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**A LIQUID IONIZATION CHAMBER USING TETRAMETHYLSILANE**

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## A LIQUID IONIZATION CHAMBER USING TETRAMETHYLSILANE

### ABSTRACT

First results with a liquid ionization chamber using Tetramethylsilane (TMS) are presented. A stack of iron plates was tested with cosmic ray muons and the charge output for minimum ionizing particles was measured.

## BETRIEB EINER IONISATIONSKAMMER MIT TETRAMETHYLSILAN

### ZUSAMMENFASSUNG

Es werden erste Ergebnisse über den Betrieb einer Ionisationskammer gegeben, die als Medium Tetramethylsilan (TMS) benutzt. Ein Satz aus Edelstahlplatten wurde mit kosmischen Myonen getestet und die Ladungsausbeute für minimal ionisierende Teilchen bestimmt.

## 1. INTRODUCTION

In high energy and nuclear physics the use of liquid ionization chambers with electron charge transport was restricted up to now to cryogenic liquids only. Liquid Xenon and Argon are widely used; for a recent review see C. Brassard<sup>1)</sup>. There are two obvious reasons to use these liquids:

- 1) In the liquid noble gases Ar, Xe, Kr, the charge output for minimum ionizing particles traversing the liquid is relatively high, and
- 2) they are cryogenic. The low temperature facilitates cleaning the liquids and maintaining the contamination below the necessary level of 1 ppm O<sub>2</sub> equivalent.

Nevertheless, for practical use, cryogenic liquids have severe drawbacks. The necessity of a cryostat in many cases prevents a compact detector design, and the cryogenic operation makes handling very cumbersome. Hence, for many applications, it is very desirable to have liquid ionization chambers operating at room temperature.

First hints of the presence of free electrons in a room temperature liquid were obtained by Tewari and Freeman<sup>2)</sup> for neopentane in 1968. Independently, and nearly at the same time, Schmidt and Allen<sup>3)</sup> observed a high mobility and a high yield of charge carriers in tetramethylsilane. Both effects were interpreted as being due to free electrons carrying the charge.

Stimulated by these observations, we intend to use warm liquids in large scale high energy detectors, for instance in calorimeters. Here we report on the first operation of a small test chamber using tetramethylsilane (TMS). The size of the chamber was chosen to represent the typical dimensions of a tower in a calorimeter, namely approximately  $10 \times 10 \text{ cm}^2$  and seven planes ganged together in the longitudinal direction. The test was done with cosmic ray muons.

## 2. ELECTRON CHARGE TRANSPORT IN LIQUIDS

When searching for candidates for free electron charge transport among the non-polar liquids, it is useful to look up the mobility and the electron affinity. These two quantities are extensively studied and give a good indication for free-moving electrons if the mobility is high and the electron affinity is negative.

### 2.1 Mobility

The mobility is defined as  $\mu = v_D/E$ , where  $v_D$  is the drift velocity and  $E$  the electric field. A compilation of mobilities for various room temperature liquids has been given by W.F. Schmidt<sup>4)</sup>. An extract is shown in Fig. 1, where the mobilities are plotted

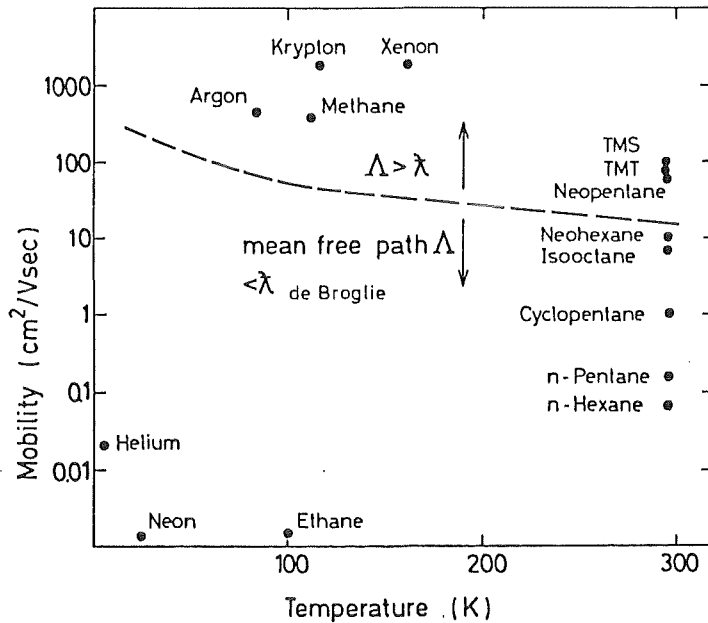


FIG. 1:  
Mobility in several liquids. The dashed line separates the region between free and bound electrons.

versus the temperature at which the mobility was measured. One observes high mobilities for the noble gases and methane and a large scale of mobilities for room temperature liquids. A plausible condition for electrons to be able to move freely in a liquid is that the mean free path  $\Lambda$  between elastic collisions with the surrounding molecules be greater than the de Broglie wavelength of thermalized electrons. In the theory of Cohen and Lekner<sup>5)</sup> on free electron charge transport, a connection between  $\Lambda$  and  $\mu$  is given by

$$\mu = \frac{2e}{3} \sqrt{\frac{2}{\pi m kT}} \Lambda ,$$

e and m are the electron charge and mass, respectively, k is the Boltzmann constant and T the temperature. Inserting in this formula the de Broglie wavelength for  $\Lambda$ , one obtains a boundary limit as indicated by the dashed line in Fig. 1. Above the line, we expect free electrons to be present, whereas below it, the electrons are supposed to be bound in localized states. In these latter liquids, the electrons are immobilized in a physical void or by polarization forces and, therefore, move only slowly.

## 2.2 Electron Affinity

The second indication for free electrons is a negative electron affinity,  $V_o$ . This quantity is also interpreted as the potential of a conduction band of an "extended electron state" in the liquid, similar to the conduction bands in metals. It is usually determined as the difference of the work function  $\phi$  of a photocathode in contact with the liquid and with the vacuum:

$$V_o = \phi_{\text{liqu.}} - \phi_{\text{vac.}}$$

A negative value is an indication that the liquid does not attract electrons and, therefore, should be a hint for free conduction electrons. A collection of  $V_o$ -values for typical liquids as compiled by W.F. Schmidt<sup>6)</sup> is given in Table 1. One observes that a high mobility corresponds in many cases with a negative affinity. When scanning through the literature more thoroughly, one finds as candidate, among the room temperature liquids, spherical molecules with the following tetrahedral structure:  $X(\text{CH}_3)_4$ . On the corners of the tetrahedron, there are four methyl radicals and, in the center, a four valent atom. So far, molecules with  $X = \text{C}$  (neopentane), Si, Ge, and Sn have been investigated. Neopentane has a boiling point of 9°C and is, therefore, not a real room temperature liquid. Ge is too expensive to be used in large quantities. Hence, for practical applications, TMS and tetramethyltin (TMT) remain. A few interesting properties of these two liquids are listed in Table 2. Due to its toxic character, we have abandoned TMT and have chosen TMS for these tests.



TABLE 1: Values for Electron Affinity  $V_0$  for Various Liquids

Liquid	T [K]	$V_0$ [eV]
Helium	4	+ 1.05
Neon	25	+ 0.67
Argon	84	- 0.20
Krypton	116	-0.40
Xenon	161	-0.67
CH <sub>4</sub>	100	- 0.25
TMS	295	- 0.57
Neopentane	295	- 0.43
Cyclopentane	295	- 0.28
Isooctance	295	- 0.18
N-Pentane	295	0.00
N-Hexane	295	+ 0.04

TABLE 2. Physical Properties of TMS and TMT

Property	Si(CH <sub>3</sub> ) <sub>4</sub>	Sn (CH <sub>3</sub> ) <sub>4</sub>
Density (g/cm <sup>3</sup> )	0.65	1.31
Boiling point at 1 bar	26.5 <sup>0</sup> C	78 <sup>0</sup> C
Melting Point	-100 <sup>0</sup> C	-54 <sup>0</sup> C
Dielectric Const. (20 <sup>0</sup> C)	1.92	~ 2.3
Rad. Length $X_0$ (cm)	52.2	9.2
Interact. Length $\lambda_0$ (cm)	130	87
$E_p$ min. ioniz. (MeV/cm)	1.03	1.72
Flammable	yes	yes
Toxic	no	yes

### 2.3 Drift Velocity

The drift velocity for TMS is shown in Fig. 2, where it is plotted versus the applied field as measured by U. Sowada<sup>7)</sup>. It is linear up to 20 kV/cm, giving a constant mobility. At higher fields, the mobility decreases due to inelastic collisions, since the electrons begin to lose the energy which they pick up from the field between collisions. We find that a field of 20 kV/cm can be realized without major complications. At this value, the drift velocity is  $2 \times 10^6$  cm/sec which corresponds to a drifttime of 50 nsec/mm. This is a factor 4 faster than in LAr where the corresponding value is 200 nsec/mm.

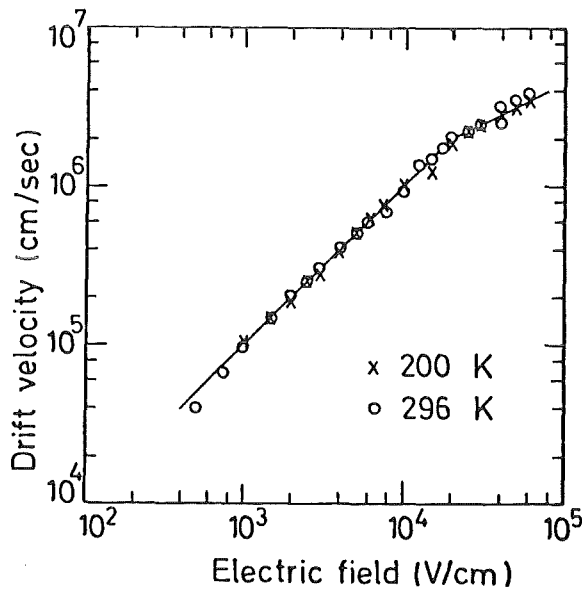


FIG. 2:  
The drift velocity in  
TMS (Ref. 7)

### 3. TEST APPRATUS

#### 3.1 Ionization Chamber

The ionization chamber is shown schematically in Fig. 3. A small stack of electrodes was chosen with dimensions which represent a typical tower arrangement in a high energy calorimeter. In order to avoid poisoning of the liquid, the only materials used for construction are stainless steel, polyethylene, teflon rods and ceramic washers. The materials used as seals are indium and PTFE-like compounds (Kalrez<sup>8</sup>).

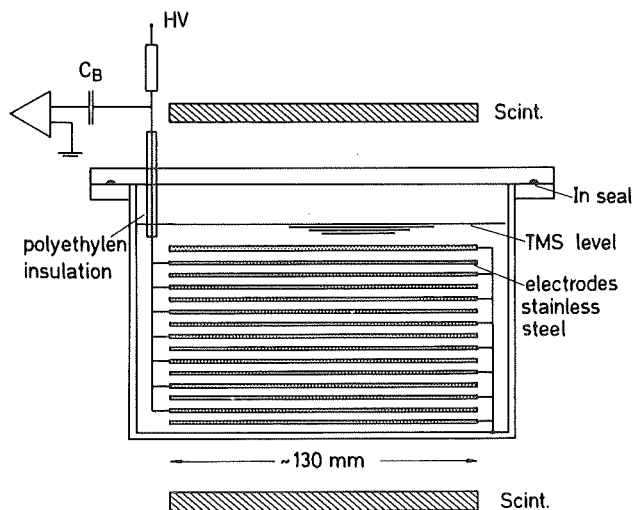


FIG. 3:  
Arrangement of the chamber

The stack is built up of 7 pairs of stainless steel plates of hexagonal shape, 1.5 mm thick and about 130 mm across. The gap height is 3.5 mm, resulting in a total of 4.9 cm liquid to be transversed by a cosmic muon. The total amount of liquid contained in the chamber is approximately 3 l.

Before assembly, the iron plates, the chamber enclosure and all parts of the support structure are electropolished and cleaned in an ultrasonic bath of methanol. After assembly, the chamber is baked out by 90°C hot water flowing through a coil soldered onto the outside of the chamber body. Bake-out time is about 20 h at a pressure of about  $10^{-4}$  mbar.

The high voltage plates are ganged together and the signal fed to a common preamplifier. Below and above the chamber, scintillators serve to trigger on cosmic muons.

### 3.2 Electronic Circuit

The readout electronics is shown in Fig. 4. The low noise charge amplifier is placed within a few cm of the chamber body and shielded in a copper housing. The total capacitive load corresponds to 800 pF and is rather high. We, therefore, use a transformer to optimize the matching to the amplifier input capacitance. An amplifier of the CELLO LAR-calorimeter was taken. For integration, we use a time constant of 800 nsec. After the input stage, the signals are amplified and processed by an integrating and differentiating circuit (ORTEC 572) with both time constants approximately 1  $\mu$ s. Finally, the signals are analyzed in a multi-channel analyzer which is triggered by NIM-based electronics.

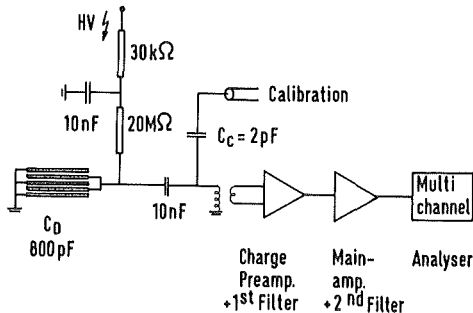


FIG. 4:  
Principle of the electronic circuit

Calibration is performed with a step voltage fed via a capacitor of 2.2 pF onto the detector capacitance. The rms noise corresponds to 0.8 fC. The high voltage is filtered carefully by RC networks, the later being located directly at the chamber entrance. As high voltage power supply we use a Spellmann RHSR 25 PN.

### 3.3 Purification System

The purchased liquid has a purity of only 99.9% and has been in contact with air. A first step in purification, therefore, is vacuum distillation. The system used is shown in its principal parts in Fig. 5. It is designed to purify quantities of 5 l of liquid. All parts in contact with the TMS liquid or vapour are made of stainless steel.

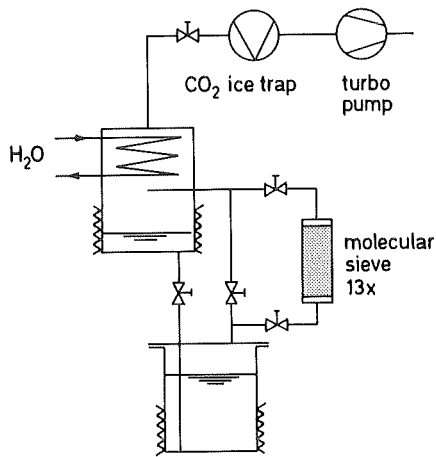


FIG. 5:  
Principle of the purification system

TMS Purification System

In the first step, the TMS is evaporated from the lower vessel into the upper one where it is recondensed by a water cooled heat exchanger coil. Part of the vapour phase, probably mainly air, is pumped away by a turbo pump. This is done very cautiously by observing in a glass ice trap whether TMS begins to condense. The vacuum distillation is repeated until no more residues are observed in the evaporation vessel. The residues normally found look different for different batches of liquid. But typically, in the first two distillation passes, we observe an oily liquid, maybe water, remaining on the bottom and dust-like particles on the walls. We find three distillation passes to be sufficient. We let the last one proceed very slowly with a temperature difference of only 2<sup>0</sup>C between the lower and the upper vessel.

After vacuum distillation, the TMS vapour passes through two chimneys filled with molecular sieves (zeolite) of type 13X. We observe electric signals already after the first passage. Normally two passes are sufficient to give full signal output.

The oxygen and moisture contamination is below the sensitivity of the monitors, which - in both cases - is approximately 1 ppm. At room temperature, the vapour pressure of TMS is around 0.9 bar. In order to avoid air leaking into the system, we apply a slight overpressure in the measuring cell by pressurizing with purified argon gas.

#### 4. RESULTS

We have measured the charge output for cosmic ray muons with respect to the applied voltage. The yield is shown in Fig. 6. As usual,  $G$  is defined as the yield of escaped electrons per 100 eV deposited ionization energy. For comparison, the yield obtained by H. Jungblut<sup>11)</sup> is given for TMS and for liquid methane. Jungblut made his measurements in a small cell of a few cubic centimeters. Our findings agree well with his results.

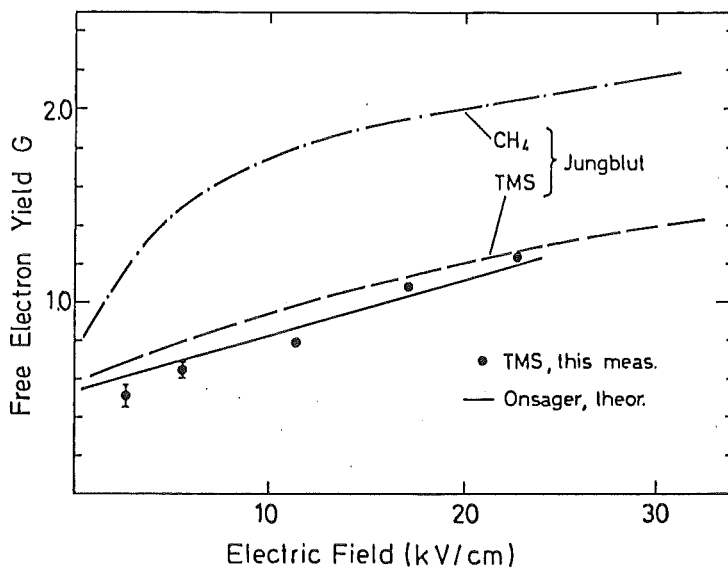


FIG. 6: Number of free electrons  $G$  yielded for 100 eV deposited ionization energy

The straight line in the figure represents the prediction of Onsager's theory<sup>12)</sup>. Onsager assumes geminate recombination of the electrons with their parent positive ion. After being thermalized by inelastic collisions with the surrounding molecules, the Coulomb attraction recombines the electron with its positive parent ion. For low fields, Onsager's calculation predicts a linear dependence with a slope-to-intercept ratio equal to  $e^3/2\epsilon k^2 T^2$ , where  $e$  is the electron charge and  $\epsilon$  the dielectric constant of the medium. Our result agrees well with this prediction. It is due to the Boltzmann factor that, for room temperature liquids, high fields are needed to reach saturated charge collection.

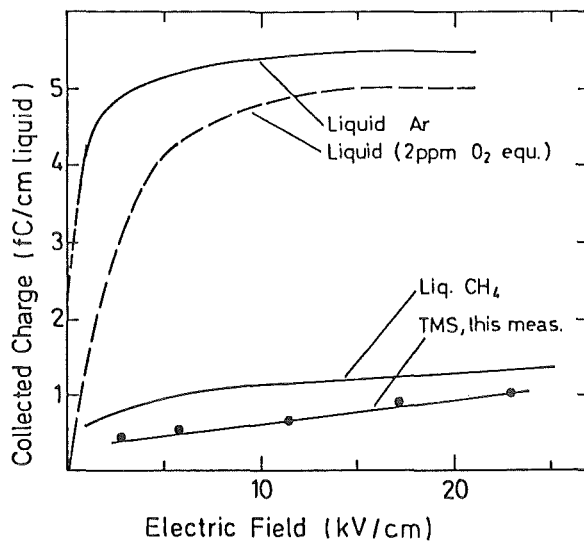


FIG. 7:

Collected charge for minimum ionizing particles traversing 1 cm of liquid. The straight line through the measured points corresponds to Onsager's theory.

In Fig 7, the charge output is plotted in a more practical form given for a minimum ionizing particle traversing 1 cm of liquid. At a typical voltage of 20 kV/cm about 1 fC/cm are collected. For comparison, the yield in LAr is given together with a typical purity level in large detector systems assumed to be 2 ppm O<sub>2</sub> equivalent. One finds that, for a given detector configuration, the pulse height in TMS is reduced by a factor 4-5 as compared to LAr.

We could typically raise the high voltage to 22 ... 28 kV/cm without observing additional noise in the preamplifier. Beyond these values, small discharges start to augment the preamplifier noise. We have not yet localized these discharges in the liquid, the decoupling capacitor or the feedthrough cable

We have repeated the measurements with two other fillings of liquid. During the measurements, the cell was decoupled from the purification system. For a period of two weeks, we could not observe any change in charge collection within our accuracy of about 5%.

## 5. CONCLUSION

In a small test chamber, we could detect cosmic ray muons with rather simple methods and obtained reasonable charge output. For the treatment of the chamber, only normal methods used in vacuum technology were applied. The purification of the liquid with a few vacuum distillations and a passage over molecular sieves is also relatively simple and straightforward. The charge output is reduced by a factor 4 ... 5 with respect to liquid argon. But for many applications - such as high energy calorimetry, where the energy deposition is dense - this is no severe hindrance. We, therefore, feel confident that warm liquid ionization chambers will add a new tool to high energy instrumentation.

We wish to express our appreciation of the skillful help of R. Dittmann in setting up the apparatus. We would like to thank Profs. G. Flügge and B. Zeitnitz for continuous interest and encouraging support.



REFERENCES

- 1) C. Brassard, Nucl.Instr. and Meth. 162 (1979) 29
- 2) P.H. Tewari and G.R. Freeman, J. Chem.Phys. 49 (1968) 4394
- 3) W.F. Schmidt and A.O. Allen, J. Chem.Phys. 50 (1969) 5037
- 4) W.F. Schmidt, Can. J. Chem. 55 (1977) 2197
- 5) M.H. Cohen and J. Lekner, Phys.Rev. 158 (1967) 305
- 6) W.F. Schmidt in "Charge Storage, Charge Transport and Electrostatics with their Applications", edited by Y. Wada, M.M. Perlman, H. Koado, Elsevier Scientific Publishing Company, Amsterdam-Oxford-New York (1979)
- 7) U. Sowada, Thesis at the Freie Universität Berlin, Fachbereich Physik (1976)
- 8) Kalrez, trade mark Du Pont de Nemours S.A., France
- 9) Oxygen monitor Teledyne 311, Bestobell Comp., USA
- 10) Moisture monitor Aquanal, Gerhard GmbH, Blankenbach, Germany
- 11) H. Jungblut, Diploma work at the Freie Universität Berlin, not published
- 12) L. Onsager, Phys.Rev. 54 (1938) 554