stopping power greater than \( S_0 \), ΔE/E0, to the total energy loss of
the charged particle, E0, is given by the equation:

\[ \frac{\Delta E}{E_0} = \frac{\int_{r_1}^{r_2} m(r) dr}{E_0} \]

where the limits of the integral, \( r_1 \) and \( r_2 \), are the solution(s) of
Eq. (5) for \( S(r) = S_0 \).

The ratios ΔE/E0 are calculated for initial energies E0 in the
range from 0.5 to 10 MeV and for stopping power values \( S_0 \) ranging
from 100 eV/μm to the maximum stopping power observed of
about 810 eV/μm. Results are presented in Fig. 8. In the same
figure, the experimentally measured ratio of Δp/E0 is plotted. The
experimental curve has a similar shape as the calculated curves for stopping powers in the region of 400–500 eV/\(\mu m\).

A more detailed analysis can be performed by examining the ratio of \(\Delta p/\Delta E_i\) as a function of \(^3\)He ion energy. This ratio is plotted for various stopping power values in Fig. 9. The mean values of \(\Delta p/\Delta E_i\) ratios for different stopping power thresholds, \(S_c\), are presented in Table 4. According to these data, the energy equivalent of the pulse height loss, \(\Delta p\), is a constant fraction of the energy, \(\Delta E_i\), deposited at a stopping power higher than about 450 eV/\(\mu m\).

As already mentioned, the recombination probability is a function of the local ion pair density, which depends on the stopping power \(S(E)\). Similar charged particles with different initial kinetic energies in the same traversing medium are slowed down roughly in the same way near the end of their trajectory where the stopping power becomes large, neglecting the energy straggling influence. Therefore, as the stopping power values become similar, the number of charge carriers that undergo recombination is essentially constant and proportional to the energy deposited at a stopping power larger than a critical value \(S_c\), above which the recombination probability becomes signifi-

![Fig. 8](image-url)

**Fig. 8.** Ratio \(\Delta E/\Delta E_0\) of the fraction of \(^3\)He energy loss with stopping power greater than the indicated values (in eV/\(\mu m\)) as a function of initial energy \(E_0\) of recoiling \(^3\)He nucleus (thin lines with small dots, left axis). Curves are calculated in steps of 25 eV/\(\mu m\). The thick line with error bars presents the experimentally determined \(\Delta p/\Delta E_0\), as a function of initial energy \(E_0\) (right axis).

![Fig. 9](image-url)

**Fig. 9.** Ratio of the difference between expected and measured recoil peak position (\(\Delta p\)) to the fraction of energy deposited (\(\Delta E\)) with stopping power larger than the values indicated in the legend, as a function of the initial \(^3\)He recoil nucleus energy. The error bars are presented only in one of the curves for presentation clarity.

![Fig. 10](image-url)

**Fig. 10.** Sketch of Bragg curve where the dark area, marked with \(E_0\), indicates the part of particle's path where recombination effects become significant.

![Fig. 11](image-url)

**Fig. 11.** Stopping power of \(^3\)He nuclei in the counter gas mixture as calculated with SRIM code and fitted with equation (9), \(R^2=99.986\%\).

<table>
<thead>
<tr>
<th>Energy (MeV)</th>
<th>(\Delta p/\Delta E_i\pm1\sigma) [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>&gt; 500 eV/(\mu m)</td>
<td>&gt; 475 eV/(\mu m)</td>
</tr>
<tr>
<td>Average</td>
<td>38.1</td>
</tr>
<tr>
<td>St. Deviation</td>
<td>4.4</td>
</tr>
</tbody>
</table>
The density normalised thermal electron mobility in Ar is $10^{10}/eV/cm^2/Vs$, while in Kr it is about eight times less, at $1.3 \times 10^{7}/eV/cm^2/Vs$ [36]. The counter used in the present study differs from the counter used in the present study in its geometrical characteristics and its quench gas (CO$_2$) concentration, which is 1% instead of 2% here. An increase of the CO$_2$ concentration in mixtures with Kr reduces electron mobility, e.g. increase in CO$_2$ concentration from 0.5% to 1% results in about a two-fold reduction in electron mobility [36]. Thus, the almost double deviation of the expected-to-measured recoil peak positions observed in the present study might be attributed to the reduced electron mobility due to an increased CO$_2$ concentration.

### 3.4. Energy calibration equation for the recoiling $^3$He nuclei

The pulse height produced by recoiling $^3$He nuclei is proportional to the charge carriers that escape recombination:

$$Q = Q_1 + Q_2(1 - R),$$

where $Q_1$ and $Q_2$ is the total charge initially formed by the $^3$He recoiling nucleus, $Q_2$ the charge fraction that is influenced by the recombination effect and $R$ is the recombination coefficient. The charge $Q_2$ is produced from the energy fraction that is deposited with a stopping power less than $S_0$, the limit above which recombination effect starts to be significant. This fraction corresponds to the total amounts of energy $E_1$ plus $E_3$ of the Bragg curve in Fig. 10, which can be calculated by integrating the Bragg curve within the appropriate limits

$$E_1 = \int_0^{E_0} S(E)dr = \int_{E_0}^{E} \frac{dr}{dE}dE,$$  

(8)

where the limits of integration are $E_0$, the initial charged particle energy, and $E$, the energy of which corresponds to a stopping power equal to $S_0$, $S(E_0) = S_0$.

The stopping power function of alpha particles with energy $<10$ MeV can be expressed by [35]

$$S(E) = (x_3/E)\ln(1 + x_4/E + x_5E)$$

(9)

The range $r_0$ of any particle with an initial energy $E_0$ is approximately described by the Bragg-Kleeman rule

$$r_0 = aE_0^{1.5}$$

(10)

For alpha particles (also known as the Geiger rule) $p = 1.5$ which is valid for energies above about 1–2 MeV. When an alpha particle has travelled a distance $r$, its energy has decreased to $E(r)$, which is sufficient to cover the remaining $r_0-r$ distance

$$r_0 - r = aE(r)^{1.5}$$

and by differentiation

$$\frac{dr}{dE} = -1.5aE^{0.5}.$$  

(11)

The integral $E_1$ can be calculated as follows:

$$E_1 = \int_{E_0}^{E} S(E)\frac{dr}{dE}dE = \int_{E_0}^{E} (x_3/E)\ln(1 + x_4/E + x_5E)(-1.5aE^{0.5})dE$$

(12)

The stopping power of $^3$He nuclei in the counter gas mixture, $S(E)$, is computed with SRIM code, and constants $x_3$, $x_4$ and $x_5$ are calculated as fitting constants of Eq. (9) (see Fig. 11). Similarly, constant $\alpha$ of the range-energy relationship is calculated to be $(3.115 \pm 0.010)10^{-5}$, where the energy is expressed in keV, and the range is expressed in mm (see Fig. 2).

According to the results presented in Section 3.3, the stopping power above which the recombination effect become significant is $S_0 = 450 \text{ eV/\mu m}$, which according to Eq. (9) corresponds to $E_0 = 2.6$ MeV. Therefore, when the initial energy $E_0$ is less than 2.6 MeV, the integral $E_1$ in Fig. 10 becomes 0 and the charged

![Fig. 12. Calculated reduction (thick line) of recoil $^3$He nucleus energy with the actually measured reduction (points) as a function of recoil energy. The dark area confines the $\pm 1\sigma$ range of calculated values.](image-url)
particle is losing all its energy (except the part of the integral marked as \( E_3 \)), traversing the counter gas with a stopping power larger than 450 eV/\( \mu \)m. Integral \( E_3 \) was calculated with numerical integration of \( S(r) \) and appears to be fairly small and constant, at 0.1 \( \pm \) 0.01 MeV for initial \(^3\)He ion energies below 7 MeV.

Finally, the recombination defect to recoiling \(^3\)He nuclei pulse height, \( \Delta p \), can be expressed by the following:

\[
\Delta p = \begin{cases} 
R(E_0 - E_1 - E_3) & E_0 > E_c \\
R(E_0 - E_1) & E_0 \leq E_c 
\end{cases}
\]

The analytical expression of \( \Delta p \) is presented in Appendix B.

A comparison of the reduction in the energy of recoil nucleus as it is calculated using Eq. (B.2) and as it is measured is presented in Fig. 12 as a function of \(^3\)He nucleus energy. The presented differences include the influences of both resolution and recombination effects. Experimental points are in the region confined between \( \pm 1 \sigma \) of the theoretically produced curve. Uncertainty was calculated with the propagation of uncertainties in fitting constants and the recombination coefficient.

4. Conclusions

The experimental results show that the relation of the energy of recoil nuclei vs. pulse height is not a linear function, in contrast to the result observed for the full energy peak. Furthermore, when the recoil nuclei pulse height is converted to energy via recombination energy calibration of the full energy peak, a significantly lower energy than the one expected from kinematics is calculated. The observed variations of the differences between measured and expected recoil nuclei energy with energy and the counter gas mixture. Comparison of the total energy loss to ionization and R-squared of the fitting. The energy calibration correction, i.e., the recombination effect. The combined effect of space charge and initial recombination variation with electric field strength cannot account for more than 5% of the measured differences when the operating voltage varies by 20%.

The experimentally determined loss in recoil nuclei energy equivalent to pulse-height loss is a constant fraction of particle energy deposited at a high-stopping power. For the commercially available He-3 counter used in the present study, the fraction of energy deposited at a stopping power larger than 450 eV/\( \mu \)m undergoes a recombination defect of 33.5 \( \pm \) 1.3%.

Regarding spectroscopy applications, in He-3 counters, gas mixtures with Kr as a high stopping power gas were abandoned due to their higher sensitivity for \( \gamma \)-rays, as compared to Ar-based mixtures [37]. The fact that electron mobility is much less important in Kr than in Ar results in increased recombination effects, which move the recoil peak backward towards a lower energy. In this way, a larger area of the spectrum is clear from counts originating from recoil events, which is an advantage for neutron spectroscopy applications.

The energy calibration correction, i.e., the recombination defect, of recoil events in a spectroscopy application can be achieved by Eq. (13). The constants of this equation can be theoretically calculated as constants of the range-energy and stopping power-energy relationships of the recoil nucleus, while the recombination coefficient, \( R \), should be determined experimentally.

Acknowledgments

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Appendix A

The stopping power, \( S(r) \), was calculated with SRIM/TRIM code as a function of the \(^3\)He nuclei path length and was fitted with Equation (5). The contribution of each term of the function is presented in Fig. 13, in the same sequence as it is written in the equation.

Fitting is performed from \( r=0 \) up to the point where range straggling influence starts, typically at values of about \(< 10 \) eV/\( \mu \)m of the stopping power. Comparison of the total energy loss to ionization as it is computed by SRIM code with the numerically integrated equation (5) is presented in Table 5. Monte Carlo calculated stopping power values (Bragg functions) and the corresponding fitted functions are presented in Fig. 14.

![Fig. 13. The contribution of each term of Eq. (5) to the total calculated stopping power (top line) of the counter gas mixture.](image)

<table>
<thead>
<tr>
<th>Initial energy (keV)</th>
<th>Total energy loss to ionization</th>
<th>( R^2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( 0.50 )</td>
<td>0.493</td>
<td>0.500</td>
</tr>
<tr>
<td>( 0.75 )</td>
<td>0.742</td>
<td>0.749</td>
</tr>
<tr>
<td>( 1.00 )</td>
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</tr>
<tr>
<td>( 10.00 )</td>
<td>9.989</td>
<td>9.970</td>
</tr>
</tbody>
</table>
Appendix B

The energy $E_1$, deposited by $^3$He recoiling nucleus with initial energy $E_0$, from the beginning of its track up to the point that kinetic energy is reduced to $E_C$ (see Fig. 10), is described by Eq. (12). The integral $E_1$ was calculated with the program language of Mathematica [38] taking into account the value of the hypergeometric function $F(0.5,0.5,1.5;x^2) = \sin^{-1}(x)/x$.

$$E_1 = 3 \frac{\sqrt{2}x_3}{\sqrt{2}x_5} + \left[ \frac{\sqrt{1-d} \sin^{-1} \left( \frac{1-d}{1+d+2x_3E} \right) }{1} \right]_{E_C}^{E_0} \left[ \frac{\sqrt{1+d} \sin^{-1} \left( \frac{1+d}{1+d+2x_3E} \right) }{1+d+2x_3E} \right]_{E_0} \left( 2 - \ln \left( 1 + \frac{2x_5}{E_C} \right) - z_1 \right)_{E_0} - \left( \frac{2x_5}{E_C} \ln \left( 1 + \frac{2x_5}{E_C} \right) - 2 \right)_{E_0}$$

$$d = \sqrt{1 - 4x_4x_5}$$

Finally, the pulse height reduction, $\Delta p$, due to the recombination effect can be calculated using Eqs. (13) and (B.1) as it follows:

$$\Delta p = \left\{ \begin{array}{ll}
\sqrt{1-d} \sin^{-1} \left( \frac{1-d}{1+d+2x_3E} \right) & \text{if } E_0 \geq E_C \\
\sqrt{1+d} \sin^{-1} \left( \frac{1+d}{1+d+2x_3E} \right) \left( 2 - \ln \left( 1 + \frac{2x_5}{E_C} \right) - z_1 \right)_{E_0} - \left( \frac{2x_5}{E_C} \ln \left( 1 + \frac{2x_5}{E_C} \right) - 2 \right)_{E_0} & \text{if } E_0 < E_C
\end{array} \right.$$