Electrical Breakdown in Helium Cells at Low Temperature

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Electrical breakdown of partially filled helium cells below 0.5 K is shown to be the result of Penning ionization of metastable triplet helium excimers bound to the surface of the liquid.

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The study of the electrical breakdown of dielectrics has a long and venerable history dating back 200 years. More recently [1], breakdown in superfluid helium has been investigated because of its importance in low-temperature, high-voltage applications where it can serve as both an electrical insulator and a thermal conductor. Breakdown in helium is also of fundamental interest because the liquid can be prepared with exceptional purity. While liquid helium under pressure is able to withstand fields in excess of 10^6 V cm^{-1} [2], it has been found [3] that cells with dimensions on the order of 1 cm, partly filled with liquid helium at temperatures below 0.5 K, may undergo electrical breakdown at very low voltages, i.e., at around 1000 volts. We have encountered this phenomenon in the development of a prototype detector of solar neutrinos using liquid helium as the target material [4]. Recoil electrons from neutrino scattering are to be detected by extracting them from the liquid and accelerating them in the vacuum by an electric field. In order to understand the possible constraints on such a particle detector using superfluid helium, we have studied electrical breakdown in partly filled cells, a process for which there appears to be no explanation in the literature.

We have performed a set of experiments to understand the nature of the breakdown. The results discussed here were carried out using a simple, cylindrically symmetric cell 0.4 cm high and 2.5 cm in diameter contained within a slightly larger vacuum-tight chamber, as shown in Fig. 1. The axis of the cylinder was vertical. The top and bottom surfaces of the cell were defined by electrodes, as was the side wall of the cylinder, called the ring electrode. Helium in measured amounts could be introduced into the cell. The current in the top and ring electrodes were measured as a function of the voltage applied to the bottom electrode. The current was found to be too small to measure ($<10^{-13}$ A) until a threshold voltage was reached. Above this voltage, a large current flowed and the temperature of the cell rapidly increased. The negative threshold voltage was approximately -1300 V, and the positive threshold voltage $+600$ V.

The observation and measurement of a threshold voltage does not provide information about the mechanism for the electrical breakdown, i.e., the way in which an initial small current grows and results in breakdown. To study this process, we have added to the cell a 0.5 mCi radioactive ^{63}Ni source ($\beta-$ emitter with an end point of 66 keV and average electron energy of 17 keV) was placed on the bottom in the center of the cell. The range of a 66 keV electron in liquid helium is 0.05 cm [5]. This source results in a measurable current before breakdown occurs, and we are able to study how this current varies with voltage to learn about the feedback mechanism that results in breakdown.

The results of three experiments, all performed at 100 mK, are shown in Fig. 2. The data in Fig. 2(a) were taken with the cell completely full of liquid. The potential is that of the bottom electrode with the ring and top electrodes grounded so that when the potential of the bottom electrode is positive (negative), positive ions (electrons) flow upwards through the liquid. The magnitude of the current is independent of the sign of the applied potential, and no measurable current flows to the ring. Within the field range of these measurements, the current to the top increases approximately linearly with increasing field. The primary electrons from the $\beta-$ source cause ionization in the liquid. Most of the secondary electrons recombine with positive ions and some are separated by the applied field. As the field increases, the number of secondary electrons

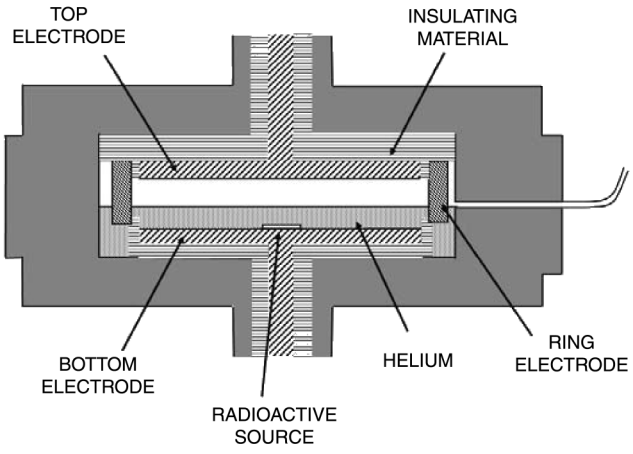

FIG. 1. Experimental cell. The inner diameter of the ring electrode was 2.5 cm, and height between top and bottom electrodes was 0.4 cm. The electrodes were made of copper and the insulator of Teflon.

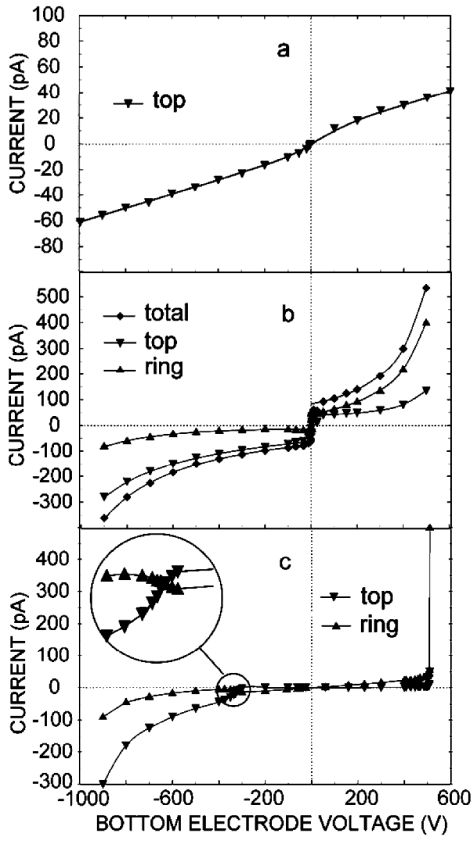

FIG. 2. Current to the top of the cell and to the ring as a function of the voltage on the bottom electrode at 100 mK. (a) Cell full of helium. (b) Cell half full of natural helium. (c) Cell half full of helium containing 6000 ppm ^{3}He.

that are separated rather than recombining increases. The saturation current, if no electron/ion pairs were to recombine, is estimated to be 600 pA. With the full cell there is no breakdown up to an applied voltage of 2000 V.

Figure 2(b) shows results with the cell half full of natural liquid helium (^{3}He content 0.1 to 0.3 ppm). At just a few volts in the half filled cell, the total current rises to a value of about 80 pA, levels off, and then rises further with increasing potential. Extrapolation of the current to higher voltage indicates that the current becomes infinite at voltages of $+670$ V and -1340 V. The current flow in the ring and top electrode is different for the two field polarities. Data have been taken up to voltages at which heat dissipation begins to raise the temperature of the cell.

We now consider possible explanations of these results. All walls above the free surface of bulk superfluid helium are covered by a He film, several hundred angstroms thick. Below 0.5 K the saturated vapor pressure of He is so low that the space above the liquid can be considered a vacuum. An electron in the vacuum and in the presence of an electric field is thus always destined to hit liquid helium, either the bulk liquid or the film. If an electron initially at the negative electrode acquires sufficient energy in traveling to the positive electrode, it can ionize helium atoms on striking the liquid (film or bulk), the ionization potential of He being 24.6 eV. The question is this: what is the mechanism that provides the positive feedback so that more electrons are created at a position in the cell such that they can be accelerated by the field and produce further ionizations? Two mechanisms can be dismissed: (1) Positive helium ions, accelerated by the field in the reverse direction, do not produce further ionizations at the point of origin of the original electron. A positive ion cannot escape the liquid at low temperatures [6,7] because of the polarization charge at the surface. Even if it were able to get to the vacuum, a He^+ ion accelerated to an energy of a few hundred eV has an extremely low probability of ionizing additional helium atoms on hitting the liquid. While there are ionization processes associated with atomic level crossings [8] when a helium atom and ion encounter each other at close range, the cross sections are very much smaller than the elastic scattering cross section [9]. (2) Photons, emitted in the radiative decay of excited-state helium atoms or excimers, do not produce electrons by photoemission that escape the negative electrode. The photon flux can be large in the presence of ionizing radiation [10]. However, the motion of a photoelectron of a few eV on entering the liquid is dominated by elastic scattering from He atoms, and its range is such that it cannot escape the attractive force of its image charge unless the electric field is large [6,11,12], the order of 10^5 V cm^{-1}.

The results for the half filled cell can be understood in terms of Penning ionization of excimers on the surface of the helium and the subsequent acceleration of electrons across the vacuum. An energetic electron in passing through helium produces ionization with a loss of energy, on average, of 43 eV per event [13,14]. Most of the electron/ion pairs, when the ionizing particle in helium is an electron, undergo geminate recombination [10], the ions first having formed He^+ excimers with a binding energy of roughly 2 eV. The number that does not recombine depends on the applied field. Very roughly, 50% of the excimers formed on recombination are in excited spin-singlet states and 50% are in spin-triplet states [10]. The ratio of the number of singlets to triplets is not $1/3$, presumably because in geminate recombination there is a correlation of the spin states of the recombining pair. Helium atoms in excited states also rapidly form excimers. Excited atoms in singlet states radiatively decay to the ground state, sometimes having formed an excimer, other times not, in less than 10^{-8} s [15]. The triplet atoms, however, all end up as excimers in the lowest $a^3\Sigma_u^+$ level, 18 eV above the dissociated ground state. The radiative lifetime for decay from this level has been measured in liquid helium to be 13 s [16]. Because of the large radius of the outer electron in the triplet excimer, it forms a bubble of radius 7 Å [17] in liquid helium. When an excimer encounters a wall, it is annihilated. When two $He_2(a^3\Sigma_u^+)$ excimers interact, they undergo the exothermic Penning reaction creating either

$3\mathrm{He}(1\,^1S) + \mathrm{He}^+ + e^-$ or $2\mathrm{He}(1\,^1S) + \mathrm{He}_2{}^+ + e^-$. Upon Penning ionization in the liquid, the separation of the charged products is small, and they recombine. The cross section for the Penning process in the liquid is estimated to be the order of $\sigma \sim 10^{-14}$ cm^2. Measured bimolecular rate constants, $\sigma\nu \sim 10^{-10}$ cm^3 s^{-1}, are consistent with this estimate [18]. Because the excimer is neutral and forms a bubble, it has a lower energy on the surface of the liquid than in the bulk [19,20]. If it reaches the surface at low temperatures, it is bound there but free to move in the plane. While Penning ionization does not create additional charge when it occurs in the bulk liquid, it certainly can do so when two excimers interact on the surface. An electron emitted into the vacuum escapes the Coulomb field of the ion. A positive ion can also escape the liquid provided its energy is sufficient to overcome the attraction of the electron remaining in the liquid. Two measurements report the ratio of the number of electrons to positive ions emitted from the surface to be 3 [6] and 1.6 [20].

Metastable triplet excimers are created in the liquid by the 0.5 mCi radioactive source at an estimated rate of $1.9 \times 10^7 \times (17\,000/43) \times (1/4) \approx 2 \times 10^9$ s^{-1}, where we have used 43 eV as the average energy to ionize a helium atom and produce an excimer. The factor of $1/4$ arises from the fact that only half of the source electrons enter the liquid and half of the excimers formed in geminate recombination in helium are in the triplet state. At 100 mK these excimers propagate with a mean free path that is determined by scattering from ^{3}He atoms in the superfluid. From a measurement of the low field current as a function of helium filling, we find that $1/4$ of the triplet excimers produced by the source reach the surface when the cell is half full ($3/4$ are annihilated on solid surfaces). The 5×10^8 excimers reaching the surface per second move about with a thermal velocity of 2×10^3 cm s^{-1} and annihilate in pairs to produce charges in the vacuum. The total surface area A_T of the He in the half filled cell is about 8 cm^2, so that the annihilation rate per unit area is $dn/dt = \alpha n^2 \sim 6 \times 10^7$ cm^{-2} s^{-1}, where n is the surface density of excimers and the rate constant is $\alpha = \lambda\nu$. The cross length λ for excimers to interact on the surface should be of the order of 10^{-7} cm, so that $\alpha \sim 10^{-4}$ cm^2 s^{-1} and $n \sim 5 \times 10^5$ cm^{-2}. The average distance an excimer travels before annihilation is $\ell = 1/(n\lambda) \sim 20$ cm, justifying the assumption that the excimers are spread uniformly over all the surfaces. The lifetime of an excimer due to two-body annihilation on the surface is $\tau = \ell/\nu \sim 10^{-2}$ s.

Current gain arises from electrons being accelerated across the vacuum from the more negative surface to the more positive surface. If the energy of the electron is sufficient to ionize He atoms, more excimers are created, some of which travel to the surface. Once bound to the surface, these mobile excimers become distributed uniformly over all surfaces. In so doing, they provide a feedback mechanism for generating additional electrons at the more negative electrode. Positive ions introduced to the vacuum and accelerated from the more positive to the more negative surface contribute to the current but produce no such feedback.

It is possible to make an estimate of the magnitude of the potential at which the current should increase without limit. The number of electrons generated in the vacuum at the more negative surface is $(1/2)A_n(dn/dt)$, where A_n is the area of the surface and the $1/2$ arises from the assumption that half the electrons from Penning ionization escape the liquid. When a potential V is applied to the bottom electrode, an electron acquires an energy of $V/2$ eV in accelerating across the vacuum of the half filled cell. If we assume that it takes 43 eV to create an ionization, that $1/2$ the ionizations create triplet excimers, and that $1/2$ the excimers reach the surface, then the total number of excimers returned to the surface where the potential is more negative (i.e., film or surface of the bulk helium) is $[(A_n/A_T)A_n(dn/dt)(V/2)(1/43)1/8]$. When this quantity is equal to the number destroyed $(A_n dn/dt)$, i.e., when the potential across the cell is $V = 690(A_T/A_n)$ V, the current should increase indefinitely. The ratio A_T/A_n depends on polarity, being 1.7 when V is positive and 2.4 when negative for the half filled cell. Hence this estimate yields "breakdown" voltages of $V_{+b} = 1200$ V and $V_{-b} = 1600$ V.

We have developed this model in more detail to calculate the current as a function of applied voltage [21]. The model includes (1) the current from positive ions produced by Penning ionization on the surface, (2) the transmission of electrons [6] but not positive ions through the surface, and (3) geometric factors related to the field profile in the cell. The model predicts the currents flowing in the ring and top electrodes to be

$$I_\pm = \pm \frac{a_\pm + b_\pm |V| + d_\pm V^2}{1 - |V|/V_{\pm b}}, \tag{1}$$

where the coefficients $a_\pm$, $b_\pm$, and $d_\pm$ have different values for positive and negative voltages ($\pm$) and for top and ring electrodes, whereas $V_{\pm b}$ depends only on the sign of the voltage. The terms have different origins. $a_\pm$ is associated with the flux of excimers that arrive at the surface produced by the radioactive source, $b_\pm|V|$ depends on the current generated by the source in the bulk liquid, and $d_\pm V^2$ is small, arising from ion production from the source-generated current if it traverses the vacuum. The curves in Fig. 2(b) are fits to the data points using Eq. (1) with $a_\pm$, $b_\pm$, $d_\pm$, and $V_{\pm b}$ treated as adjustable parameters. The best fit values of these parameters agree within a factor of 2 with what is expected from the model. The values obtained for the breakdown potentials for the half filled cell are $V_{+b} = 668$ V and $V_{-b} = 1340$ V. These values are somewhat smaller than the rough estimates made above, while their relative magnitudes are approximately in the ratio expected. The model does not capture particularly well the

division of current between the ring and the top electrodes in the vicinity of the step at low voltage. This discrepancy cannot be seen from the plots in Figs. 2(b) and 2(c). We believe this discrepancy may be due to the presence of space charge, in particular, electrons trapped above the liquid surface when the field is pressing them against the liquid.

The result of adding 6000 ppm of ^{3}He to the liquid is illustrated in Fig. 2(c). At low positive voltages no measurable current is observed in the top electrode. Excimers created by the radioactive source are scattered in the liquid to such an extent that in undergoing random walk they are more likely to encounter the bottom electrode, near their point of origin, than reach the free surface. Also at positive voltages, the current in the ring electrode is the same as in the full cell since the positive ions cannot penetrate the surface. At low negative potentials the same arguments hold. An electron bubble, because of scattering by ^{3}He, cannot obtain sufficient velocity to create a vortex ring to carry it through the surface. At a potential of -300 V, however, the current switches from the ring to the top electrode [see inset of Fig. 2(c)], as the electrons can gain sufficient momentum to create vortices. Once the crossover occurs, the current is amplified through the creation of excimers by the electrons accelerated across the vacuum.

When the potential is positive, the onset of a runaway current is abrupt. The voltage dependence of the current is that given by Eq. (1) with an extremely small numerator. The current only becomes large when the potential is very close to V_{+b}. This configuration most nearly approximates breakdown in the absence of a radioactive source. No electrons are introduced into the vacuum by the source, and few excimers reach the surface in the presence of the ^{3}He. For negative potential the current rise is much more gradual, resulting from the source current traversing the vacuum. The breakdown voltages obtained for the half filled cell with 6000 ppm are $V_{+b} = 510$ V and $V_{-b} = 1135$ V, which are lower than those found with natural helium. The most likely reason for this difference is a change in the number of excimers, created by the energetic electrons, that reach the surface. These excimers are created very near the surface. Because of their shortened mean free path, they are more likely to encounter the surface rather than wander off into the bulk liquid. If we assume that all the excimers reach the surface (rather than $1/2$ as assumed above), then the estimate of the positive breakdown voltage would be 600 V, accidentally close to the measured value. The results of other measurements, such as the dependence of the I-V curves on helium level in the cell, on ^{3}He concentration and on temperature, support the model developed here. These experiments will be reported in a lengthier publication.

In summary, the electrical breakdown in partially filled helium cells at low temperatures depends upon several properties of helium: (1) a film covers electrodes above the free surface, (2) triplet helium excimers form mobile bound states on the helium surface, and (3) Penning ionization of excimers on the surface inject electrons into the vacuum. Breakdown also depends on geometry. With a modest potential across the cell, electrons produced by Penning ionization are accelerated and, upon hitting the liquid, produce additional excimers in sufficient quantity to replace those lost in the annihilation process. With this understanding of breakdown it is possible to design a helium-based neutrino detector in which events are marked by detecting electrons calorimetrically above the liquid.

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