Average energy expended per ion pair in liquid xenon*

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(Received 28 April 1975)

Measurements were made of the average energy W per ion pair formed in liquid xenon by internal-conversion electrons from ^{207}Bi. We observed voltage pulses resulting from electron collection either in liquid xenon or in a gaseous mixture of argon (95%) and methane (5%). The relative pulse heights for the two materials determine the ratio of the W values. Using the known W for the gaseous mixture, we obtained a liquid-xenon W of 15.6 ± 0.3 eV. This value is considerably smaller than the gas-phase values, 21.5 or 21.9 eV. For interpretation, we adapted Platzman's energy-balance equation to liquids, assuming a conduction-band picture. Theoretical values thus calculated agree well with experiment.

I. INTRODUCTION

In the field of radiation physics, it is an important question whether the average energy required to form an ion pair in the liquid state (W_l) is smaller than that in the gaseous state (W_g). In 1969, one of the authors (T.D.) predicted that the W value in the liquid state of rare gases (Ar, Kr, and Xe) might be smaller than that in the gaseous state, assuming that the electronic band structure in the liquid rare gas is the same as that in the solid.[1] Recently, it was shown by several investigators that the band-gap energy E_g in the liquid state of xenon is almost the same as that in the solid state, in corroboration of Doke's assumption.

To ascertain Doke's prediction for liquid rare gases, we measured W_l in liquid argon more precisely than in earlier investigations, and demonstrated that W_l(Ar) (=23.6 eV) is clearly smaller than that (=26.4 eV) in gaseous argon. The result was reported in a previous paper[2] (referred to as I). According to Doke's prediction, such a reduction in the W value for the liquid state is expected to be more significant for xenon. So far, no measurements on xenon precise enough to distinguish a meaningful difference between W_l and W_g have been made.

From measurements of steady conduction currents produced by irradiation with x rays, Robinson and Freeman[3] obtained for the G value of free electrons in liquid xenon a value of 13.7, which corresponds to W_l(Xe) of 7.3 eV. This value is smaller than the band-gap energy of liquid xenon obtained from reflection spectra[4] (9.3 ± 0.4 eV) or from photoconductivity [8.9 eV (Ref. 5) or 9.22 ± 0.01 eV (Ref. 6)]. On the other hand, some authors measured W_l(Xe) and found 23.4 eV (Ref. 7) or 23 ± 2 eV (Ref. 8), i.e., values which are similar to the W value in gaseous xenon [21.9 eV (Ref. 9) or 21.5 eV (Ref. 10)]. From the analysis of pulse-height distributions produced by α particles, Konno and Kobayashi[11] estimated W_l(Xe) to be 17.3 eV, assuming that the extrapolation based on Jaffe's theory[12] is valid. By measuring steady conduction currents produced by a β source, Takahashi, Konno, and Doke[13] obtained W_l(Xe) to be 16.4 ± 1.4 eV. This value might have been influenced, however, by the effect of backscattering of β particles to the source plate.

For precise determination of W in liquid rare gases, α particles are not advantageous, because the complete collection of the electronic charges is far from being achieved and the application of Jaffe's theory to the estimation of produced electronic charges cannot theoretically be justified for liquid rare gases.[13,14] Accordingly, the use of charged particles with low ionizing power avoids the difficulty mentioned above. Also, the steady-conduction-current method sometimes causes large uncertainties because of the low accuracy in the estimation of absorbed energy. If electron pulses produced by conversion electrons or γ rays

are observed, as in the measurements of W_l(Ar),[2] the energy for each induced pulse can easily be determined without any ambiguity.

Very recently, Gadenne, Lansiart, and Seigneur[15] constructed a liquid-xenon gridded ionization chamber of centimeter size to detect γ rays. The value of W_l(Xe) measured with this chamber was 16.5 ±0.7 eV, which is very close to that of Ref. 13. However, this value seems to be unreliable, because the measurement was done at a comparatively weak electric field, and because the corrections for attachment and grid trapping of electrons appear uncertain.

As can be seen from the foregoing discussion, the literature values of W_l(Xe) are scattered. Therefore, it is desirable to measure more precisely W_l(Xe) with the electron-pulse method using an internal-conversion electron source, as done for liquid argon.

II. EXPERIMENTAL

In a so-called fast ionization chamber, operated with electron collection, the amplitude of the voltage pulse at the collector depends on the initial location of a disintegration track in the chamber. This can be attributed to the image charge induced at the collector by positive ions, which remain almost stationary during the electron collection time. To eliminate the effect of the positive ions, it is necessary to introduce a shield grid, kept at an intermediate potential, between the collector and the cathode. The use of such a gridded ionization chamber makes possible the precise determination of the electronic charges produced by ionizing radiation.

In the present set of measurements, we have used gridded ionization chambers again. The collector and the cathode of the liquid chamber were circular disks 17 mm in diameter, and a grid was placed between them. The collector was separated from the grid by 1.8 mm, and the grid from the cathode by 2.2 mm. The grid is an array of gold-plated stainless-steel wires with a diameter of 11.5 μm, strung with a 150-μm spacing onto a flange. The grid shielding inefficiency was calculated to be 1.88%, and the critical ratio of fields, at which the grid begins to trap electrons, was estimated to be 1.64 from the formula given by Bunneman, Cranshaw, and Harvey.[16]

The chamber was fixed with three feed-through seals in a stainless-steel vessel. The chamber and gas-filling system were evacuated at a pressure of 2×10^{-7} Torr and baked out at about 120°C for longer than 40 h.

The xenon gas, which was purified with a copper catalyzer and a barium-titanium getter, was supplied by Toshiba Electric Co.[17] It was further purified by a purifier containing about 20 000 pieces of barium-titanium getter[18] maintained at about 600°C. The gas was circulated by convection in the purifier for over 72 h. The vacuum system and the purifier were made of stainless steel. To connect the stainless-steel flanges, we used copper gaskets. No filter was adopted in our system, because fine powder was not expected to be produced from the getter. By using this purifier, satisfactory and reproducible saturation characteristics were obtained, as will be described below. The getter seems to work much more stably than the hot calcium, which was used in the measurement of the W value for liquid argon.[2] The purified xenon was condensed into the stainless-steel vessel, which was cooled by an n-propyl alcohol bath maintained at 183 ±1 K. A ^{207}Bi conversion-electron source (1-mm-diam spot) was deposited chemically on the center of the cathode.

The gas chamber and the mixed α source used in the present experiment were entirely the same as reported in the measurements of the W value for liquid argon.[2] The gas chamber was filled with an Ar(95%) + CH_4(5%) gas mixture at 2 atm.

The amplification system, with a differential time constant of 2 μsec and an integral time constant of 2 μsec, was used for ionization chambers filled either with liquid or gas.

III. RESULTS

The output pulse height of the preamplifier attached to the liquid-filled chamber V_l was compared with that of the same preamplifier attached to the gas-filled chamber V_g. From the ratio V_g/V_l, W_l(Xe) was calculated by means of Eq. (5) of I:

$$W_l(\mathrm{Xe}) = W_g \frac{V_g}{V_l} \frac{E_c}{E_\alpha} \frac{V_{tl}}{V_{tg}}, \qquad (1)$$

where E_c is the energy of the conversion electron (=0.976 MeV), E_α is the energy of α particles emitted from ^{241}Am (=5.486 MeV), and W_g is the W value in the gas mixture[10] (=26.09 ±0.13 eV). Further, test pulses of the same amplitude from a mercury-relay pulse generator[19] are fed to the preamplifier through a small capacitance. V_{tl} or V_{tg} is the pulse height obtained at the output stage of the preamplifier, when the preamplifier is attached to the liquid- or gas-filled chamber.

A typical pulse-height spectrum of ^{207}Bi for liquid xenon is shown in Fig. 1. For liquid argon, the peaks of K conversion electrons were higher than those of L conversion electrons (see Fig. 2 in I). For liquid xenon, the situation is reversed: the peaks corresponding to L conversion electrons

are higher than those of K conversion electrons. This fact may be due to the larger photoelectric absorption coefficient of liquid xenon for γ rays.

Corrections for energies of K conversion electrons in the liquid chamber due to the shielding inefficiency[16] were estimated to be less than 0.37%. Corrections arising from the source thickness and variation of the rise time were considered to be negligible, as explained in I. Apart from these corrections, we cannot expect any cause of energy shift for the peaks of K conversion electrons (0.976 and 0.48 MeV). On the other hand, if the other two pronounced peaks are mainly due to γ rays, these peaks might be shifted to the low-energy side, because of the escape of x rays originating from the xenon atoms. Thus, we assume that the peak of K conversion electrons at 0.976 MeV is not shifted, and take it as an energy standard. Energies shown in parentheses in Fig. 1 are thus determined.

Saturation characteristics for observed peaks are shown in Fig. 2. The pulse height of the 0.976-MeV peak of K conversion electrons was constant within 0.9% while the electric field was varied from 12.1 to 17.3 kV/cm, and the saturation characteristics were reproducible. Thus, we adopted the value of V_i at the maximum field strength as the saturated pulse height.

The same pulse-height spectrum as shown in the previous paper[2] was obtained for the gas chamber. The pulse height was constant within 0.5% while the electric field was varied from 0.4 to 1.0 kV/cm. In order to obtain V_g, we made the corrections[2] for the shift of the pulse height of ^{241}Am α particles (45 keV).

From the ratio V_i/V_g, W_i was calculated to be $W_i(\mathrm{Xe}) = 15.6 \pm 0.3$ eV, according to Eq. (1).

The extrapolated pulse height V_i^{ext} for infinite field strength was also estimated from a least-squares analysis of the "$1/E - 1/V$ plot," which has been often used for the liquid ionization chamber.[13,20,21] The value of W_i(Xe) calculated from V_i^{ext}/V_g is 15.3 eV. But this value might be an underestimate for reasons discussed in Ref. 13. Therefore, we evaluated the value of W_i(Xe) to be 15.6 ± 0.3 eV, a result probably more reliable than the extrapolated value.

IV. DISCUSSION

We have adopted the peak of 0.976-MeV K conversion electrons to evaluate W_i(Xe), because we find no reason for an energy shift, except for the source thickness, the shielding inefficiency, and variation of rise time. Because we found the linearity of the electronic system to be satisfactory, we examined whether the observed energy of each peak was consistent with that in published tables,[21] taking the 0.976-MeV peak as an energy standard.

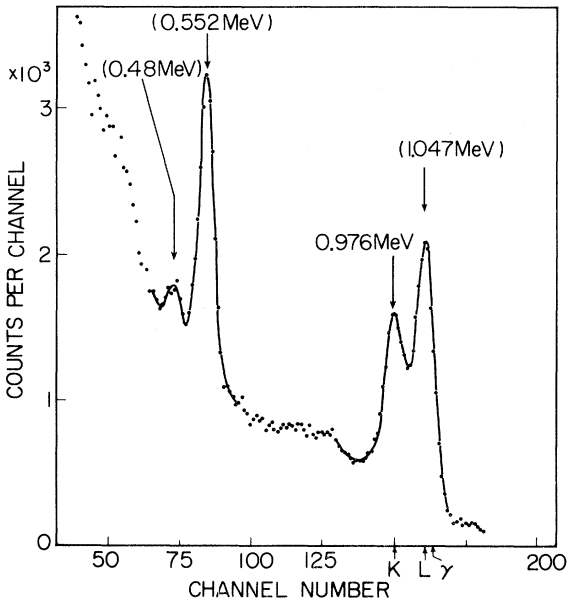

FIG. 1. Pulse-height spectrum of conversion electrons and γ rays emitted from a ^{207}Bi source, measured by the liquid ionization chamber filled with liquid xenon. Energies shown in parentheses are calibrated by the peak at 0.976-MeV K conversion electrons. The arrows on the horizontal axis indicate known energies of K conversion electrons, L conversion electrons, and γ rays.

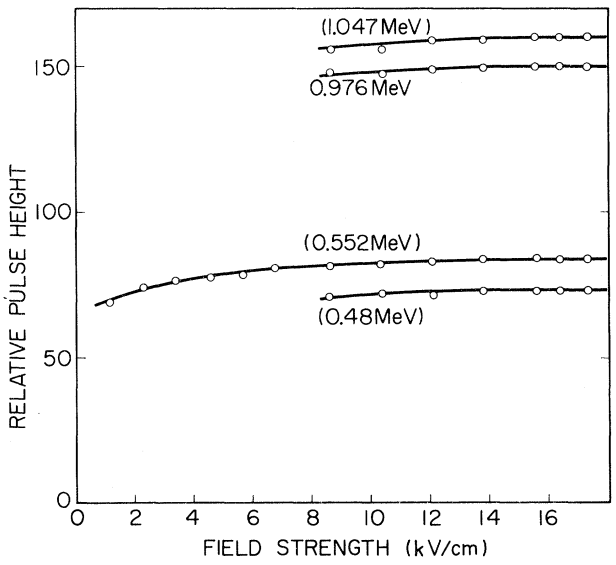

FIG. 2. Saturation curves of the pulse heights for a ^{207}Bi source obtained by the chamber filled with liquid xenon. For the field strengths of 8 kV/cm and lower, only the peak at 0.552 MeV was separately observed.

TABLE I. Quantities appearing in the energy-balance equation (2) for argon, krypton, and xenon in the condensed state. The W_I value and the ratio W_I/E_g for xenon were obtained from the present experiment, and those for argon and krypton were given by Refs. 2 and 13. The ratio W/E_g was calculated by means of Eq. (2).

Liquid	E_g (eV)	Calculated E_i/E_g	Calculated $(E_{ex}/E_g)(N_{ex}/N_i)$	Calculated ϵ/E_g	Calculated W/E_g	Experiment W_I (eV)	Experiment W_I/E_g
Ar	14.3[a]	1.08	$0.89\times0.21=0.19$	0.36	1.63	23.6 ± 0.3[b]	1.65 ± 0.02[b]
Kr	11.7[a]	1.11	$0.9\times0.08=0.07$	0.47	1.65	20.5 ± 1.5[c]	1.75 ± 0.13[c]
Xe	9.28[a]	1.13	$0.9\times0.06=0.05$	0.48	1.66	15.6 ± 0.3	1.68 ± 0.03

[a]These values were evaluated by Baldini (Ref. 26) for the solid state. For liquid xenon, almost the same values were recently obtained by several authors (Refs. 4–6).
[b]Reference 2.
[c]Reference 13.

The energy (0.480 MeV) of our lowest peak agrees well with the energy value for K conversion electrons (0.482 MeV) recommended by Lederer, Hollander, and Perlman.[22] On the other hand, the energies of the other two peaks, 1.047 and 0.552 MeV (Fig. 2), were smaller by 17–18 keV than the known energies of γ rays, i.e., 1.064 and 0.570 MeV. The energy shift may be attributable to the contribution of L conversion electrons and the escape of K x rays originating from xenon atoms. An energy shift due to the grid shielding inefficiency for 1.064-MeV γ rays is slightly larger than that for conversion electrons, but we cannot explain the energy shift of 17 keV by this effect.

Assuming that the band structure found in solid argon, krypton, and xenon also applies in the liquid state, we consider possible theoretical values of W_I. For this purpose, we may use Platzman's energy-balance equation

$$\frac{W_I}{E_g}=\frac{E_i}{E_g}+\frac{E_{ex}}{E_g}\frac{N_{ex}}{N_i}+\frac{\epsilon}{E_g}, \tag{2}$$

where N_i is the number of electron-hole pairs produced at an average energy expenditure of E_i, N_{ex} is the number of excitons at an average energy expenditure of E_{ex}, ϵ is the average kinetic energy of subexcitation electrons, and E_g is the band-gap energy.

If Shockley's model is used as in I, the quantity ϵ is given as

$$\epsilon=\int_0^{E_1} E\frac{dN}{dE}\,dE\Big/\int_0^{E_1}\frac{dN}{dE}\,dE, \tag{3}$$

where E_1 is the energy of the lowest exciton state and dN/dE the density of states. The value of ϵ/E_g for argon calculated from Eq. (3) is smaller than that in I, in which E_i was adopted as the limit of integration. As the upper limit of integration in Eq. (3), E_1 should be adopted rather than E_i. For krypton and xenon, however, ϵ values are almost unchanged. The ratio of the number of excitons to that of electron-hole pairs, N_{ex}/N_i, was also reexamined and the refined values are shown in Table I. The experimental value of $W_I/E_g=1.68$ for liquid xenon agrees well with the theoretical value (1.66).

The recent interest in the use of liquid xenon as a detector medium[1,7,8,15,20,23,24] is motivated by the large electron mobility and the large atomic number. In addition to these desirable properties, the value of N_{ex}/N_i is extremely small. This suggests that the Fano factor[25] of liquid xenon would be very small. Consequently, liquid xenon should be advantageous for nuclear-detector material.

Investigations of Fano factors for liquid rare gases are in progress.

ACKNOWLEDGMENT

The authors wish to thank Dr. Mitio Inokuti for his careful reading of the manuscript.

*Research supported by the Mitsubishi Foundation.

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