Average energy expended per ion pair in liquid argon*

M. Miyajima,† T. Takahashi, S. Konno, and T. Hamada
Institute of Physical and Chemical Research, Wako-shi, Saitama, Japan

S. Kubota
Department of Physics, St. Paul's University, Nishi-Ikebukuro, Tokyo, Japan

H. Shibamura and T. Doke
Science and Engineering Research Laboratory, Waseda University, Shinjuku, Tokyo, Japan

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The measurement of the W value, the average energy expended per ion pair, in liquid argon for internal conversion electrons emitted from ^{207}Bi, is carried out by the electron-pulse method. Comparing with the W value (26.09 eV) for α particles in a gas mixture of argon (95%) and methane (5%), the W value in liquid argon is determined to be $23.6^{+0.5}_{-0.3}$ eV. This value is clearly smaller than that in gaseous argon (26.4 eV). Such a reduction of W value in liquid argon can be explained by assuming the existence of the conduction band in liquid argon as in solid argon. Under this assumption, the W value in liquid argon is estimated by applying the energy-balance method, which was used for the rare gases by Platzman. The calculated value thus obtained is in good agreement with the experimental result. Following the same method, the W values for liquid krypton and liquid xenon are also estimated.

INTRODUCTION

In 1969, one of the authors (T.D.)[1] suggested that the W value, the average energy expended per ion pair, in the condensed state of rare gases may have a lower value than that in the gaseous state, assuming the presence of a conduction band in the condensed state. Recently, a number of experimental studies[2,3] have shown that in the liquid rare gases the negative charge carriers are electrons with a large mobility. Further, Wannier excitons, already discovered in solid xenon, are also observed in liquid xenon.[4,5] These facts may suggest the existence of the conduction band in the liquid rare gases. Therefore, it is very interesting to study the W values in the liquid rare gases. However, these values are unknown except for roughly measured values on liquid argon.

In 1965, Swan[6] obtained the W value for liquid argon of $W_i(A) = 26.0$ eV, by applying the columnar recombination theory of Jaffe to the analysis of the pulse-height distributions produced by α particles. He considered that $W_i(A)$ agreed with the W value for gaseous argon $W_g(A)$, i.e., 26.4 eV.[7,8] In 1966, Ullmaier[9] obtained $W_i(A) = 25.7 \pm 3$ eV from the measurement of the ionization current produced by x rays. His result was almost the same as $W_g(A)$. In 1969, Klassen and Schmidt[10] also measured the ionization current produced by energetic x rays and obtained $W_i(A) = 22.5 \pm 3$ eV. Their result may indicate that $W_i(A)$ is smaller than $W_g(A)$, but their error is too large to distinguish a meaningful difference between $W_i(A)$ and $W_g(A)$. Such a relatively large error in the steady current method arises from uncertainties in the estimation of the energy deposited in the detector volume by external radiation.

The aim of our studies is to investigate the ionization and excitation phenomena in the condensed state of the rare gases and their applicability to nuclear-radiation detection.

As the first step in our studies, we report here $W_i(A)$ determined relative to $W_g(A)$ by the electron-pulse method using a source of energetic conversion electrons. One of the advantages of the pulse method is in the ability to determine the energy of individual pulses without any ambiguity, and furthermore to obtain the saturation of the ionization pulse easily, since the specific ionization of the energetic electrons is relatively low.

Moreover, we describe the estimation of the W value in the condensed state of the rare gases from the energy-balance equation that was applied to the rare gases by Platzman[8], using recent information on the band structures in the solid rare gases, and we also describe the comparison of the estimated values with the experimental results.

METHOD OF DERIVATION OF W_i

In this study a gridded liquid-ionization chamber and a gridded gas-ionization chamber are used as experimental apparatus. These are shown schematically in Fig. 1 together with the block diagram of the electronic circuits. Signal pulses

from both ionization chambers are amplified by a charge sensitive preamplifier and a main amplifier and are analyzed by a multichannel pulse-height analyzer. The test pulse from a mercury relay test pulser is fed to the preamplifier input stage through a small capacitor C_{in} in order to calibrate the measuring system. Measurements are made using a conversion-electron source in the liquid chamber and a mixed α source in the gas chamber, respectively. The steps in the measurements are the following. The amplifier system is connected with the liquid chamber and the spectrum of the conversion electrons with energy E_c is measured. The saturated pulse height V_l is expressed as:

$$V_l = \frac{eE_c}{W_l} \frac{A}{C_l + C_s + (1+A)C_f} G , \tag{1}$$

where e is the electronic charge, C_s the stray capacity of the preamplifier input stage, C_l the capacity of the liquid chamber, C_f the feedback capacity of the preamplifier, A the open-loop gain of the preamplifier, and G the effective gain of the main amplifier. Furthermore, the test pulse V_t is fed to the preamplifier as the system is connected with the liquid chamber. The pulse height V_{tl} obtained at the output stage of the amplifier system, is given in the following form:

$$V_{tl} = \frac{C_{in}(C_l + C_s)V_t}{C_{in} + C_l + C_s} \frac{A}{C_l + C_s + (1+A)C_f} G . \tag{2}$$

Next, the amplifier system is connected with the gas chamber and the spectrum of the α particles with energy E_a is measured. The saturated pulse height V_g produced by the α particles in the gas chamber is obtained by changing suffixes l to g, and c to a, in Eq. (1). Similarly, the pulse height V_{tg} of the test pulse at the output of the amplifier system is obtained by replacing the suffix l by g in Eq. (2). Consequently, from Eqs. (1) and (2), W_l is written by using V_t as

$$W_l = eE_c \frac{V_{tl}}{V_l} \frac{C_{in} + C_l + C_s}{C_{in}(C_l + C_s)} \frac{1}{V_t} . \tag{3}$$

Also, W_g is written in the same manner by changing the suffixes l to g, and c to a, in Eq. (3). Then W_l can be rewritten by using W_g as

$$W_l = W_g \frac{V_g}{V_l} \frac{E_c}{E_a} \frac{V_{tl}}{V_{tg}} \left(\frac{1 + [C_{in}/(C_l + C_s)]}{1 + [C_{in}/(C_g + C_s)]} \right) . \tag{4}$$

In this equation, the value of parenthesis can be put equal to unity if $C_{in} \ll C_l + C_s$ and $C_{in} \ll C_g + C_s$. Actually, C_{in}, C_l, and C_g were measured to be 1.1 ± 0.5, 28.0 ± 0.5, and 32.0 ± 0.5 pF, respectively, by a capacitance bridge. In this case the deviation of the above value from unity is at most 0.7%, even though C_s vanishes. Accordingly, W_l can be reduced to a simple form within the above accuracy as

$$W_l = W_g \frac{V_g}{V_l} \frac{E_c}{E_a} \frac{V_{tl}}{V_{tg}} . \tag{5}$$

Thus, we can determine W_l relative to W_g within the accuracy limit mentioned above, if the pulse heights V_l, V_g, V_{tl}, and V_{tg} are measured.

EXPERIMENTAL APPARATUS AND PROCEDURE

Liquid chamber

The collector and the cathode are made of flat surfaced stainless-steel disks with diameters of 15 mm. The grid is an array of wires with a diameter of 10 μm strung with a 100 μm spacing onto a square flange. The distance between the

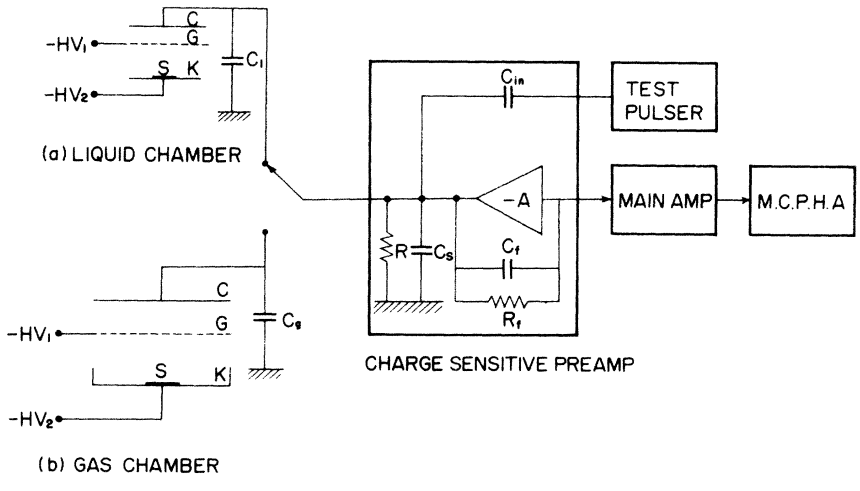

FIG. 1. Schematic diagram of experimental apparatus and block diagram of electronic circuits: (a) liquid chamber; (b) gas chamber.

cathode and the grid is 4 mm, and 2 mm between the grid and the collector. The shielding inefficiency of the grid is estimated to be 0.92% and the critical ratio of fields, at which the grid begins to trap electrons, to be 1.92, using the formulas given by Bunemann, Cranshaw, and Harvey.[11]

The chamber was fixed with three feed-through seals in a glass vessel. The vessel and the gas-filling system were evacuated to a pressure of 2×10^{-7} Torr. During the evacuation, the chamber and the gas-filling system were baked out at about 100 °C for more than 16 hours. High-purity argon gas (99.999%), which was purified in a hot calcium (700 °C) purifier for more than 48 hours, was condensed into the chamber, which was cooled by liquid nitrogen. Then the reservoir of the liquid nitrogen was replaced by a cryostat to keep the chamber at liquid-oxygen temperature in the course of the experiment. A ^{207}Bi conversion-electron source (1-mm-diam spot), which emits electrons with energies of 1.05, 0.976, 0.55, and 0.48 MeV, was deposited chemically on the center of the cathode surface.

The differential and the integral time constants of the main amplifier were selected to be 2 μsec each, in order to achieve the optimal condition for the comparison of the pulse heights from the liquid chamber and the gas chamber, since the use of longer time constants gave rise to large microphonic effects in the liquid chamber. The typical pulse-height spectrum of ^{207}Bi is shown in Fig. 2. The full width at half-maximum (FWHM) for the 0.976 MeV conversion-electron peak was 39 keV. The resolution showed a great improvement compared with that obtained by Marshall.[12] The saturation curve of the pulse height for the 0.976 MeV peak is shown in Fig. 3. The output pulse height was constant to within 0.5%, while the field was varied from 15 to 22 kV/cm.

Gas chamber

The collector and the cathode are both brass circular disks with diameters of 190 mm. A guard ring of 15-mm height is fixed on the edge of the cathode. The grid is an array of wires with a diameter of 100 μm strung at a spacing of 1 mm onto a flange of 190-mm diameter and placed between the collector and the cathode at a distance of 24.5 mm from the cathode and 18 mm from the collector. The shielding inefficiency of the grid is estimated to be 1.02%. The mixed α source of ^{239}Pu, ^{241}Am, and ^{244}Cm, of which the energies are 5.156, 5.486, and 5.806 MeV, respectively, was placed at the center of the cathode. The chamber was evacuated to less than 1×10^{-6} Torr and filled with the argon (95%)–methane (5%) mixture at 2 atm. The pulse-height spectrum of α particles is shown in Fig. 4. The FWHM for the peak of ^{241}Am was 60 keV. The saturation curve of ^{241}Am peak is shown in Fig. 5.

CORRECTIONS AND RESULT

In the electron-pulse method using a gridded ionization chamber, a correction is needed for the shift of the peak position to the low-energy side. The shift of the peak position originates from the energy loss of particles within the source, the shielding inefficiency, and the variation of the rise time due to the emission angles of particles.

In the gas chamber, each component of the shift is estimated to be 14 keV from the source thickness, 15 keV from the shielding inefficiency, and 16 keV from the rise-time effect, respectively. The shift correction amounts to 45 keV, corresponding to 0.82% of the energy of ^{241}Am α particle.

In the liquid chamber, on the other hand, the

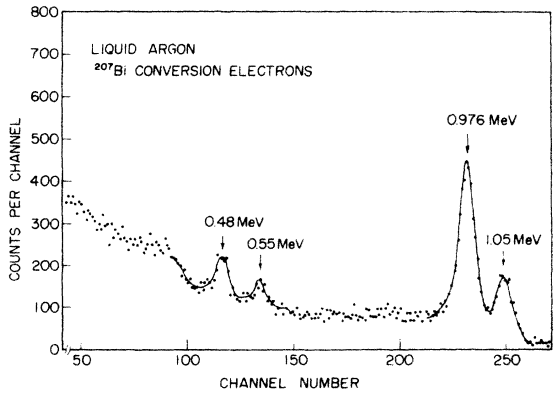

FIG. 2. Pulse-height spectrum of ^{207}Bi conversion electrons measured by the liquid ionization chamber.

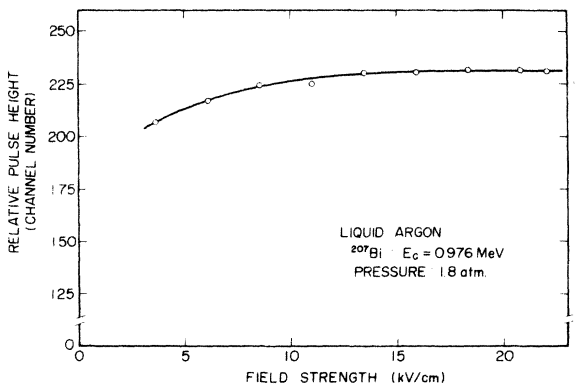

FIG. 3. Saturation curve of ionization for 0.976 MeV conversion electrons.

correction due to the above effects can almost be neglected for the conversion electrons for the following reasons. The correction due to the source thickness for the conversion electron is negligible, since the specific ionization of the energetic electrons is very small. The rise-time effect is also negligibly small owing to the large saturated value of electron drift velocity in the liquid argon. It is difficult to estimate the correction due to the shielding inefficiency of the grid, since the trajectory of electrons in the liquid is not straight. The correction, however, is considered to be negligible even if a straight trajectory is assumed.

After having made the correction to the shift of peak position, W_i was determined to be $23.6 \pm^{0.5}_{0.3}$ eV from Eq. (5) by using 26.09 eV for the W value in the argon (95%)–methane (5%) mixture.[7] This value is clearly lower than the W value for gaseous argon.

DISCUSSION

For all five rare gases, the ratio of the W value to the corresponding ionization potential W_g/I is equal to 1.7 with a few percent divergence. According to Platzman,[8] the ratio can be expressed from the energy balance of absorbed energy as

$$W_g/I = E_i/I + (E_{ex}/I)(N_{ex}/N_i) + \epsilon/I, \qquad (6)$$

where N_i is the number of ions produced at an average energy expenditure of E_i, N_{ex} is the number of excited atoms at an average expenditure of E_{ex}, and ϵ is the average kinetic energy of subexcitation electrons. Here, we assume that the band structures found in solid argon, krypton, and xenon also exist in the liquid states for the following reasons: (i) Mobility values of electrons in the liquids are of the same order of magnitude as those in the solids[13]; (ii) Wannier excitons are observed in liquid xenon and the "effective" energy gap of liquid xenon is estimated to be almost the same as that of solid xenon,[14] although the band edges may become smeared by the increased lattice disorder.[15] Accordingly, we can reasonably expect that I in Eq. (6) corresponds to the band-gap energy E_g for liquid or solid rare gases. In the condensed state, therefore, I and W_g should be substituted by E_g and W_i, respectively:

$$W_i/E_g = E_i/E_g + (E_{ex}/E_g)(N_{ex}/N_i) + \epsilon/E_g, \qquad (7)$$

where N_{ex} and N_i are considered to be the numbers of excited atoms including excitons and electron-hole pairs, respectively.

Since E_i is an average energy loss per ionizing collision, we used a mean value of gap energy in the band gap as E_i. Thus, the ratio E_i/E_g for argon was estimated to be 1.08 from the band structure given by Rössler,[16] assuming the width of the valence band is negligibly small.

The ratios E_{ex}/E_g and N_{ex}/N_i were estimated from the optical approximation to be 0.89 and 0.26, respectively, using the absorption spectrum of solid argon measured by the DESY group.[17] This estimation was also done on the assumption that all the excitons that lie in the continuum above E_g dissociate to electron-hole pairs immediately.

The ratio ϵ/E_g was estimated by using the following two models. (i) The "gas model" is based on the fact that the lowest excitation level in the solid state is not so much different from that in the gaseous state.[18] Since the lowest level is the main factor which affects the ϵ value, the ϵ values in both states can be considered to be equal, provided that energies of the lowest levels are equal for both states. Analyzing the data on the additional ionization yield for rare-gas mix-

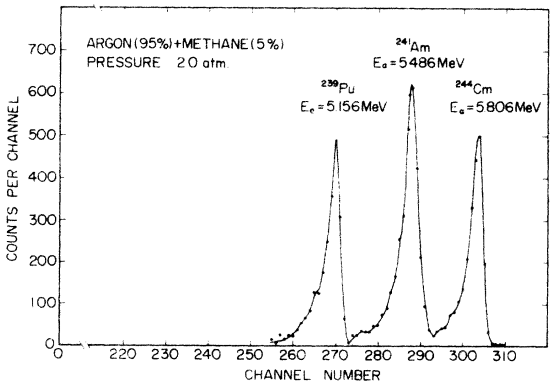

FIG. 4. Pulse-height spectrum of ^{239}Pu, ^{241}Am, and ^{244}Cm α particles measured by the gas ionization chamber.

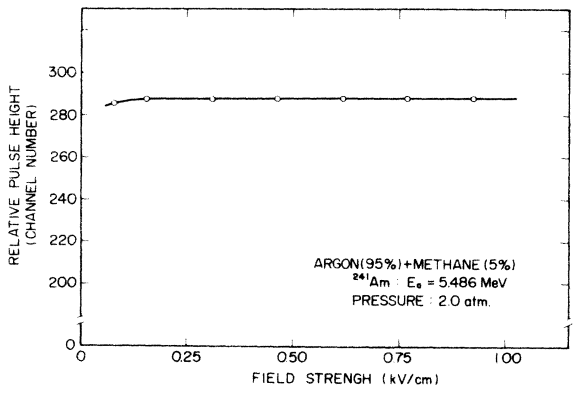

FIG. 5. Saturation curve of ionization for ^{241}Am α particle (5.486 MeV).

tures measured by Jesse and Sadauskis,[19] Platzman estimated ϵ/I to be 0.3 for all rare gases.[8] Using this value, the ratio ϵ/E_g was calculated from the following relation:

$$\epsilon/E_g = (\epsilon/I)(I/E_g) = 0.3(I/E_g). \quad (8)$$

(ii) The "solid model" is based on the Shockley model[20] for the estimation of the W value for semiconductor materials. Namely, the energy ϵ is taken to represent an average kinetic energy of final electrons with energy less than E_i which no longer create electron-hole pairs; furthermore, it is assumed that such electrons distribute proportionally to the state density dN/dE for the energy level. Thus, ϵ is given as

$$\epsilon = \int_0^{E_i} E\frac{dN}{dE}\,dE \Big/ \int_0^{E_i} \frac{dN}{dE}\,dE. \quad (9)$$

The ratio ϵ/E_g for argon was estimated by using the state density for solid argon given by Rössler.[16] Since Rössler does not give the density of states for energies above 9 eV from the bottom of the conduction band, in this estimation the density of states for argon was assumed to keep the same value as that at 9 eV for the range from 9 eV to $E_i = 1.08\ E_g(14.3\ \text{eV}^{18}) = 15.4$ eV. The ratio ϵ/E_g for argon, obtained from these models, is 0.33 for the gas model and 0.43 for the solid model, respectively.

Using the values as derived above, we obtained a value of 1.64 from the gas model and 1.73 from the solid model, respectively, for W/E_g in argon. Although a difference of about 5% between both models is found, the agreement between these estimated values is fairly good, if the roughness in the estimation is taken into consideration. Also, these values are in good agreement with the experimental value of 1.65. For krypton and xenon, the estimate of the ratio W/E_g was done in the same manner.

Table I shows the result of calculation of each value for argon, krypton, and xenon. From the table, it is clear that the ratio N_{ex}/N_i for the condensed state is much lower than that for the gaseous state (0.4); in particular, the reduction of the ratio for krypton and xenon is remarkable.

The ratios ϵ/E_g for krypton and xenon from the solid model were estimated by taking 9 eV as the upper limit of the integration in Eq. (9). This calculation is a good approximation for xenon, but not so good for krypton because the true upper limits of the integral E_i are $1.08\ E_g(9.28\ \text{eV}^{18}) = 10.5$ eV for xenon and $1.11\ E_g(11.7\ \text{eV}^{18}) = 13$ eV for krypton. As in the case of argon, the value of W/E_g for xenon obtained from the solid model is about 5% smaller than that from the gas model.

The experimental values for krypton and xenon obtained by the steady current method,[21] are also shown in Table I. These observed values are clearly lower than those for the gaseous state and are in fairly good agreement with those estimated from the solid model rather than those from the gas model.

From the viewpoint of the variance in the ionization fluctuations, which is expressed as the Fano factor, it is interesting to note that the ratios N_{ex}/N_i for krypton and xenon are very small. Alkhazov, Komar, and Vorob'ev[22] concluded that the Fano factor depends upon the distribution of numbers of excited and ionized atoms and decreases with the decrease in N_{ex}/N_i. This means that the Fano factors for krypton and xenon in the condensed state may be considerably lower than those in the gaseous state.

Further investigations of the W values and the Fano factors for the liquid state are in progress and will be reported in the near future.

TABLE I. Quantities appearing in the energy-balance equation (7) for argon, krypton, and xenon in condensed state. The W_l value and the ratio W_l/E_g for argon obtained from the present experiment and those of krypton and xenon given by Ref. 21 are compared with ratios (W/E_g) calculated by using Eq. (7).

		Calculated					Experiment	
				ϵ/E_g				
Liquid	E_g (eV)	E_i/E_g	$(E_{ex}/E_g)(N_{ex}/N_i)$	Gas model	Solid model	W/E_g	W_l (eV)	W_l/E_g
A	14.3 [a]	1.08	0.89× 0.26=0.23	0.33	0.43	1.74 1.64	$23.6^{+0.5}_{-0.3}$	$1.65^{+0.02}_{-0.01}$
Kr	11.7 [a]	1.11	0.9 ×0.1=0.09	0.36	0.42	1.62 1.56	20.5 ± 1.5 [b]	1.75 ± 0.13 [b]
Xe	9.28 [a]	1.13	0.9 ×0.06=0.05	0.39	0.51	1.69 1.57	16.4 ± 1.4 [b]	1.73 ± 0.15 [b]

[a] Reference 18.

[b] Reference 21.

CONCLUSIONS

The W value in liquid argon for conversion electrons was measured by the electron-pulse method and was found to be $23.6 \pm {}^{0.5}_{0.3}$ eV, which is clearly lower than that for gaseous argon.

The W value is in good agreement with the estimated value obtained from the energy-balance equation, assuming that the band structure in solid argon can be applied to the liquid state and all the excitons excited to the continuum above E_g dissociate to electron-hole pairs immediately.

Using the same method, the W values for krypton and xenon in the condensed state were estimated and the results showed that the estimated values were in fairly good agreement with those measured by the steady-conduction-current method. In the estimation, it was found that values of N_{ex}/N_i for krypton and xenon in the condensed state were much lower than those in the gaseous states. This suggests that the Fano factor in the condensed state of those rare gases may be considerably lower than that in the gaseous state.

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†Present address: National Laboratory for High Energy Physics, Tsukuba, Ibaraki, Japan.

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Erratum

Erratum: Average energy expended per ion pair in liquid argon [Phys. Rev. A 9, 1438 (1974)]

M. Miyajima, T. Takahashi, S. Konno, T. Hamada, S. Kubota, H. Shibamura, and T. Doke

In this experiment, a terminator of 100 Ω (not shown in Fig. 1) was used at the receiving end of the test pulser in the charge-sensitive preamplifier. Then the terminator side of the capacitor C_{in} can be regarded as grounded because of the small impedance of the terminator. Thus, Eq. (1) should be written as

$$V_i = \frac{eE_c}{W_i} \frac{A}{C_{in} + C_s + C_i + (1+A)C_f} G . \qquad (1')$$

Also, Eq. (2) should be written in the following precise form:

$$V_{ti} = C_{in} V_t \frac{A}{C_{in} + C_s + C_i + (1+A)C_f} G . \qquad (2')$$

Accordingly, from Eqs. (1′) and (2′), Eq. (3) is written as

$$W_i = eE_c \frac{V_{ti}}{V_i} \frac{1}{C_{in} V_t} . \qquad (3')$$

Then W_i can be written as in Eq. (5) without any discussion of the accuracy limit concerning Eq. (4). As a result, W_i determined for the liquid argon should be 23.6 ± 0.3 eV, not $23.6^{+0.5}_{-0.3}$ eV.