Estimation of the ionisation quenching factor in gases from W-value measurements

I. Katsioulas,1,∗ P. Knights,1,2 and K. Nikolopoulos1
1 School of Physics and Astronomy, University of Birmingham, B15 2TT, United Kingdom
2 IRFU, CEA, Université Paris-Saclay, F-91191 Gif-sur-Yvette, France

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The effect of ionisation quenching for ions is critical for experiments relying on the measurement of low energy recoils, such as direct Dark Matter searches. We present ionisation quenching factor estimates over a range of energies for protons, α-particles, and heavier ions in H2, CH4, N2, Ar, CO2, and C2H6 gases, estimated from the respective reference W-value measurements. The resulting ionisation quenching factors are compared with predictions from SRIM.

Keywords: ionisation quenching factor, mean ionisation energy, W-value, nuclear recoil, dark matter

I. INTRODUCTION

Direct searches for Dark Matter (DM) particles employ a range of detector technologies, including liquid noble gases [1-4], crystals [5], and semiconductors [6]. Gaseous detectors are used to expand DM searches below the Lee-Weinberg mass bound of about 2 GeV [7], through the use of light-nuclei gases, and for directional DM searches [8-10].

The NEWS-G experiment, using a spherical proportional counter filled with a neon-methane gas mixture [10], set the most stringent exclusion limit at the time in the DM-nucleon spin-independent interaction cross section for a 500 MeV mass DM candidate. The TREX experiment [11], using a gaseous Time Projection Chamber (TPC) with argon-isobutane and neon-isobutane gas mixtures, is projected to achieve competitive sensitivities in the DM candidate mass region below a few GeV. Directional DM searches such as DRIFT [12], CYGNUS [13], MIMAC [8], and NEWAGE [14] employ low-pressure (below a few hundred millibar) TPCs with the ability to infer the ionisation track direction. Additionally, many of these detectors utilise gas mixtures containing odd-proton, e.g. 19F, or odd-neutron isotopes, e.g. 3He, to achieve world-leading sensitivity to spin-dependent DM-nucleon interactions.

Knowledge of the expected amount of ionisation and scintillation induced by nuclear recoils is important for these searches, which rely on the detection and measurement of nuclear recoils with energy from a few eV to hundreds of keV, induced by coherent elastic scattering of DM particles on nuclei. As a result, several investigations of the corresponding ionisation quenching effects have been performed [15-21].

Low energy nuclear recoils dissipate energy in a medium through inelastic Coulomb interactions with atomic electrons, referred to as electronic energy losses, and elastic scattering in the screened electric field of the nuclei, referred to as nuclear energy losses. Electronic energy losses dominate for fast ions, while nuclear energy losses are increasingly important for decreasing ion kinetic energy \( E_R \), and become dominant for ion velocities smaller than the electron orbital velocity [22]. The produced secondary recoil atoms and electrons may also undergo scattering, further transferring energy to other particles.

The ionisation quenching factor, \( q_f \), concept was introduced by Lindhard et al. [23], and remains a field of intense study [24]. The electronic and nuclear stopping power are estimated assuming they are uncorrelated. Integrating the electronic and nuclear energy losses until the ion stops provides the energies \( v \) and \( \eta \) given to atomic motion and electrons, respectively. The maximum available energy for ionization and scintillation is \( \eta \). The Lindhard quenching factor is defined as \( \eta/E_R \), the ratio of energy given to electrons over the ion kinetic energy.

SRIM [25] (Stopping and Range of Ions in Matter) is a computational tool primarily used for the estimation of ion ranges in matter. TRIM (TRansport of Ions in Matter), a module of SRIM, is often used to estimate the ionisation quenching factor in materials as it provides estimates for the fraction of ion kinetic energy dissipated in electronic and nuclear energy losses, including contributions of secondary recoils. In Fig. 1, SRIM simulations of the spatial profile of energy dissipation for light and heavy ions in a medium are shown. Heavier ions, like Ar+ shown in Fig. 1b), are subject to a higher rate of elastic scattering compared to lighter ions, such as protons shown in Fig. 1a), of the same kinetic energy. SRIM estimates of the ionisation quenching factor of heavy ions in their own gas are presented in Fig. 2.

In the literature, multiple definitions of the ionisation quenching factor may be found depending on the application:

1. the fraction of the ion kinetic energy that is dissipated in a medium in the form of ionisa-
Figure 1: Tracks of (a) 10 keV $\text{Ar}^+$ and (b) 10 keV protons in Ar gas simulated using SRIM [25].

Figure 2: Quenching factor for ions in their own gas estimated using SRIM [25].

takes the form,

$$ q_f = E_d/E_R , $$

where $E_d$ is the amount of electronic energy losses “visible” to a detector [8, 9].

For these measurements, ions with precisely known kinetic energy are required, along with the capability to measure small energy deposits. As a result, although the ionisation quenching factor is critical for the modeling of the detector response to nuclear recoils — and, thus, for the sensitivity to discover DM — relevant measurements are scarce for low energy ions in gases. Santos et al. [26], investigated the quenching of He$^+$ in He/C$_4$H$_{10}$ gas mixtures using a table-top low energy ion-electron accelerator [27]. The measurements were performed with a Micromegas proportional counter [28]. The ionisation quenching factor was estimated as the ratio of the energy measured in the detector for ions to their initial total kinetic energy. Beyond measurements, prompted by the requirements of the DM community, Hitachi estimated the dependence of the quenching factor on energy in liquid noble gas detectors [29], and attempted to expand the work of Lindhard et al. to atomic gases and molecular gases, and their binary mixtures [30].

In this article, ionisation quenching factors are estimated for a range of gaseous targets by exploiting available measurements of the mean ionisation energy $W$ for electrons and ion species. Measurements of $W$ have been the topic of intense investigations for over a century [31, 32], with improved experimental methods and increased precision over time.

In the above, the first definition coincides with that of Lindhard. Moreover, the term “visible” refers to energy dissipated in a detectable form for the specific detection system, i.e. ionisation for detectors measuring charge.

For the experimental determination of the ionisation quenching factor in a material, the amount of ionisation induced by an ion species has to be measured as a function of its kinetic energy and compared to the corresponding ionisation by electrons of the same initial energy. In this case the ionisation quenching factor
II. THEORETICAL BACKGROUND

In the following, a brief description of the theoretical investigations on $W$ is provided and the connection to the ionisation quenching factor is discussed.

A. Description of the W-value

The average energy expended by ionising radiation to produce an electron-ion pair inside gaseous and liquid media, or an electron-hole pair in solids, is denoted by $W$. When all of the kinetic energy $E$ of the ionising particle is deposited in a medium the $W$-value is:

$$W = E/N_i$$  \hspace{1cm} (2)

where $N_i$ is the mean number of electron-ion (or electron-hole) pairs created. It is assumed that any secondary particles produced during the energy loss process, e.g. $\delta$-electrons, bremsstrahlung photons, or electron and nuclear recoils, also dissipate their energy in the medium and the produced electron-ion pairs are included in $N_i$ [31].

The $W$-value contains all the required information for converting the kinetic energy dissipated by ionising radiation in a medium to electric charge, and vice versa. The magnitude of the $W$-value is the outcome of the competition between ionising and non-ionising processes, such as excitation of atomic electrons and molecular degrees of freedom, dissociation processes (neutral fragmentation) in molecular gases, and production of neutral atomic recoils. As a result the $W$-value also depends on the interacting particle species and, in general, tends to a constant value for increasing energy.

1. The $W$-value in pure gases

An intuitive explanation of the $W$-value magnitude for the simplest case of fast electrons in pure gases was provided by Platzman [33], based on energy balance arguments. The kinetic energy is apportioned in three parts: a) production of electron-ion pairs; b) production of discrete excited states or neutral dissociation fragments; and c) production of sub-excitation electrons, with kinetic energy below the lowest electronic excitation energy of the medium.

This relation is written as:

$$E = N_i \bar{E}_i + N_{ex} \bar{E}_{ex} + N_i \epsilon$$  \hspace{1cm} (3)

where $\bar{E}_i$ is the mean energy transfer per electron-ion pair, $N_{ex}$ is the mean number of excited states produced, $\bar{E}_{ex}$ is the mean energy transfer per excited state, and $\epsilon$ is the mean kinetic energy carried by sub-excitation electrons. Dividing by $N_i$ results in:

$$W = \bar{E}_i + \frac{N_{ex}}{N_i} \bar{E}_{ex} + \epsilon$$  \hspace{1cm} (4)

The $W$-value is more naturally expressed relative to the first ionisation threshold $I$ of the gas:

$$\frac{W}{I} = \frac{\bar{E}_i}{I} + \frac{N_{ex}}{N_i} \frac{\bar{E}_{ex}}{I} + \frac{\epsilon}{I}$$  \hspace{1cm} (5)

Since every term on the right-hand side of Eq. 5 depends on the energy of the ionising particle, the $W$-value also exhibits an energy dependence, which becomes weaker for $E \gg I$. Platzman uses helium as an example to demonstrate the completeness of the approach, where the magnitude of each term for high energies is:

$$1.71 \approx 1.06 + 0.85 \cdot 0.40 + 0.31$$  \hspace{1cm} (6)

Each term on the right-hand side of Eq. 5 has been derived from methods other than the absolute measurement of the $W$-value, which is shown on the left-hand side. The magnitude of these terms provides a sense of the apportionment of the ionising electron's kinetic energy. The value of 1.06 for the ratio $\bar{E}_i/I$ is greater than one because of the production of excited and multiply charged ions. This effect is present in all noble gases and is further enhanced in molecules thanks to the larger number of available degrees of freedom. The term $\bar{E}_{ex}/I$ is in general less than one but close to it because the energy levels of most excited states lie below or close to $I$. The ratio $N_{ex}/N_i$ depends strongly on the atomic and molecular dynamics, taking values of approximately 0.5, 1, and 2 for closed-shell atoms, molecules, and free radicals, respectively [33]. Finally, the ratio $\epsilon/I$ is smaller than unity because by definition sub-excitation electrons have energy lower than $I$, and this is the part dissipated as heat. The value of approximately 0.3 applies to most noble gases but is less than that for molecules because molecular ions retain more energy than atomic ions [34]. Overall, high energy ionising particles have a $W/I$ ratio of 1.7–1.8 in noble gases and 2.1–2.5 in molecular gases.

2. $W$-value dependence on particle species and energy

The $W$-value depends on the ionising particle species and energy. For electrons with energy lower than a few keV, the cross section for electronic excitation increases with respect to that for ionisation as the electron energy decreases [34]. The electron W-value as a function of kinetic energy is, typically, described as:

$$W(E) = \frac{W_o}{1 - U/E}$$  \hspace{1cm} (7)
where $W_a$ is the asymptotic $W$-value for $E \gg I$ and $U$ is a constant close to the average energy of sub-excitation electrons [35]. This relation is in agreement with measurements in a number of gases including nitrogen, methane, and propane, down to low energies [36–38], although some measurements suggest a stronger dependence [39]. This is discussed in detail in Refs. [31, 32].

The energy dependence of the $W$-value for charged hadrons, such as protons, $\alpha$-particles, mesons, and heavy ions is more complicated as it involves a larger number of reactions compared to electrons. During the energy loss process, in addition to secondary electron generation, electron capture and loss cycles are taking place. Moreover, collisions of ions with atoms and molecules result in energy transfers that may lead to further ionisation when ion velocities become comparable with electron orbital velocities. When ions carry electrons, as expected for slow ions, collisions with atoms and molecules give a number of products, including energetic ions and molecular fragments, that may result in further ionisation. A theory description that reliably predicts the $W$-value is not yet available and cross sections for the individual process are under investigation [22].

The energy balance formulation of Platzman can be extended for ions by adding the contribution of neutral recoils created during the cascade [23]:

$$E = N_i \bar{E}_i + N_{ex} \bar{E}_{ex} + N_{\epsilon} \bar{\epsilon} + N_{nr} \bar{E}_{nr}$$

(8)

Dividing by $N_i$ gives:

$$W_{ion} = \bar{E}_i + \frac{N_{ex}}{N_i} \bar{E}_{ex} + \bar{\epsilon} + \frac{N_{nr}}{N_i} \bar{E}_{nr}$$

(9)

where $N_{nr}$ is the number of neutral recoils produced and $E_{nr}$ is their average kinetic energy. Other terms keep their original meaning as in Eq. 3. The $N_i \bar{E}_i$ and $N_{ex} \bar{E}_{ex}$ terms include ionisation and excitation, respectively, induced by the secondary ions in addition to that induced by the secondary electrons.

### B. Relation of the $W$-value and the ionisation quenching factor

The $W$-values for different ionising radiations encapsulates all necessary information to estimate differences in the amount of induced ionisation. The quenching factor parameterises these differences. In Section III, three definitions of the quenching factor were outlined. The first quenching factor definition is experimentally accessible only in the case where a detector is able to measure the total amount of energy deposited by an ion in electronic energy losses, ionisation and excitations, as a fraction of its total kinetic energy. The two other definitions can be measured with a detector relying on ionisation.

For a particle of energy $E$ depositing all of its energy in the detector, there will be $N_i = E/W$ primary electrons produced. The detector will record a signal $S$ proportional to $N_i$ where the constant of proportionality $G$ includes the response of the read-out electronics and any electron multiplication. The detector can be calibrated with electrons of known kinetic energy in order to relate the signal recorded with the corresponding energy deposited in the detector, $S \rightarrow E$. In the case of electrons, the corresponding signal for a given energy deposited will be $S_e = G \cdot N_i^e = G \cdot E/W_e(E)$. For ions of the same kinetic energy the corresponding signal will be $S_i = G \cdot N_i^i = G \cdot E/W_i$, which, through the calibration, would be interpreted as an electron equivalent energy $E_{ee} = N_i^i \cdot W_e(E)$. $E_{ee}$ is the energy visible to the detector, thus, from Eq. 1, the quenching factor can be written as:

$$q_f(E) = \frac{E_{ee}}{E} = \frac{N_i^i \cdot W_e(E)}{E} = \frac{W_e(E)}{W_i(E)}$$

(10)

When the kinetic energy is over a few keV, far away from the energy region where the $W$-value for electrons is energy dependent, the quenching factor can be derived from the ratio of the asymptotic $W$-value for electrons $W_a$ over the $W$-value for ions $W_i$ in the same medium. This is the case in the second definition, and is equivalent to Eq. 10 when $W_e \rightarrow W_a$.

In the third definition of the quenching factor, the energy dependence of $W_e$ is retained, and thus accounts for the effective quenching of electrons at energies below a few keV.

### III. QUENCHING FACTOR ESTIMATION

The estimation of the quenching factor, using Eq. 10 for different ion species relies on measurements of the $W$-values for ions and electrons, the latter used to obtain the asymptotic $W$-values for electrons. In this work, the measurements were selected based on two community reports produced by ICRU (1971) and IAEA (1995) international commissions, which surveyed multiple studies of $W$-values. The gases studied are $H_2$, $CH_4$, $N_2$, Ar, CO$_2$, C$_2$H$_6$ and $W$-values for electrons, protons, $\alpha$-particles, and constituent ions are included. These gases are frequently used as counting gases and quenchers in proportional counters [42], and as components of tissue-equivalent gases, used to emulate energy deposition in the human body. Furthermore, these are of particular interest as targets for rare event searches with gaseous detectors.
A. W-value measurements for electrons and ions in various gases

The electron measurements by Combecher et al. \cite{37}, Waibel and Grosswelt \cite{36, 40, 41} are presented in Fig. 3 and demonstrate the discussed energy dependence of the W-value. CO₂ data from Ref. \cite{37} were not used due to their discrepancy with other published measurements and calculations \cite{32}. The data for each gas are fitted with Eq. 7 using \( W_a \) and \( U \) as free parameters to estimate the asymptotic W-value for electrons in each gas. Only measurements for electron kinetic energies substantially larger than the first ionisation threshold are included in the fit. The measurement uncertainties are dominated by systematic effects, which are assumed to be fully correlated across the considered energy range. These results are summarised in Table I. The resulting asymptotic W-values appear higher than those suggested by ICRU \cite{31} for high energy electrons. The recommended ICRU values are an arithmetic average of measurements published by a number of authors before 1971, using different energies and methodologies, and the assigned uncertainties were chosen to embrace most of the available results. The results presented here are based on the latest measurements recommended by the IAEA report \cite{32}. The difference between the derived asymptotic W-value and the corresponding ICRU recommended value is assigned as a systematic uncertainty in the quenching factor estimate.

The W-values for protons and heavier ions in the aforementioned gases were taken from various sources. Specifically, Chemtob et al. \cite{43} and Phipps et al. \cite{44} were used for the W-values of protons, \( \text{H}_2^+ \), \( \text{H}_3^+ \), \( \text{He}^+ \) and \( \text{Ar}^+ \) in Ar gas; Huber et al. \cite{45}, Boring et al. \cite{46}, Nguyen et al. \cite{47}, Waibel et al. \cite{48} for protons, \( \text{H}_3^+ \),

Table I: W-values for electrons in various gases. The asymptotic W-values are derived from the fits shown in Fig. 3.

| Gas        | W [eV]        | ICRU Asymptotic |
|------------|---------------|-----------------|
| H₂         | 36.5±0.7      | 38.0±0.8        |
| CH₄        | 27.3±0.6      | 27.90±0.01      |
| N₂         | 34.8±0.7      | 34.91±0.17      |
| Ar         | 26.4±0.5      | 28.5±0.6        |
| CO₂        | 33.0±0.7      | 33.02±0.12      |
| C₃H₈       | 24.0±0.5      | 26.4±0.5        |

Figure 3: W-values versus electron kinetic energy for the gases studied. Data from Ref. \cite{37} were used for H₂, Ar and C₃H₈. For CH₄, data from Refs \cite{37} and \cite{36} are combined. The data from Refs \cite{37} and \cite{40} were combined for N₂. Only the data from Ref. \cite{41} was used for CO₂. The curves are fitted using Eq. 7 to estimate the asymptotic W-value (\( W_a \)) for each gas.
He⁺, N⁺ and N₂⁺ in N₂ gas; Huber et al. [45], Nguyen et al [47] and Waibel et al. [48] for protons, H₂⁺, He⁺, C⁺ and O⁺ in CO₂ gas; Huber et al. [45], Nguyen et al [47] and Waibel et al. [48] for protons, H₂⁺, He⁺, and C⁺ in CH₄ gas; Willems et al. [49], and Posny et al [50] for protons, He⁺ and C⁺ in C₃H₈ gas; Grosswelt et al [51] for protons in H₂ gas. The W-value for electrons and ions in the gases studied are shown in Fig. 4. In the case of molecular ions the energy mentioned is per atom. In the case of proton and H₂ curves, it is observed that they, practically, coincide. This is explained by the molecules in the ion beam dissociating in the initial collisions with the gas constituents [45].

In Fig. 5 the difference between the measured electron W-value to the corresponding asymptotic value, normalised to the latter, is shown.

B. Results

The quenching factor for each ion species in a gas was estimated by dividing W_a in that gas by the W-value of an ion species, as per Eq. [10]. The results are displayed in Fig. 6. The uncertainties were calculated using the uncertainty in the estimation of the W_a for electrons and the uncertainty for the W-values for ions provided in the respective publications. For reference, the estimated W-values for each ion species are presented along with the W-value provided by SRIM.
Figure 6: Ionisation quenching factor versus ion energy for several gases. The quenching factors estimated by SRIM are also provided for comparison. \( \sigma_{W_{\text{sys}}} \) is the systematic uncertainty on \( W_{\alpha} \), which is taken to be the difference between the fitted and ICRU recommended \( W_{\alpha} \).
IV. DISCUSSION

The quenching factor for gases, like the W-value, depends on the incoming particle species and kinetic energy. It is independent of the gas density, up to the formation of electronic bands at elevated densities. This allows for W-values and quenching factors for gases to be estimated with specifically designed experiments, and then be applied to other experimental configurations in a wide range of densities. The method presented is based on this principle, utilising dedicated W-value measurements to estimate the quenching factor in pure gases.

A. Energy dependence of the quenching factor

The results display the expected energy dependence of the quenching factor which is a consequence of the energy dependence of the W-value for ions. It is noted that for electron kinetic energies lower than a few keV (Sec.IIA2), their W-value is also energy dependent, as shown in Fig. 4 introducing an effective electron ionisation quenching. This additional reduction in ionisation has to be taken into account when converting between nuclear recoil energies and electron equivalent energies. In order to accurately determine the electron equivalent energy of the ion, the energy dependent electron W-value should be taken into account. The difference between $W_A$ and the measured W-value for electrons in various gases is shown in Fig. 6. At an energy of 500 eV there is approximately a 3% difference between the two, which increases with decreasing energy.

B. Proton quenching factor greater than unity

The W-value for high energy protons, $\mathcal{O}$(MeV) kinetic energy, in any gas is in general larger than or equal to the one for electrons in the same gas. However, in Fig. 6 the quenching factor for protons in H$_2$, Ar, N$_2$, and CO$_2$ is found to be greater than unity in some energies. This counter intuitive effect is a result of the minimum in the proton W-value curve which is observed in those gases, and can be seen in Fig. 4. This is attributed to charge exchange reactions involving H and the molecule (atom) of the gas. These reactions are dominant in the energy range around the minimum. Although commonly referred to as the ionisation quenching factor, a more appropriate term would be ‘ionisation yield’, similar to the term ‘scintillation yield’ used in scintillation detectors.

As an example the case of N$_2$ is discussed, while a detailed discussion can be found in Ref. [53]. The charge exchange runs in a cycle which starts with the proton capturing an electron resulting in the production of neutral H and a N$_2^+$,

$$H^+ + N_2 \rightarrow H + N_2^+$$  \hspace{1cm} (11)

The H atom can further undergo one of the following reactions:

$$H + N_2 \rightarrow H + N_2^+$$  \hspace{1cm} (12)
$$H + N_2 \rightarrow H + N_2$$  \hspace{1cm} (13)
$$H + N_2 \rightarrow H + N_2^+ + e^-$$  \hspace{1cm} (14)

The H atom can go through the N$_2$ excitation reaction in Eq. 12 or the elastic scattering reaction in Eq. 13 before ending the cycle by the ionisation reaction in Eq. 14. This cycle converts an initially pure proton beam into a mixture of H$^+$ and H, where the latter dominates for energies above ~10 keV. At low energies, the channels of N$_2$ ionisation and excitation compete almost equally. The addition of secondary electrons, arising primarily from the charge exchange reactions and the large number of ions produced, results in an increased ionisation yield. Above about 100 keV the charge exchange channel shuts off and results in a rise of the W-value, which is lowered again near the MeV-scale due to an increase of proton induced ionisation.

C. Comparison with predictions from SRIM

In Fig. 6 the estimated quenching factor curves are compared with quenching factor from SRIM calculations. The level of agreement with SRIM varies and depends on ion type, energy range and gas. SRIM predictions seem to be in reasonable agreement with results for N$_2^+$ in N$_2$, C$^+$ and O$^+$ in CO$_2$ over 10 keV. SRIM predictions are also comparable to the results for He$^+$ in N$_2$, CO$_2$, Ar, as well as Ar$^+$ in Ar over 10 keV. Below 10 keV SRIM predictions are consistently higher than the estimated quenching factors. This effect may be attributed to the overestimation of the electronic stopping power in low energies by SRIM.

Two further points of disagreement between SRIM and the experimental results involve protons in all gases, except organic molecular gases, and all ion types in organic molecular gases. In the case of protons in H$_2$, Ar, N$_2$, and CO$_2$ the discrepancy originates from the charge exchange cycle, discussed in section IVB, resulting in additional ionisation with respect to the electronic stopping power estimated by SRIM. In hydrocarbons, as shown in Fig. 7 the order of quenching factors is inverted with respect to what would be anticipated by molecular mass arguments. For example, as predicted also by SRIM, ions in CO$_2$ would be expected to have a
lower quenching factor than in CH₄ or C₃H₈. This behaviour has been previously reported \[30\], but its source remains unclear.

Figure 7: Quenching factor for C⁺ ions in CO₂, CH₄, and C₃H₈. The energy in CH₄ data has been increased by 3 keV for visibility.

V. SUMMARY

Ionisation quenching is a crucial effect on the measurement of low energy ions in a number of applications, including direct dark matter searches. Due to the scarcity of measurements in relevant materials and energy ranges, one often has to rely on simulations, theoretical models, or extrapolation of measurements, leading to large uncertainties.

In this article, a distinction between the several definitions of the quenching factor found in literature is made, the importance of the increased electron W-value for low kinetic energies is highlighted, and estimates of the ionisation quenching factor for protons, α-particles, and heavier ions are presented using measurements of their W-values and W-values of electrons in H₂, CH₄, Ar, N₂, CO₂, and C₃H₈ gases. The obtained quenching factor estimates are discussed in terms of ion species and kinetic energy in a given gas, along with gas-specific effects, for example additional ionisation induced by charge exchange reaction cycles. Moreover, comparisons with simulations are included, where such effects are not currently included for the calculation of the electronic stopping power.

The presented method uses data from dedicated high-precision experiments and is directly applicable to any other gas for which W-value measurements become available. Since the quenching factor is an intrinsic property of a given gas, obtained estimates may be applied to any measurement using said gas.

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