Scintillation Light, Ionization Yield and Scintillation Decay Times in High Pressure Xenon and Xenon Methane

K. N. Pushkin, *Student Member, IEEE*, D. Y. Akimov, A. A. Burenkov, V. V. Dmitrenko, *Senior Member, IEEE*, A. G. Kovalenko, V. N. Lebedenko, I. S. Kuznetsov, V. N. Stekhanov, C. Tezuka, S. E. Ulin, Z. M. Uteshev, and K. F. Vlasik

***Abstract*—Scintillation light, ionization yield and scintillation decay times have been measured in xenon and in its mixture with a 0.05% concentration of methane as a function of the reduced electric field (E/N)—the ratio of the electric field strength to the number density of gas—at a pressure of 21 atm. The measurements of scintillation decay times in the xenon-methane mixture have been made for the first time as a function of the reduced electric field (E/N). An ionization chamber has been constructed to simultaneously measure electrons and photons from a ^{239}Pu source, which is placed in the center of a cathode in the chamber. The main peculiarity of the chamber is a movable cathode to measure scintillation light and ionization yield at various distances from the anode and monitor the purity of the investigated gas. It has been observed that both scintillation light and ionization yield decrease when methane is added into the xenon gas. Scintillation decay times in the xenon-methane mixture are observed to be longer than in the pure xenon when the electric field strength increases.**

***Index Terms*—High pressure xenon detectors, ionization yield, scintillation light, time protection chamber, xenon-methane mixture.**

I. Introduction

MUCH research has been done to investigate extensively scintillation and ionization processes in noble gases (Xe, Ar, He and etc) and in their admixtures during the last four decades [1]–[6]. New developments of the detectors based on high-pressure xenon allow one to determine the interaction point of a charged particle with the medium in the detector using the scintillation light as a trigger [7], [8]. Another point to use the prompt scintillation light has been proposed recently to correct the ionization signal [9]. Xenon is characterized by low drift velocity of the "hot" electrons that results in large diffusion. Admixtures of light molecular gases (H_2, CF_4, CH_4) into the xenon gas allow one to increase substantially drift velocity of the electrons [10]–[13]. The wide use of xenon detectors has regained a new interest to studies of scintillation and ionization processes in xenon gas admixtures. Some works have been performed before on the study of xenon-methane mixtures for proportional counters, but the processes taking place in the mixture and influencing deterioration of both energy and spatial resolution are not completely understood [14], [15]. The other experiments on the measurements of scintillation light in the xenon-methane and argon-methane mixtures revealed that scintillation light decreased when the concentration of methane increased [16], [17], but the real reason of the light decrease was not considered. A few works have been carried out recently on the study of scintillation and ionization processes in the xenon-methane and xenon-hydrogen mixtures [18]–[20]. We continue doing this research and present new experimental data on measurements of scintillation light and ionization yield in pure xenon at a pressure of 21 atm as a function of the reduced electric field (E/N)—the ratio of the electric field strength (E, V/cm) to the number density of gas (N, 1/cm^3)—and in xenon doped with a 0.05% concentration of methane. We have measured scintillation decay times in the pure xenon and xenon-methane mixture (0.05%) at low electric fields to investigate the process of excitation and recombination luminescence. The goal of this work was initiated to clearly understand the effect that a methane admixture exerts on time and coordinate characteristics of high-pressure xenon detectors.

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K. N. Pushkin, V. V. Dmitrenko, S. E. Ulin, Z. M. Uteshev, and K. F. Vlasik are with the Moscow Engineering Physics Institute (State University), 31 115409 Moscow, Russia (e-mail: p.kirill@rambler.ru).

D. Y. Akimov, A. A. Burenkov, A. G. Kovalenko, I. S. Kuznetsov, and V. N. Stekhanov are with the Institute of Theoretical and Experimental Physics, 25 117218 Moscow, Russia.

V. N. Lebedenko is with Blacket Laboratory, Imperial College London, SW7 2BW London, U.K.

C. Tezuka is with the Advanced Research Institute for Science and Engineering, Waseda University, Tokyo 169-8555, Japan.

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II. Method

A. Apparatus

A high-pressure xenon ionization chamber utilizing a Frisch grid electrode has been constructed to measure both scintillation photons and ionization electrons produced by 5.1 MeV alpha particles from a ^{239}Pu source. The ionization chamber is shown in Fig. 1. The chamber made of stainless steel has an inner diameter of 130 mm and a height of 115 mm. The cathode electrode is mechanically connected to a linear motion feedthrough to adjust the distance between the anode electrode with a magnet and measure scintillation light and ionization yield at various distances. The distance between the Frisch grid electrode and the anode is 0.5 cm. The distance between the Frisch grid electrode and the cathode could be adjusted from 0.5 to 5 cm. Thus, the peculiarity of the chamber construction allowed us to determine the purity of the investigated gas by changing the distance between the electrodes and measuring the amplitudes of

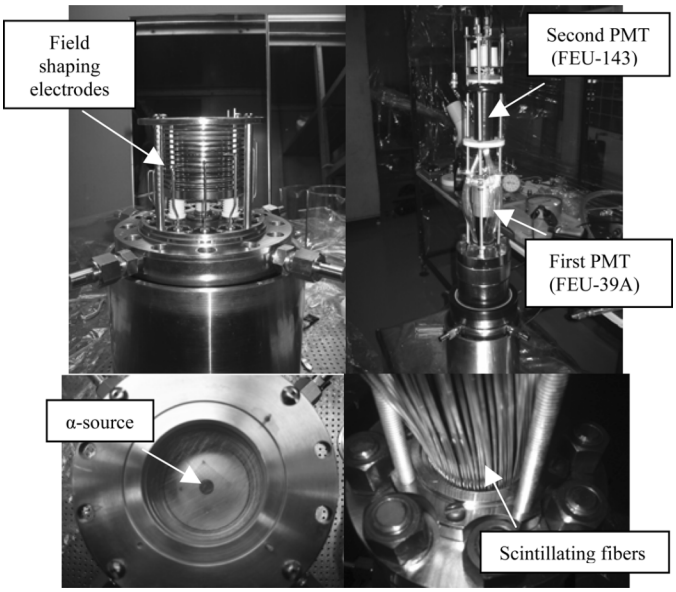

Fig. 1. A high-pressure ionization chamber with a Frisch grid electrode.

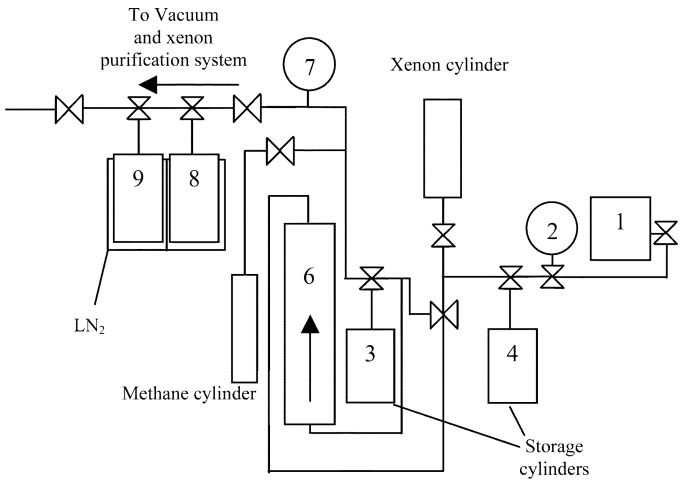

Fig. 2. A schematic view of a methane purification system.

ionization signals. The ^{239} Pu alpha source is deposited on the cathode electrode and coated with a thin layer of a MgF_2 dielectric. A tiny stainless steel grid is attached to the layer of the alpha source to prevent collection of positive ions on the surface of the MgF_2 dielectric and to prevent as a consequence distortion of the electric field in the chamber.

The thickness of the MgF_2 layer has not been precisely ascertained but it is assumed to be of the order of a few microns that could result in partial loss of alpha particle energy when the particles come out of the source. SRIM software [21] was used to simulate alpha particle energy losses, which confirmed that an alpha particle loses about 1.1 MeV energy when it penetrates through the MgF_2 layer with a thickness of 10 microns for the case of the vertical tracks. The ionization chamber incorporates a view port made of fused silica (KU1), which has a transmission of 65% for the 175 nm VUV light. The view port is 80 mm in diameter and 30 mm in thickness. The Frisch grid and the anode electrodes are an array of wires strung on a ring to provide transparency for scintillation light. A VUV photomultiplier tube (PMT; FEU-39A, Russia) with a spectral sensitivity of 160–600 nm and with a quantum efficiency of 10–15% was placed on the view port to detect the 175 nm scintillation light. Another type of a photomultiplier tube (FEU-143, Russia) with the nanosecond time resolution and spectral sensitivity of 300–650 nm was used for scintillation counting utilizing scintillating fibers to collect a small portion of scintillation light (single-photon counting method) to measure decay times in the pure xenon and in xenon-methane mixture.

B. Gas Purification System

A new gas system has been built to purify methane separately from xenon. Fig. 2 shows a schematic view of the methane purification system. Before assembling the plumbing, all parts of the system and ionization chamber were ultrasonically cleaned with petroleum and then with ethanol in an ultrasonic bath. The system was baked at 180°C for 90 hours and pumped out with both activated carbon (8) and zeolite cryogenic (9) pumps. When the level of evacuation was high, the system was pumped out with a magneto-discharge pump NMD 0,16-1 (Russia). A mass spectrometer (Residual Gas Analyzer, RGA200, Stanford Research Systems) allowed us to detect and analyze the outgassing from the lines of the methane purification system and from the ionization chamber (1). Electronegative impurities such as O_2 were below detectable level. The system was leak checked with helium. The xenon gas was first purified with a calcium high-temperature getter at 650°C and then transferred through a titanium getter. As mentioned above, the purity of the xenon gas was constantly monitored by measuring the ionization signals at various cathode-anode distances. The purity of the xenon gas determined by electron lifetime was equal to 3 ms at an electric field of $\mathrm{E} = 100$ V/cm. We bought a cylinder with methane gas with an initial purity of 99.9%. The methane gas was recycled several times from cylinder (3) to cylinder (4) through a high pressure Oxisorb cartridge (Messer Greisheim) (6) by cooling down the cylinders alternately with liquid nitrogen. The purification system was pumped out and baked after each cycle. The system and the chamber were tested with the mass spectrometer after each purification cycle on presence of known electronegative impurities, which were not observed. Finally, the methane gas was frozen with liquid nitrogen at cylinder (4) and the residual vapor pressure was pumped out with the zeolite cryogenic pump. The gas mixture was prepared in the system including only cylinder (4), the pipelines and the ionization chamber (1). To obtain the desired concentration, the volumes of cylinder (4), the ionization chamber (1) and the pipelines, as well as the compressibility factor of the gas, were taken into account. A vacuum gauge sensor (2) with the pressure range of (0–1 atm) was used to evaluate the amount of methane introduced into the xenon gas.

C. Experimental Method

Scintillation light was detected with the PMT (FEU-39A) and recorded with a digital oscilloscope (LeCroy LT344, 500 Ms/s). Scintillation decay time characteristics of the xenon and xenon-methane mixture were measured with a single-photoelectron technique [22]. The second PMT (FEU-143),

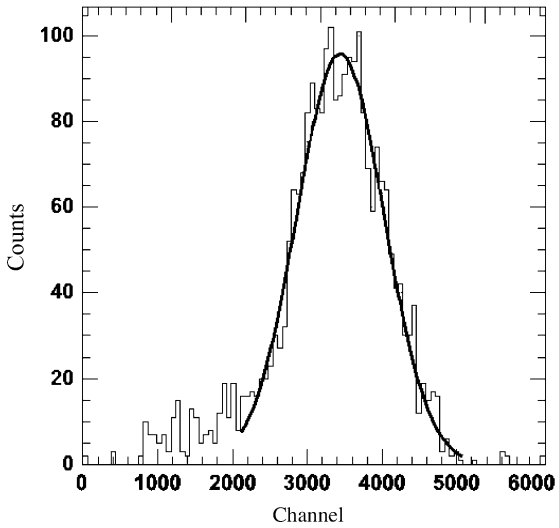

Fig. 3. A Gaussian pulse height distribution of a scintillation signal obtained in $\mathrm{Xe} + \mathrm{CH_4}(0.05\%)$ mixture at a pressure of 21 atm and at $\mathrm{E/N} = 0.36 \times 10^{-17}$ Vcm^2.

coupled with the chamber optical window through the scintillating fibers, recorded a signal in coincidence with the signal detected with the first PMT (FEU-39A). A scintillation photon was reemitted inside of the fiber core with p-terphenyl and POPOP when it entered a scintillating fiber. Since the photoelectron collection efficiency of the fiber system is low, the time distribution of the single photoelectron signals reflects a decay curve of scintillation.

The ionization charge was collected and integrated with a charge sensitive amplifier and sent to the digital oscilloscope. The data were recorded and stored in the computer. The gain of the PMT (FEU-39A) was proven to be stable during the measurements by using a green LED.

The experimental data were analyzed with a Data Analysis Framework ROOT Software (CERN) [23].

III. Results & Discussion

A. Measurements of Scintillation Light Yield

Scintillation light, ionization yield and scintillation decay times have been measured in the high pressure xenon and xenon doped with methane (0.05%). A typical Gaussian pulse height distribution of scintillation light is shown in Fig. 3. The peak channel is proportional to the number of scintillation photons.

A typical scintillation pulse in the pure xenon is shown in Fig. 4(a). The scintillation and electroluminescence pulses are clearly seen in the figure. The electroluminescence coincides in time with the ionization pulse.

In order to estimate the purity of methane, scintillation light and ionization yield were measured in the pure methane at pressures of 2.4 atm and 8.8 atm. The signals are shown in Fig. 4(b). Unlike the pure xenon, neither scintillation light nor electroluminescence was observed in the xenon-methane mixture (0.05%).

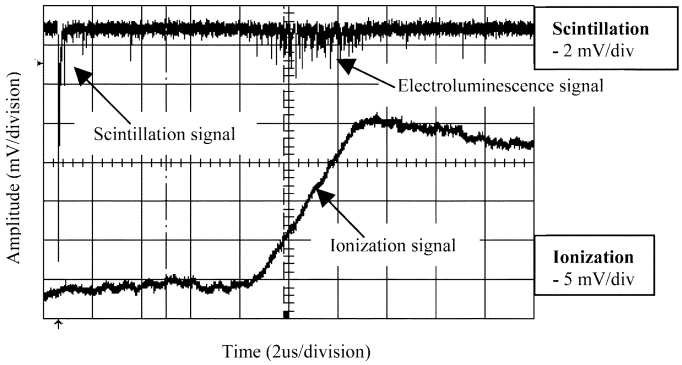

Fig. 4(a). Ionization signal, scintillation and electroluminescence signals in pure xenon at a pressure of 21 atm.

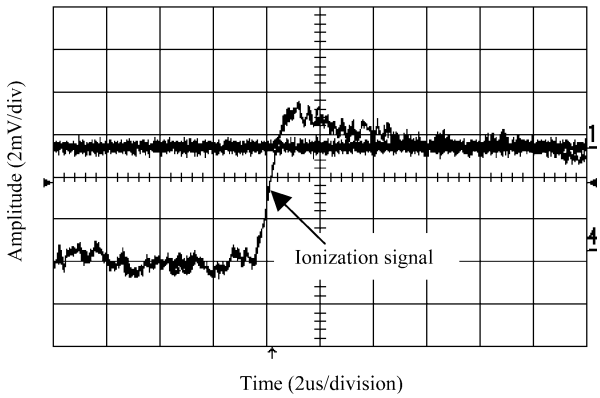

Fig. 4(b). Ionization signal in the pure methane at a pressure of 8.8 atm [neither scintillation light nor electrolumenescence was observed with a PMT (FEU-39A)].

The following processes take place in methane.

$$\mathrm{e}, \alpha + \mathrm{CH_4} \rightarrow [\mathrm{CH_4^*}] \quad \text{(a)}$$
$$[\mathrm{CH_4^*}] \rightarrow \mathrm{CH^*} + \mathrm{H_2} + \mathrm{H} \quad \text{(b)}$$
$$[\mathrm{CH_4^*}] \rightarrow \mathrm{C^*} + 4\mathrm{H} \quad \text{(c)}$$
$$[\mathrm{CH_4^*}] \rightarrow \mathrm{H^*} + \mathrm{CH} + \mathrm{H_2} \quad \text{(d)}$$
$$[\mathrm{CH^*}] \rightarrow \mathrm{CH} + \mathrm{h}\nu(\lambda = 431.4\mathrm{nm}) \quad \text{(e)}$$
$$\mathrm{C^*} \rightarrow \mathrm{C} + \mathrm{h}\nu(\lambda = 165.7\mathrm{nm}, 175.1\mathrm{nm}) \quad \text{(f)}$$
$$\mathrm{H^*} \rightarrow \mathrm{H} + \mathrm{h}\nu(\lambda = 121.5\mathrm{nm}). \quad \text{(g)}$$

All three $[\mathrm{CH_4^*}]$ decay routes (b), (c), (d), are possible following high-energy impact excitation of a $\mathrm{CH_4}$ molecule (process a) with alpha particles or with electrons in the region between the Frisch grid electrode and the anode. These decays will generate the CH* 431.4 nm emission (e) and also produce a small amount of H Lyman-α at 121.5 nm (g) and CI line emission at 165.7 nm and 175.1 nm (f). The Lyman-α will be promptly absorbed by methane causing methane dissociation with only a small amount of reemission from CH* and $\mathrm{CH_2}$ at 390 nm, 431 nm and 500 nm respectively [24], [25]. It should be noted that the excited states of CH* as well as C* states can be readily quenched with $\mathrm{CH_4}$ molecules that leads to loss of scintillation light as the radiative lifetime of CH* state is of the order of 500 ns and

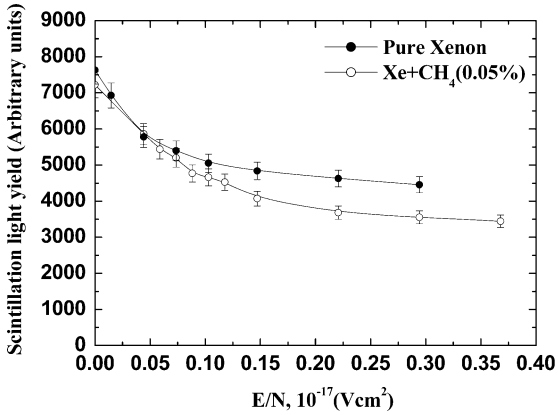

Fig. 5. Scintillation light yield as a function of the reduced electric field (E/N) at a pressure of 21 atm.

its quenching rate constant is $(2.24 \pm 0.07) \times 10^{-11}$ cm^{3}s^{-1} for $\mathrm{CH}(\mathrm{A}^2\Delta) + \mathrm{CH}_4 \rightarrow \mathrm{CH} + \mathrm{CH}_4^* \rightarrow$ CH+non-radiative product process [26].

The plot of scintillation light measured in the pure xenon and xenon-methane (0.05%) mixture at a pressure of 21 atm as a function of the reduced electric field is shown in Fig. 5.

Scintillation light in the pure xenon decreases by 42% when the electric field increases to a value of 0.3×10^{-17}Vcm^2. The pure xenon and xenon-methane mixture have the same scintillation light value at the electric field of 0.05×10^{-17} Vcm^2 and light drops in relative to the pure xenon when the electric field is continually increased. Scintillation light in the xenon-methane admixture decreases by 23% in relative to light in the pure xenon at the electric field of 0.3×10^{-17} Vcm^2.

Scintillation light in xenon is emitted due to excitation $\mathrm{Xe}_2^* \rightarrow 2\mathrm{Xe} + \mathrm{h}\nu$ and recombination processes $\mathrm{Xe}_2^+ + \mathrm{e} \rightarrow \mathrm{Xe}_2^*$ that were described elsewhere [27]. Thus, scintillation light decrease in the pure xenon with increasing the electric field is due to the decrease of recombination processes. Scintillation light in the xenon-methane mixture decreases in relative to light in the pure xenon due to the quenching mechanism of xenon atoms and xenon molecules (dimmers) with methane molecules as the mechanism was described in works [28], [29].

B. Measurements of Scintillation Decay Times

Scintillation decay times in the pure xenon and xenon-methane mixture (0.05%) have been measured. It was shown in previous work that decay times in the pure xenon decrease as the medium density increases [30]. As a charged particle passes through the gas it creates excitation and ionization along its path. The resulting positive ion and free electron called an ion pair recombine, if there is no electric field, forming a neutral atom. The decay of recombination scintillation is mainly attributed to initial (geminate) recombination that takes place in the high-density core of electrons, which shortly recombine with ions. The next more slow process takes place outside of the core and is called volume recombination. This type of recombination is accounted to electrons Brownian motion, which leave the core and become thermalized far away from the core where the Coulomb attraction is negligible. Scintillation decay times decrease with the increase of the gas density

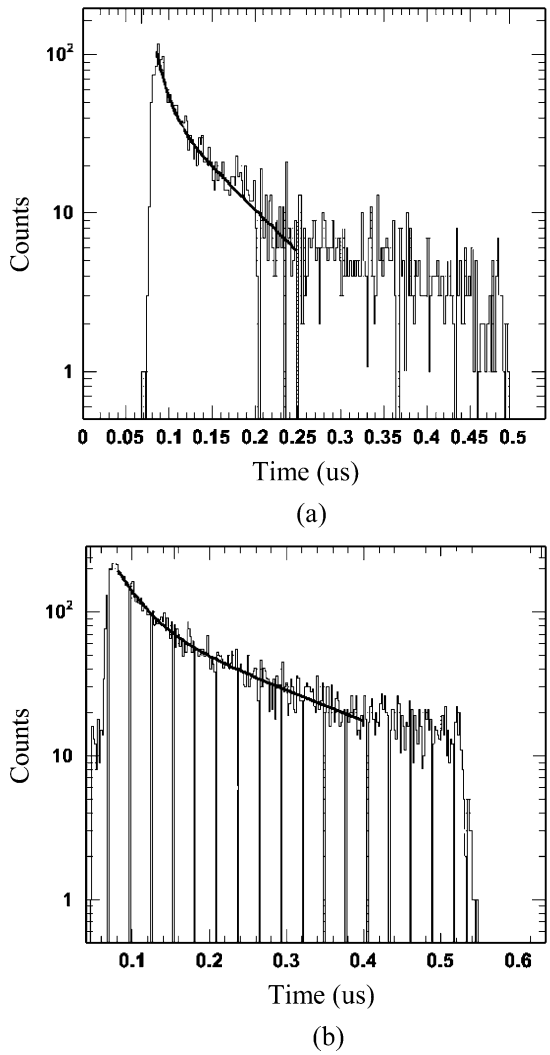

Fig. 6. Scintillation decay times in the pure xenon (a) and in xenon-methane (0.05%) (b) measured at a pressure of 21 atm and at an electric field of $\mathrm{E/N} = 0.01 \times 10^{-17}$Vcm^2.

because volume recombination contributes significantly less. Decay times decrease substantially when the electric field is applied. This reduction is also more likely related with the decrease of volume recombination processes. Though, the applied electric field erodes the core of the electrons, the initial (geminate) recombination still takes place contributing to scintillation light. It enabled us to measure scintillation decay time characteristics in the pure xenon and in xenon-methane mixture. The measurements of scintillation decay times in the xenon-methane mixture (0.05%) have never been done before. Scintillation decay times measured at a pressure of 21 atm are shown in Fig. 6.

The distributions were fitted with a sum of two exponential functions (slow components). Here we give our preliminary analysis without taking into account the fluorescence decay time of p-terphenyl and POPOP (≈ 1.8 ns) in the fiber. It is known that recombination luminescence contributes a large part to the slow $^3\Sigma_\mathrm{u}^+$ states and rather a small part to the fast $^1\Sigma_\mathrm{u}^+$ states [22]. Decay times of the slow component as a function

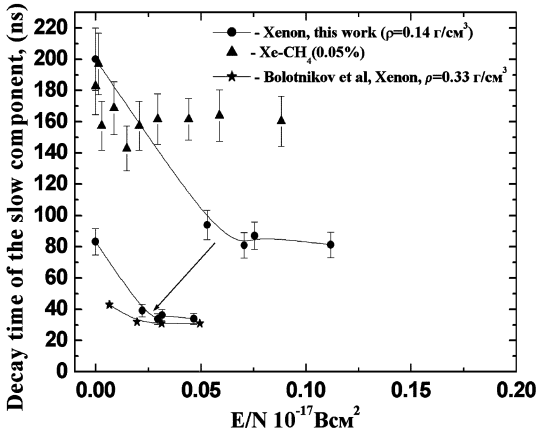

Fig. 7. Decay time of the slow component as a function of the reduced electric field (E/N) in the pure xenon and xenon-methane (0.05%) at a pressure of 21 atm.

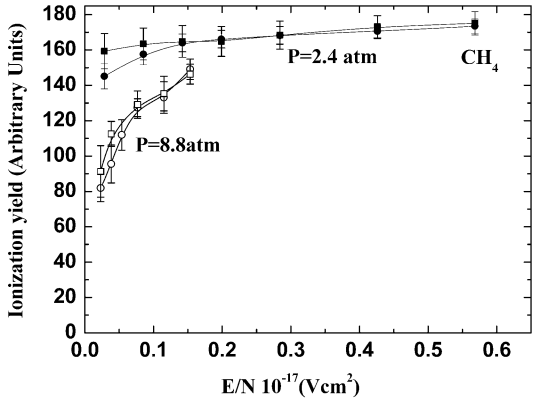

Fig. 8. Ionization yield in the pure methane at pressures of 2.4 atm and 8.8 atm as a function of the reduced electric field (E/N).

of the reduced electric field (E/N) in the pure xenon and in xenon-methane mixture (0.05%) are shown in Fig. 7.

The slow component decreases considerably in the pure xenon as the electric field strength increases. The slow component decreases from 200 ns to 94 ns when the electric field increases from $\mathrm{E/N} = 0$ to $\mathrm{E/N} = 0.05 \times 10^{-17}\mathrm{Vcm}^2$ and becomes saturated in the range of 80-85 ns. This decrease is related with the decrease of volume recombination.

The experimental data by Bolotnikov *et al.* [30] were obtained in the pure xenon at a density of 0.33 g/cm^3 ($\sim$45 atm) (solid line, stars). We assume that if decay time of the slow component decreases proportionally with the increase of the gas density then our data (solid line, black circles) will be scaled with the data obtained by Bolotnikov *et al.* at their density.

We found that the behavior of the slow component in the xenon-methane mixture is completely different from that one in the pure xenon. The slow component decay seems to stay approximately constant when the electric field increases. The electrons are still cold in this range of the small electric fields due to loss of the average energy when they collide with methane molecules. The energy of the electrons in the $\mathrm{Xe{-}CH_4}(0.05\%)$ mixture at $(0{-}0.08 \times 10^{-17}\mathrm{Vcm}^2)$ is approximately equal to the average energy of the electrons in the pure xenon at $\mathrm{E/N} = 0$, which results in recombination of the electrons and thus in the decay time delay of the slow component in the $\mathrm{Xe{-}CH_4}(0.05\%)$. We presume that the decay time in the $\mathrm{Xe{-}CH_4}(0.05\%)$ mixture will be of the same order with the decay time in the pure xenon at high electric fields [31].

C. Measurements of Ionization Yield

Ionization yield in the pure methane at pressures of 2.4 atm and 8.8 atm as a function of the reduced electric field (E/N) has been measured. The plot of ionization yield in the pure methane versus the reduced electric field is shown in Fig. 8. As it was pointed out before, we measured ionization yield in the pure methane at various distances between the electrodes to estimate the purity of the gas. Ionization yield was measured at a 1.5 cm distance between the electrodes (black and white squares) and at a 5 cm distance between the electrodes (black and white circles). It is clear from the figure that ionization yields both at a pressure of 2.4 atm and at a pressure of 8.8 atm are practically independent of the distance between the electrodes. There is only a slight dependence at the range of the electric field of $0 - 0.1 \times 10^{-17}\mathrm{Vcm}^2$ that could be negligible taking into account the amount of methane we added into the xenon gas. Consequently, the purity of the methane was relevant, from the point of view of electron attachment processes, to use it in the xenon gas as an admixture.

When the pressure is increased from 2.4 atm to 8.8 atm ionization yield drops substantially by factor of 1.7 at an electric field of $0.03 \times 10^{-17}\mathrm{Vcm}^2$ and it increases when the electric field is continuously increased. Two-body (h) and three-body (i) recombination processes take place in methane:

$$\mathrm{CH_4^+ + e^- \rightarrow products} \quad \text{(h)}$$

$$\mathrm{CH_4^+ + e^- + CH_4 \rightarrow products.} \quad \text{(i)}$$

The process (h) is associated with two-body dissociative recombination, which is independent of pressure.

On the contrary, three-body recombination (i) causes large dependence on pressure [31]. Thus, that noticeable drop of the ionization yield with the pressure increase is related with three-body recombination process.

Ionization yield in the pure xenon and xenon doped with a 0.05% concentration of methane is plotted in Fig. 9. Ionization yield in the pure xenon increases with increasing of the electric field that is attributable to the reduction of recombination processes. Ionization yield decreases over the whole range of the electric field when methane is added into the xenon gas.

Since we added methane into the xenon gas we decided to estimate the purity of the xenon-methane mixture. Fig. 10 shows ionization yield in the xenon-methane mixture (0.05%) measured at a 1.5 cm distance between the electrodes (black circles) and at a 5 cm distance between the electrodes (white circles).

It is clearly seen from the figure that ionization yield drops at the low electric fields when the distance between the electrodes is increased from 1.5 cm to 5 cm and it subsequently increases to the same charge value with the xenon-methane (1.5 cm electrode distance) when the electric field is steadily increased to 0.25×10^{-17} Vcm^2.

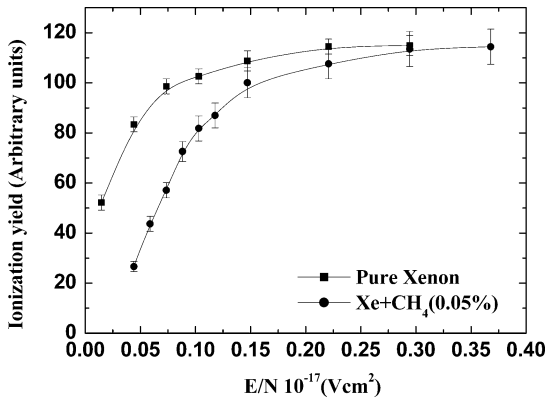

Fig. 9. Ionization yield as a function of the reduced electric field (E/N) at a pressure of 21 atm.

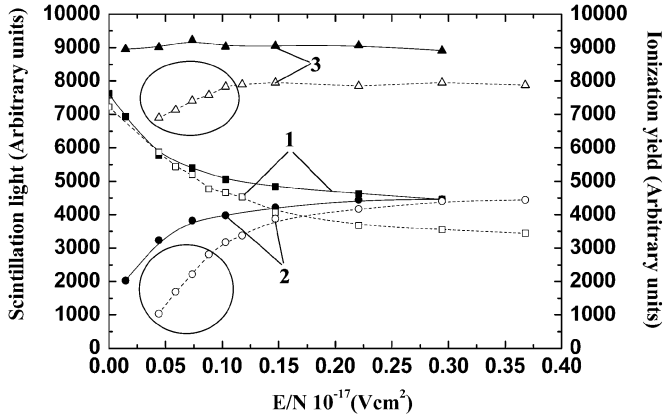

Fig. 11. Scintillation light yield (1), ionization yield (2) and their sum (3) in the pure xenon (solid lines) and xenon-methane mixture (0.05%) (dash lines) at a pressure of 21 atm as a function of the reduced electric field (E/N).

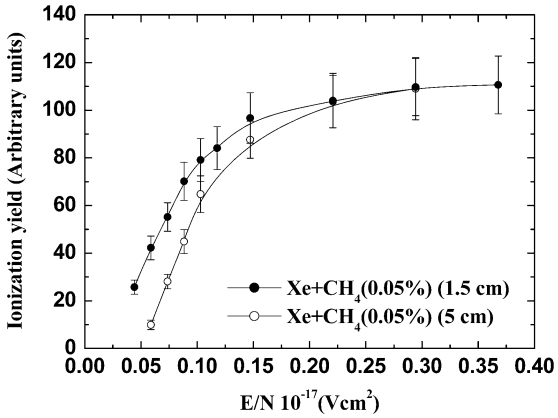

Fig. 10. Ionization yield in the xenon-methane mixture (0.05%) at a distance of 1.5 cm (black circles) and 5 cm (white circles) between the electrodes as a function of the reduced electric field (E/N) at a pressure of 21 atm.

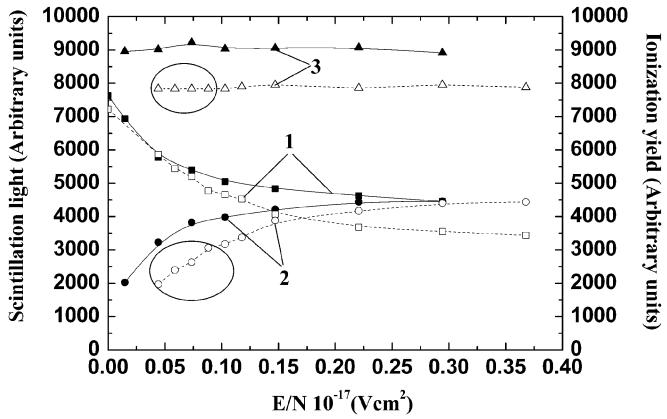

Fig. 12. Scintillation light yield (1), corrected ionization yield (2) and their sum (3) in the pure xenon (solid lines) and xenon-methane mixture (0.05%) (dash lines) at a pressure of 21 atm as a function of the reduced electric field (E/N).

This typical loss of the electrons is related to presence of some electronegative impurities, which were not evident in the pure xenon and pure methane before making the mixture. Consequently, the decrease of ionization yield in the xenon-methane mixture in relative to the pure xenon can be attributed to a significant increase of the recombination processes and electron attachment with some electronegative impurities. According to a mechanism postulated by Bloch and Bradbury [32] the probability of electron attachment with electronegative impurities increases especially in mixtures with polyatomic gases in three body reactions. The experiments on measurements of ionization yields in $\mathrm{Ar} + \mathrm{CH_4}$ and $\mathrm{Ar} + \mathrm{NH_3}$ showed that the probability of electron attachment increases with increasing of the concentration of molecular admixtures [33].

Scintillation light yield (1), ionization yield (2) and their sum (3) in the pure xenon (solid lines) and xenon-methane mixture (0.05%) (dash lines) as a function of the reduced electric field (E/N) are plotted in Fig. 11. It is clearly seen that the sum in the pure xenon is almost constant with increasing of the electric field strength. The constant behavior indicates a right balance of energy between scintillation and ionization yields. Unlike the pure xenon, the sum gradually decreases in the xenon-methane mixture (0.05%) at low electric fields and becomes constant at the electric field over 0.10×10^{-17} Vcm^2.

This maybe attributed with electron attachment with some unknown to us impurities, which have been observed in the measurements of ionization yield in the xenon-methane (0.05%) mixture measured at two various electrodes distances (see Fig. 10).

The experimental points of the ionization signal in the Xe-$\mathrm{CH_4}$ mixture were normalized to the experimental points obtained at an electric field over $\mathrm{E/N} = 0.15 \times 10^{-17}$ Vcm^2 in accordance with electron attachment length (see Fig. 11). The normalized points of the ionization signal are plotted in Fig. 12. Thus, we show that the constant behavior of the sum (3) in the whole range of the electric fields clearly indicates the right balance between scintillation and ionization yields for the xenon-methane mixture without loss of electrons due to electron attachment processes. Decrease of the sum in the xenon-methane mixture in relative to the sum in the pure xenon is due to quenching of xenon atoms and xenon molecules (dimmers) with methane molecules that leads to decrease of the 175 nm VUV scintillation light. We presume that the sum of scintillation and ionization yields for the xenon-methane

mixture will continue to decrease in relative to the pure xenon with increasing of the methane concentration.

IV. Conclusion

Scintillation light, scintillation decay times and ionization yield in the pure xenon and xenon doped with a 0.05% concentration of methane at a pressure of 21 atm as a function of the reduced electric field (E/N) have been measured. The measurements have revealed that both scintillation and ionization yield decrease in the whole range of the electric fields when methane is added into the xenon gas. The principal reason of scintillation light loss can be caused by the de-excitation of xenon atoms and xenon molecules (dimmers) with methane molecules.

Scintillation decay times (the slow component) decrease in the pure xenon as the electric field strength increases. This obvious decrease of the slow component with the increase of the electric field is related to reduction of volume recombination. Scintillation decay times seems to stay approximately constant in the whole range of the electric fields and are observed to be longer when methane is added into the xenon.

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