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W value in liquid krypton

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The *W* value, or the average energy required to produce an electron-ion pair, was measured in liquid krypton with a pulsed ionization chamber and ^{207}Bi 976-keV internal-conversion electrons. From the charge collected at the highest electric field of 16 kV/cm, the *W* value was determined to be 18.4±0.3 eV. Extrapolation of the charge to infinite field strength yields a smaller *W* value, reduced by an amount which depends on the recombination model used to fit the data. The δ-electron model gives the lowest value of 16.8 eV. An estimation of the *W* value in liquid krypton, using Platzman's energy-balance equation, is in good agreement with the experimental result, but is not precise enough to rule out model-dependent lower values.

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I. INTRODUCTION

The *W* value, or the average energy required to produce an electron-ion pair, in liquid rare gases is a measure of the ionization efficiency of these materials in practical radiation detectors. The *W* values in liquid argon and xenon have been measured by several investigators in the last two decades, both by measuring the steady conduction current produced by irradiation of the liquids by x rays or electrons, and by measuring the electron pulses in the liquids using internal-conversion electron sources. The determination of the *W* value in liquid argon and liquid xenon with a pulsed ionization chamber, was made for the first time by Takahashi *et al.* [1] and Miyajima *et al.* [2], respectively. From the charge collected at the highest applied electric field (22 kV/cm for liquid argon and 17.3 kV/cm for liquid xenon), these investigators reported a *W* value of $23.6^{+0.5}_{-0.3}$ eV in liquid argon and 15.6±0.3 eV in liquid xenon. By the same method, smaller *W* values in liquid xenon have been subsequently reported by other investigators. [3,4].

In comparison, there have been far fewer experiments in liquid krypton, and the only two measurements of the *W* value available to date are not in good agreement. The *W* value obtained by Huang and Freeman [5] is 16.7 eV, while Takahashi *et al.* [6] reported 20.5±1.5 eV, as an average between the value from the measurement at the highest applied field and the value extrapolated to infinite field. Both *W*-value results were obtained by measuring the steady conduction current produced by irradiation of the liquid with x rays or electrons from a β source. It is well known that unlike in the "electron-pulse" mode, the operation of an ionization chamber in the "steady-current" mode suffers from the problem of accurately determining the total energy absorbed in the liquid, which translates into a large uncertainty on the *W* value. A more precise determination of the *W* value in liquid krypton is, therefore, highly desirable.

In this paper we report the results of a measurement of the *W* value in liquid krypton obtained with a gridded ionization chamber operated in the "electron pulse" mode, using ^{207}Bi internal-conversion electrons. Both the experimental *W* value, as obtained from the charge collected at the highest applied field of 16 kV/cm, as well as the extrapolated *W* value derived from the charge expected at infinite field using different recombination models, are presented. The results are then compared to an estimation of the *W* value based on Platzman's energy-balance equation adapted to liquids. For the estimation, the results of recent measurements of reflectivity spectra and photoconductivity in fluid krypton were taken into account. These measurements give direct evidence of the band structure in liquid krypton, as well as the band-gap energy.

II. EXPERIMENTAL PROCEDURE

The gridded ionization chamber used for the *W*-value measurement was similar to the one we used in previous studies of the charge and energy-resolution response of liquid krypton to electrons, γ rays and α particles [7]. Modifications were made to optimize the high-voltage feedthroughs and electrode structure so that high electric fields could be applied. The collection and drift gaps were 2.4 and 2.7 mm, respectively. The ^{207}Bi source, used to irradiate the liquid, was chemically deposited on the center of the cathode plate. The electrodes were stainless-steel disks of diameter 2.5 cm. The effective area of the anode was decreased from the one used earlier [7], to reduce both the background from ^{85}Kr and the contribution of γ rays to the ^{207}Bi spectrum. The grid was an electroformed mesh made out of nickel and had a shielding efficiency of 99%. A ratio of 2 between the collection and drift field was used to provide 100% electron transmission. The total volume of the ionization chamber was approximately 0.3 l.

Before filling the ionization chamber, research-grade krypton gas was purified by passing it sequentially through an Oxisorb cartridge [8], a cold molecular-sieve trap [9], and two high-temperature getters [10]. The purification system was similar to the one used for xenon and is discussed in detail elsewhere [11]. The chamber and the gas-filling lines were baked out at about 100 °C in a vacuum better than 10^{-7} torr for at least 48 h before each experiment. A higher temperature, typically 300 °C, was used for the regeneration of the molecular sieves. The typical outgassing rate was better than 10^{-9} torr l s^{-1}. The purified krypton gas was condensed in the test chamber by using a cryogenic bath of liquid nitrogen and isopentane. The temperature of the cooling bath was 121 K, close to the triple point of krypton. At this temperature the vapor pressure of krypton is less than 2 atm.

Figure 1 shows a block diagram of the chamber and the associated electronics. Negative high voltage (HV) was independently supplied to the cathode and grid of the ionization chamber. The ^{207}Bi ionization pulses from the anode were analyzed with a charge-sensitive preamplifier, followed by a spectroscopy amplifier (MA) for further amplification and shaping. The pulse height of the output signal was analyzed with a multichannel analyzer (MCA). For charge calibration a test pulse from a precision pulse generator was injected into the preamplifier input stage through a test capacitor.

The W value in liquid krypton is defined as $W_{\rm LKr}=E_p/N$, where E_p is the known energy of the primary ionizing particle (976 keV in our case) and N is the total number of electron-ion pairs produced. Since the total charge is given by $Ne=CV_{\rm Kr}$, where e is the charge of the electron, $V_{\rm Kr}$ is the saturated pulse height measured for the 976-keV line, and C is the total capacitance in the amplifying network, $V_{\rm Kr}$ can be expressed as

$$V_{\rm Kr}=\frac{eEp}{W_{\rm LKr}}\frac{AG}{C_{\rm in}+C_{\rm Kr}+C_s+(1+A)C_f}\,, \tag{2.1}$$

where A is the open loop gain of the preamplifier, G is the gain of the main amplifier, $C_{\rm in}$ is the input capacitance of the preamplifier, $C_{\rm Kr}$ is the capacitance of the liquid-krypton detector, C_f is the capacitance of the feedback network, and C_s is the stray capacitance. Similarly, for a test pulse of amplitude V_t, injected into the preamplifier input stage through $C_{\rm in}$, the measured pulse height V_{t1} may be expressed as

$$V_{t1}=C_{\rm in}V_t\frac{AG}{C_{\rm in}+C_{\rm Kr}+C_s+(1+A)C_f}\,. \tag{2.2}$$

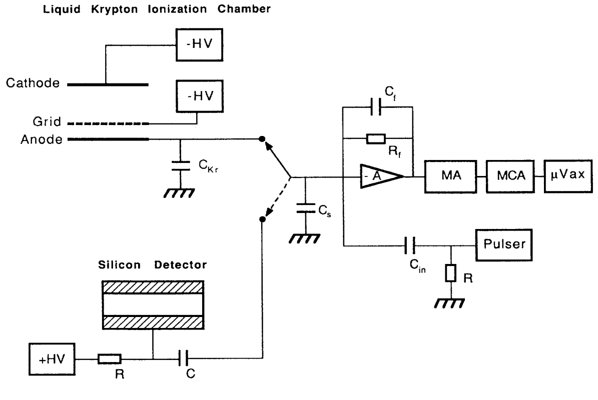

FIG. 1. Block diagram of the experimental setup used to measure W value in liquid krypton.

Taking the ratio of (2.1) and (2.2) the W value in liquid krypton can be determined from the relation

$$W_{\rm LKr}=eE_p\frac{V_{t1}}{V_{\rm Kr}}\frac{1}{C_{\rm in}V_t}\,. \tag{2.3}$$

Since the error on the measured pulse heights can be made negligibly small, the precision on $W_{\rm LKr}$ is mostly limited by the systematic error on the value of $C_{\rm in}$.

To measure $C_{\rm in}$ precisely, the ^{207}Bi pulse height spectrum was measured both with the liquid-krypton ionization chamber and with a silicon detector [12], using the same signal-readout electronics at the same temperature conditions, as shown in Fig. 1. The silicon detector used for this calibration was operated at room temperature. Following the same steps used to arrive at (2.3), the W value in silicon $W_{\rm Si}$ may be expressed as

$$W_{\rm Si}=eE_p\frac{V_{t2}}{V_{\rm Si}}\frac{1}{C_{\rm in}V_t}\,, \tag{2.4}$$

where $V_{\rm Si}$ and V_{t2} are the measured pulse heights for the 976-keV electrons and test pulse, respectively. Using the above equation, $C_{\rm in}$ can, therefore, be determined with the same precision as the known W value in silicon ($W_{\rm Si}=3.65\pm0.05$ eV [13]), if the error on the other measured quantities is negligible in comparison. An independent measurement of this capacitor with a precise capacitance meter [14] gave a consistent result within 1% of the value obtained with the silicon detector.

III. EXPERIMENTAL RESULTS

The principal radiation from ^{207}Bi consists of internal conversion electrons of energy 482, 554, 976, and 1048 keV, and γ rays of energy 570, 1064, and 1770 keV. The intensity of the 1064-keV γ-ray line is about an order of magnitude larger than the 976-keV electron line. However, due to the shallow depth of the liquid-krypton chamber used in our measurements, the γ-ray detection efficiency is rather poor, and the electron peaks are expected to dominate the pulse height spectrum.

Figure 2 shows a typical ^{207}Bi pulse-height spectrum measured in liquid krypton. The rightmost peak is the test-pulse distribution, which corresponds to a noise level of 700 electrons full width at half maximum (FWHM). The 976-keV K conversion electron line is the dominant feature of the spectrum. It becomes clearly distinguishable from the higher-energy peak produced by 1048-keV L conversion electrons and 1064-keV γ rays at fields higher than 3 kV/cm. The low-energy electron and γ-ray lines are visible, even at high fields, only as a small enhancement, as shown in the inset of Fig. 2. This is due to the background from ^{85}Kr. Krypton, in fact, contains small amounts of the radioactive isotope ^{85}Kr that has a half-life of 11 years. It decays by β decay with an endpoint energy of 670 keV and releases an average energy of 250 keV. We observed a background rate of approximately 500 counts s^{-1} cm^{-3}.

The ^{207}Bi pulse height spectrum in liquid krypton was measured as a function of electric-field strength, in the range 0.1–16 kV/cm. The peak location and the width

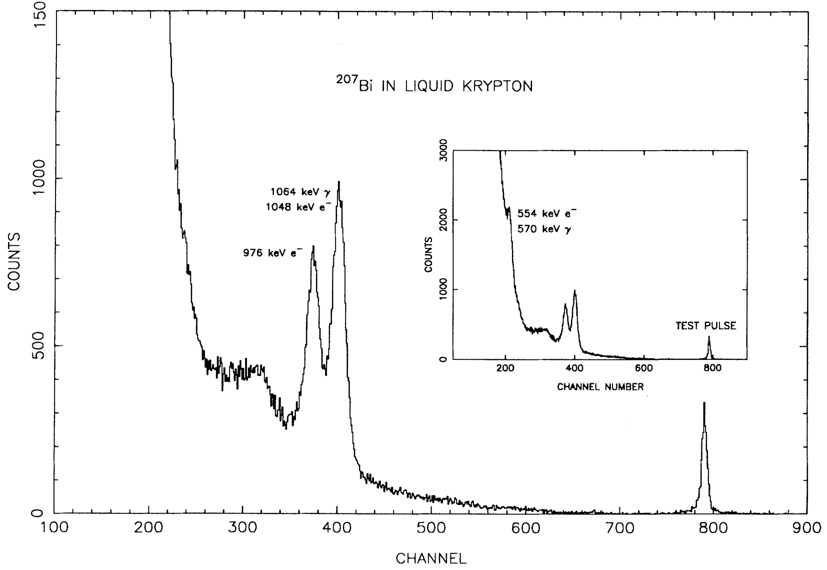

FIG. 2. Energy spectrum of ^{207}Bi in liquid krypton showing the 976-keV line and the combined 1048 and 1064-keV lines. The inset shows the low-energy lines in the range 554–570 keV.

of the measured lines were determined, for each field setting, by a least-squares fit with Gaussian functions over a constant background. The best, noise subtracted, energy resolution in liquid krypton is 3.1% FWHM at 1 MeV [7], compared to 4.5% [15] and 2.8% [16] FWHM in liquid xenon and argon, respectively, at similar field strength. The measured pulse height was converted into number of electrons using the absolute charge calibration obtained with the silicon detector. Figure 3 shows the field dependence of the collected charge for the 976-keV line and the combined 1048 and 1064-keV lines. Experiments were repeated many times and gave reproducible results. In the high-field range from 12 to 16 kV/cm the collected charge was found to remain constant to within 0.5%, indicating apparent saturation.

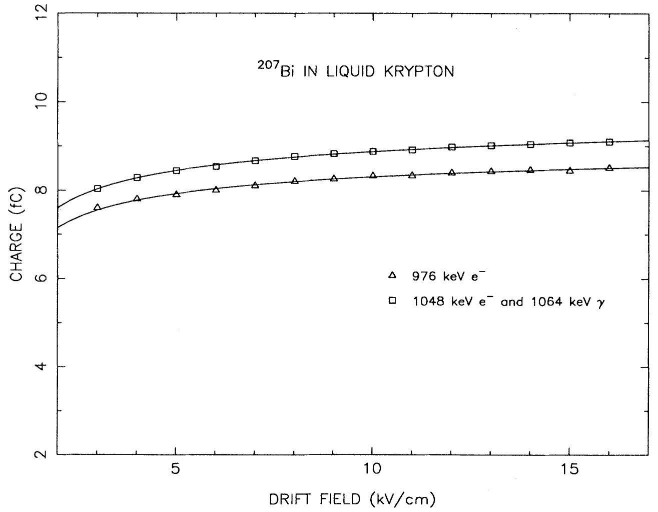

FIG. 3. Collected charge for the 976-keV line (△) and the combined 1048 and 1064-keV lines (□). The solid line is a fit with Eq. (3.1).

For the determination of the W value in liquid krypton we used the data for the 976-keV internal conversion electrons, since the energy of this line is well defined. A shift in energy due to finite source thickness, grid-shielding inefficiency or variation in pulse rise time was estimated to be negligible. Assuming that the charge measured at 16 kV/cm is saturated, (2.3) yields $W_{\rm LKr} = 18.4 \pm 0.3$ eV.

According to recombination models, however, saturation can only be achieved at infinite electric field strength. To describe the observed field dependence of the collected charge in liquid rare gases, the following equation resulting from the theory of Jaffe [17], in the high field approximation, has been conventionally used: $Q(E) = Q_0/(1+k/E)$, where $Q_0 = eE_P/W_{\rm LKr}$ is the charge at infinite field and k is the parameter which expresses the recombination rate, assumed constant along the column of uniform charge density produced by the primary ionizing particle of energy E_P. As we have previously reported [15], this equation does not fit well the experimental results in liquid argon or xenon, underestimating the charge collected at high fields. When used to fit the liquid-krypton data of Fig. 3, this equation yields an extrapolated W value of 17.8 eV. The results of the fit are shown in Table I (fit A).

To take into account the different rate of recombination in the high-charge-density regions of low-energy secondary electrons or δ electrons as well as near the end of the primary ionizing track, the following modified Jaffe equation [18] has been suggested and successfully used to interpret the ionization data in liquid argon [16,19] and in liquid xenon [15]:

$$Q(E) = \frac{(Q_0 - Q_\delta)}{1+k/E} + \frac{Q_\delta}{1+k_\delta/E} \, . \tag{3.1}$$

Here Q_δ is the fraction of the total charge produced by δ electrons and k_δ is the stronger rate of recombination along these δ tracks. The above equation was used to fit,

TABLE I. Parameters for fits.

Fit A	Fit B	Fit C
$Q_0(976)=8.77\pm0.02$ fC	$Q_0(976)=9.14\pm0.18$ fC	$Q_0=9.32\pm0.15$ fC
$Q_0(1064)=9.38\pm0.03$ fC	$Q_\delta(976)=0.88\pm0.29$ fC	$a=0.121\pm0.022$
$k=0.55\pm0.02$ kV/cm	$k=0.34\pm0.08$ kV/cm	$\xi_0 E=60.14\pm3.75$ kV/cm
	$k_\delta=18.00\pm12.27$ kV/cm	$\xi_1 E=0.696\pm0.001$ kV/cm
	$Q_0(1064)=9.88\pm0.22$ fC	$b=0.544\pm0.02$
	$Q_\delta(1064)=1.12\pm0.29$ fC	
$W=17.8$ eV	$W=17.1$ eV	$W=16.8$ eV

simultaneously, the liquid-krypton data for the 976-keV and the combined 1048- and 1064-keV energies, shown as solid lines in Fig. 3. From the charge at infinite field obtained from the fit of the 976-keV line the extrapolated W value obtained is 17.1 eV. The results of the fit are shown in Table I (fit B). Since k_δ is much larger than k, the contribution of δ electrons to the total charge has a very slow dependence on the applied field. Hence, based on this model, the observation of a constant charge output at high fields, to within 0.5%, cannot be indicative of complete saturation.

The same argument that the production of δ electrons affect the recombination rate along the primary particle track, and hence the energy resolution, Thomas *et al.* [20] suggested an alternative equation for the field dependence of the collected charge Q and energy resolution, E_{FWHM}, namely,

$$Q=Q_0\left[a\frac{\ln(1+\xi_0)}{\xi_0}+(1-a)\frac{\ln(1+\xi_1)}{\xi_1}\right], \tag{3.2a}$$

$$E_{\mathrm{FWHM}(\%)}=\frac{235.5}{\sqrt{E_P}}b\left[a\frac{\ln(1+\xi_1)}{\xi_1}-\frac{\ln(1+\xi_0)}{\xi_0}\right]\frac{Q_0}{Q}, \tag{3.2b}$$

where ξ_1 describes the charge density of the minimum ionizing region of the particle track and ξ_0 describes the more heavily ionizing regions of the δ electrons. The parameters a and b are functions of E_P and the δ-electron minimum and maximum energies. The above equations were used to fit our charge and energy-resolution data simultaneously. The W value from the charge extrapolated to infinite field is 16.8 eV. The results of the fit are shown in Table I (fit C). It should be noted that this double-density model contains in first approximation the simpler Eq. (3.1), and since it is also derived from Jaffe's theory, it has the same basic limitations.

IV. DISCUSSION

The ratio of the W value to the ionization potential I_g in the gas phase has been expressed by Platzman [21] as the sum of the energies required to produce electron-ion pairs, excited atoms and the kinetic energy of subexcitation electrons. The existence of a band structure in liquid rare gases is by now well confirmed experimentally and direct measurements of the band-gap energy have become available. For example, recent photoconductivity measurements have given a band-gap value of $E_g=11.55$ eV for liquid krypton [22] and $E_g=9.22$ eV for liquid xenon [23]. It is therefore reasonable to rewrite the Platzman energy balance equation for the liquid state as follows, with the band-gap energy replacing the ionization potential of the gas:

$$W_l/E_g=(\bar{E}_{\mathrm{ex}}/E_g)(N_{\mathrm{ex}}/N_i)+\bar{E}_i/E_g+\varepsilon/E_g\ . \tag{4.1}$$

Here N_i is the number of ions produced at an average expenditure of energy $\bar{E}_i$, N_{ex} is the number of excited atoms produced at an average expenditure of energy $\bar{E}_{\mathrm{ex}}$, and ε is the average kinetic energy of subexcitation electrons.

In earlier estimations of the W value in liquid rare gases using (4.1) [1,2], $\bar{E}_{\mathrm{ex}}$ and N_{ex}/N_i were obtained using the absorption spectra of solid rare gases [24]. Since the reflectivity spectra of fluid argon, krypton, and xenon have been measured since [25,26], we have used these data to estimate $\bar{E}_{\mathrm{ex}}$ and the ratio N_{ex}/N_i. The reflectivity spectrum of liquid krypton measured by Laporte *et al.* [25] is found to be as rich in excitonic structure as that in the solid state. Using this data, we estimated $\bar{E}_{\mathrm{ex}}$ and the ratio N_{ex}/N_i to be 10.15 eV and 0.26, respectively. The value of N_{ex}/N_i obtained by this method is, however, overestimated, since the reflectivity measurement is not sensitive to associative ionization in the liquid, which takes place after the primary excitation. It is known, in fact, that an excited krypton atom can further collide with a neutral atom to give rise to more electron-ion pairs,

$$\mathrm{Kr}^*+\mathrm{Kr}\rightarrow\mathrm{Kr}_2^++e^-\ .$$

We believe that this channel of ionization is important in liquid rare gases.

A more direct estimate of the ratio N_{ex}/N_i in liquid krypton can be made from ionization and scintillation data using the relation

$$\frac{N_{\mathrm{ex}}}{N_i}=\frac{L_{\mathrm{ex}}}{L_r}\left[1-\frac{N_i^*}{N_i}\right]. \tag{4.2}$$

Here L_{ex} and L_r are the intensities of excitation and recombination luminescence, respectively, and N_i^* is the number of electrons escaping geminate or initial recombinations. The ratio N_i^*/N_i is equal to $G_{\mathrm{FI}}^0/G_{\mathrm{tot}}$, where G_{FI}^0 is the free-ion yield at zero electric field and G_{tot} is the total ion yield, per 100 eV of absorbed energy. A fit to our data with the Onsager model [27], using the computer program of Dodelet *et al.* [28], yielded $G_{\mathrm{FI}}^0=3.4$,

$G_{\mathrm{tot}}=5.6$ and $b=164$ nm, where b is the most probable separation distance between the thermalized electron and ion pair. Using these values and a ratio of $L_{\mathrm{ex}}/L_r=0.49$ from Kubota *et al.* [29], we get $N_{\mathrm{ex}}/N_i=0.19$.

For the estimation of the remaining quantities, ε and $\bar{E}_i$, in (4.1), the ionization properties of the gas state were used, based on the fact that the estimated ratio W_l/E_g for rare-gas liquids is closer to the ratio $W/I_g=1.7$ for gases than it is to that observed, for example, in solid-state detectors like semiconductors. The reason for this similarity is probably in the fact that the bonding in liquid rare gases are van der Waals and not covalent like in semiconductors. For gases, $\bar{E}_i/I_g$ is equal to 1.06. Assuming that this ratio is independent of the state of the material and replacing the ionization potential in the gas by the band-gap energy in the liquid, we obtain $\bar{E}_i=1.06E_g$ for liquid krypton. In comparison, Takahashi *et al.* [1] estimated $E_i=1.11E_g$, from the band structure given by Rössler [30], assuming the width of the valence band is negligibly small.

The average energy of the subexcitation electrons ε is mostly affected by the lowest excited state, which in liquid krypton has a value of 9.92 eV [25], nearly the same as that in gas (10.0 eV) [31]. From the relation $\varepsilon/E_g=(\varepsilon/I_g)(I_g/E_g)$, using $\varepsilon/I_g=0.31$ [21] and $I_g=14.0$ eV, we estimate $\varepsilon=0.38E_g$ for liquid krypton.

Substituting $\bar{E}_i/E_g=1.06$, $\bar{E}_{\mathrm{ex}}/E_g=0.88$, $N_{\mathrm{ex}}/N_i=0.19$ and $\varepsilon/E_g=0.38$ in (4.1), we get $W_l=18.6$ eV. This estimate, obtained by applying the solid model approximation to the excitation process and the gas model approximation to the ionization process, compares well with the experimentally measured value of 18.4±0.3 eV. For comparison, the estimated *W* value in liquid krypton reported by Takahashi *et al.* [1] based solely on the solid model approximation is 19.3 eV. We recognize, however, that due to the relatively large uncertainties on the various terms on the right-hand side of the Platzman equation, this kind of estimate has a limited value in constraining the true *W* value in liquid rare gases.

V. CONCLUSION

By varying the applied electric field in the range 0.1–16 kV/cm, the saturation characteristics of a liquid-krypton gridded ionization chamber were measured repeatedly and yielded reproducible results. From the charge collected at the highest field, assumed saturated, the *W* value was determined to be 18.4±0.3 eV. Since recombination theories predict that complete charge collection can only be achieved at infinite field strength, we prefer to consider the experimental *W* value of 18.4 eV as the most precise upper limit on this quantity to date. This remark should also apply to the *W* value in liquid xenon and argon reported by other investigators using the same method. For the case of liquid krypton, the extrapolated *W* value is found to be between 3% and 8% smaller, depending on the model used to fit the data. The large uncertainty on the estimated *W* value obtained from Platzman's energy balance equation makes it in good agreement with both the experimental and the extrapolated *W* value.

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