

## Ion Emission from Liquid Lithium for Inertial-Confinement-Fusion Applications

P. R. Schwoebel

*Division 1275, Sandia National Laboratories, Albuquerque, New Mexico 87185*

J. A. Panitz

*Department of Physics, University of New Mexico, Albuquerque, New Mexico 87131*

(Received 8 April 1991)

The first experimental studies of pulsed electrohydrodynamic ion emission from molten lithium are reported. Ions are produced at the apices of cusps formed on the surface of liquid lithium by applying a pulsed electric field of nanosecond duration. The ion species are dominated by  ${}^7\text{Li}^+$  and have a turn-on time of less than 10 ns as measured by time-of-flight mass spectroscopy. Current densities of  $\sim 300\text{ A/cm}^2$  have been observed. Implications for the use of this ion source in light-ion inertial-confinement-fusion reactors are discussed.

PACS numbers: 79.70.+q, 47.65.+a, 68.10.Jy

The study of electrohydrodynamic instabilities arising on the surface of liquids has been the subject of both experimental and theoretical investigations for a number of years [1–5]. Taylor's investigations [2] of a liquid cone and a jet extrusion under an applied electrostatic field stress led to the development of the liquid-metal ion source (LMIS) [6]. This source has had wide ranging applications in ion-beam technology, and motivated theoretical investigation of the physics involved in the formation of the "Taylor cone" and the ion emission processes at its apex [7]. Taylor's studies of single cone formation and jetting were followed by investigations of a related instability that manifests itself as a two-dimensional array of cusps on the surface of field-stressed liquids [3,4]. This two-dimensional instability arises due to the application of a dc electric field perpendicular to the surface of a liquid,  $E$ , which exceeds a critical electric field  $E_c$ . For  $E > E_c$ , a two-dimensional array of cusps is created on the surface of the fluid with a characteristic wavelength [3] (i.e., a distance between cusps),  $\lambda$ , that varies as  $E^{-2}$ . One would expect that, for conducting fluids undergoing such an instability, field enhancement at the apex of the cusps would lead to the field evaporation of ions similar to the case of single-Taylor-cone liquid-metal ion-source operation. Under pulsed-electric-field conditions theoretical considerations [8] have shown that the spatial characteristics of the fluid instability depend on the wavelength and growth time of the most rapidly growing instability mode, in addition to the magnitude of the applied electric field.

The possibility of employing a two-dimensional electrohydrodynamic (EHD) instability to drive a large-area lithium-ion source for light-ion inertial-confinement fusion (ICF) under pulsed-electric-field conditions is being explored [9]. Singly charged lithium is the ion of choice for light-ion ICF [9(b)]. Ion-beam-target coupling characteristics require a source that produces a relatively pure lithium beam ( $> 90\% \text{ Li}^+$ ), with a nanosecond-scale turn-on time. Since focused ion-beam inten-

sities of  $100\text{ TW/cm}^2$  are required to initiate a significant thermonuclear burn, the ion source is required to yield  $\text{Li}^+$  current densities on the order of  $5\text{ kA/cm}^2$  for 10–20 ns [9(b)].

In principle, an EHD ion source operating in the pulsed emission mode would satisfy these criteria by creating a two-dimensional array of Taylor cones on the anode surface in response to an applied voltage pulse [9(b)]. If each cusp emits  $\sim 10\text{ }\mu\text{A}$  of  $\text{Li}^+$ , and the cusps are spaced  $\sim 2000\text{ }\text{\AA}$  apart, then the required current densities can be reached.

In this paper, the first experimental studies of ion emission from a two-dimensional array of cusps formed on the surface of liquid lithium metal under *pulsed-field* conditions are reported. The purpose of this research is to (1) demonstrate the existence of pulsed, electrohydrodynamically driven two-dimensional lithium-ion emission, (2) characterize several physical parameters of the ion emission process, and (3) assess the applicability of this source for light-ion ICF applications.

The experimental arrangement is an all-metal ultra-high-vacuum imaging atom-probe [10] field-ion microscope operating in the low  $10^{-10}$ -torr range. Lithium was vapor deposited *in situ* from SAES [11] getters (containing  $\text{Li}_2\text{CrO}_4$ ), onto type 304 stainless-steel anode emitter tips with radii of  $\sim 1\text{--}2\text{ }\mu\text{m}$ , heated to  $\sim 500\text{ K}$ . The emitter tip anode allows one to achieve the anode surface fields expected in PBFA II ( $\sim 0.2\text{ V/\AA}$ ) with only modest applied voltages (tens of kV). Stainless steel was chosen as the anode emitter tip material because of its wettability by, and corrosion resistance to, liquid lithium. Prior to lithium deposition, the emitter tips were cleaned and smoothed by annealing in vacuums of  $10^{-10}$  torr at  $\sim 1200\text{ K}$  for 20 min. In addition to cleaning and smoothing of the emitter tips the annealing treatments are believed to improve the wettability of the stainless steel by providing a chromium-depleted surface [12]. Ion emission studies were conducted by pulsing a single lithium-coated emitter tip positive (from ground poten-

tial) using a spark-gas-switched cable discharge pulse generator. The pulse generator provides up to 25-kV pulses with a 3-ns rise time and a 20-ns duration. The voltage pulse rise time and duration were chosen to mimic those achieved by PBFA II, the existing light-ion particle beam fusion accelerator under development at Sandia National Laboratories [9(a)]. Further details of the experimental arrangement will be published elsewhere [13].

The ions produced at the anode tip are accelerated radially to a single-channel plate detector backed by a phosphor screen which allows the spatial distribution of the ions to be observed. The current pulse arriving at the screen was digitized, thereby recording the travel time of the ions and providing for time-of-flight mass analysis of the ion species.

The presence of a liquid lithium layer on the tip surface was verified prior to pulsed operation by observing the spatial distribution of the ion emission and the  $I$ - $V$  characteristic of the source indicative of Taylor-cone ion emission [14] under dc conditions. Typically, a discontinuous increase in current is observed with increasing voltage, accompanied by the appearance of a single ion emission site on the liquid surface. For the tip radii employed in these experiments ( $\sim 1$ – $2\ \mu\text{m}$ ) the voltage at which dc ion emission appeared,  $V_T$ , was  $\sim 4\ \text{kV}$ . A discontinuous drop in ion current to zero accompanied by the disappearance of the ion emission site is observed when the dc bias is reduced below  $V_T$ . Reversal of the tip polarity (emitter tip negative), and application of  $\sim 3$ – $5\ \text{kV}$ , results in a roughly periodic series of bright flashes on the phosphor screen. These bursts of field electron emission are also consistent with the presence of a liquid layer on the emitter tip surface [7(a)].

A typical Li-ion distribution under pulsed-field conditions is shown in Fig. 1. In this case the magnitude of the voltage pulse was 17 kV and the measured tip radius was  $\sim 2.2\ \mu\text{m}$ . Several emission sites are always observed, in agreement with theoretical considerations: Since the growth time of instability modes having a wavelength on the order of the emitter tip radius is longer than the applied voltage pulse duration of 20 ns, single-site emission will not be observed [8]. Using the field-voltage relation [15],  $E \sim V/5r$  (accurate to  $\sim 20\%$ ), yields  $E \sim 0.15\ \text{V}/\text{\AA}$ . The existence of relatively bright emission points interspersed with less intense emission sites on the liquid surface suggests the presence of cusps having slightly different aspect ratios (thereby field enhancement factors) on the liquid Li surface. However, such images consistently demonstrate that the ion emission per cusp is roughly uniform. The mean cusp spacing can be estimated by assuming an image magnification at the phosphor screen of  $D/\beta r$ , where  $D$  is the tip to screen distance (5.8 cm),  $r$  is the tip radius, and  $\beta$  is an image compression factor ( $\sim 1.5$ ) [15]. Applying this estimate to the emission sites shown in Fig. 1 results in a mean cusp spacing of  $\sim 4000\ \text{\AA}$ . A theoretical calculation [8] of the cusp spacing for liquid Li (in a parallel-plate geometry), in a



FIG. 1. A photograph of the pulsed ion emission distribution as it appears on the phosphor screen ( $V=17\ \text{kV}$ ,  $E \sim 0.15\ \text{V}/\text{\AA}$ ). The magnification indicated on the micrograph was calculated as discussed in the text. The intensity variation of the emission sites is consistent with ion emission from cusps having slightly nonuniform aspect ratios.

field of  $0.15\ \text{V}/\text{\AA}$ , yields  $\sim 2500\ \text{\AA}$ . The difference between the theoretical and experimental values is likely due to the boundary conditions imposed by the “hemispherical” anode used for the experiments as opposed to the parallel-plate geometry employed in the theoretical calculations. The ion-beam divergence from each cusp can also be estimated from Fig. 1. The average value of the half-angles we observe is  $\sim 6\ \text{mrad}$ , at accelerating voltages in the range of 15–25 kV.

The emission regions discussed above appear in different locations on the liquid surface from pulse to pulse, as would be expected for random EHD emission from a uniform liquid layer. It is interesting to note that other distinct modes of ion emission have also been observed in the course of these experiments and are apparently related to the lithium film thickness and its uniformity on the tip surface. For relatively thin films a two-dimensional array of ion emission sites is observed in pulsed fields of  $\sim 0.2\ \text{V}/\text{\AA}$ ; however, their location on the tip surface does not change from pulse to pulse. In this case the location of the emission sites is apparently controlled by the atomic-scale roughness of the emitter tip surface which remains even following high-temperature annealing. In the other distinct mode of ion emission, single, large “flashes” of ion emission are observed upon voltage pulsing. Since these intense bursts of ion emission usually saturate the channel-plate detector, ion species determination is difficult. The appearance of the emission pattern is similar to that which is seen when pulsed electron emission is drawn from a liquid metal. Occasionally, following repeated pulsing in this mode of

operation, one can observe a two-dimensional array of emission sites, fixed in location from pulse to pulse. It is not clear how this "burstlike" mode of ion emission arises, but we suspect that nonuniformities in the lithium film near the emitter tip apex (possibly due to incomplete wetting of the surface) may be responsible. Similar results for these two ion emission modes were obtained when voltage pulsing was done from a dc holding bias just below that required for single-Taylor-cone emission.

The ion species emitted during pulsed EHD operation were analyzed by time-of-flight (TOF) mass spectrometry. A representative mass spectrum is shown in Fig. 2. The dominant mass species is  $\text{Li}^+$  (the small peak corresponding to  $^6\text{Li}^+$  could not always be resolved from the  $^7\text{Li}^+$  peak). The presence of  $\text{Cr}^+$  in the mass spectrum suggests trace Cr contamination from the Li vapor deposition process which involves the thermal decomposition of a getter material containing  $\text{Li}_2\text{CrO}_4$ . Dissolution of chromium from the surface of the stainless-steel tip is another possible source of the  $\text{Cr}^+$  impurity. However, this is unlikely since the emitter tip annealing treatments employed for cleaning and smoothing result in a surface region depleted of Cr [12].

Time-of-flight spectra (in combination with simultaneous viewing of the ion spatial distribution) allow for an estimate to be made of the average absolute ion current produced per cusp. By taking the *average* current observed in the  $\text{Li}^+$  peak, and dividing by the total emitter tip area observed by the detector [15], the typical current density value is measured to be  $\sim 300 \text{ A/cm}^2$ , translating

into  $\sim 1 \mu\text{A}$  of ion current per cusp. Because of uncertainties in the rise time of the detector electronics and the exact voltage pulse shape at the emitter tip apex, we estimate that our current density measurement is accurate to within  $\sim \pm 10\%$ .

Finally, the TOF spectra allow us to estimate the turn-on time of the ion emission (i.e., the cusp development time). The appearance of a sharp, rising detector signal for the lithium ion species indicates that the turn-on time is less than 5 ns, the upper limit to the rise time of our detector electronics. This is consistent with theoretical calculations that yield a cusp formation time on the order of 0.5 ns [8].

These experimental investigations indicate that two-dimensional EHD ion emission sources could play an important role as an ion source in light-ion ICF reactors. Although the tip-to-plane geometry employed in the present experiments is different from the large ( $\sim 800 \text{ cm}^2$ ) anode geometry under investigation in Sandia's ICF reactor, there is no reason not to expect the underlying physical principles leading to cusp formation and ion emission to be the same in both cases. The spatial uniformity of the cusp formation over areas of  $\sim 800 \text{ cm}^2$ , however, has not been documented and must be addressed experimentally in the future. Our experiments have also shown that the turn-on time ( $< 10 \text{ ns}$ ) and beam divergence per cusp ( $\sim 6 \text{ mrad}$  at  $\sim 20 \text{ kV}$ ) are acceptable for a light-ion ICF source [9(b)]. In addition, the beam purity requirements for ICF ( $> 90\% \text{ Li}^+$ ) will easily be met by a lithium EHD ion source as demonstrated by the representative TOF spectra of Fig. 2.

One fundamental question remains to be addressed: the available current density of this source. The measured value of  $300 \text{ A/cm}^2$  in the present experiments is an order of magnitude lower than that required by PBFA II, but this is a lower bound on the current densities possible in the ICF accelerator's barrel diode. Our investigations have been conducted near the threshold field required for multiple-cusp formation and ion emission. Increasing the applied electric field above threshold values should lead to a greater surface density of cusps and an increase in the ion current emitted per cusp. In the present experiments the electric field is limited by our maximum pulse voltage of 25 kV and minimum tip radius of  $\sim 1\text{--}2 \mu\text{m}$ . For tip radii less than  $\sim 1 \mu\text{m}$  the lithium surface tension forces exceed the field stresses at  $\sim 0.2 \text{ V/\AA}$ , preventing cusp formation. Even at the cusp density observed in the present experiments, increasing the current per cusp to  $\sim 10 \mu\text{A}$  (a value easily reached in single-dc-Taylor-cone liquid-metal ion sources) yields current densities close to those required for a light-ion ICF ion source. Limitations on the source lifetime due to background vacuum contamination is an important practical consideration. Studies of the effects of  $\text{H}_2\text{O}$  contamination on pulsed ion emission from lithium have been completed, and will be published elsewhere [13].

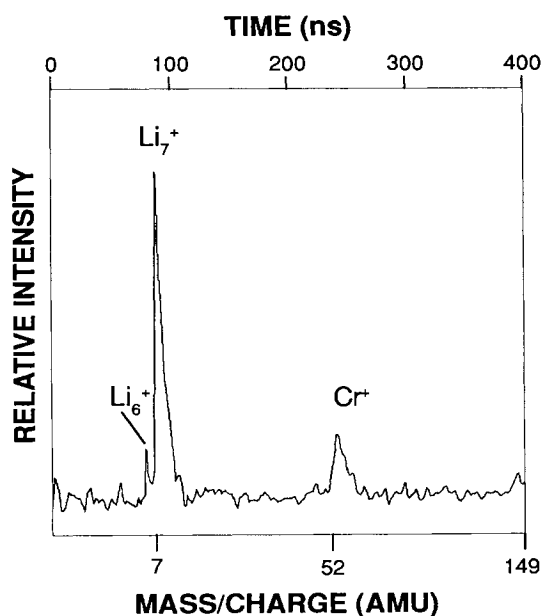


FIG. 2. A time-of-flight mass spectrum obtained from liquid lithium in the pulsed ion emission mode ( $V = 16.2 \text{ kV}$ ,  $E \sim 0.15 \text{ V/\AA}$ ). The dominant ion species produced is  $\text{Li}^+$ .

The authors would like to thank J. M. Sanchez and J. Hontas of the University of New Mexico (UNM) physics machine shop for their expertise in fabricating the ion source used for these investigations. In addition, we express our appreciation to R. A. Gerber, J. R. Woodworth, and A. L. Pregonzer for helpful discussions. This work was supported by the U.S. Department of Energy under Sandia Contract No. 75-3537 (to UNM).

- 
- [1] Lord Rayleigh, *Philos. Mag.* **14**, 184 (1882); J. Lamor, *Proc. Cambridge Philos. Soc.* **7**, 69 (1890).
  - [2] G. I. Taylor, *Proc. R. Soc. London A* **280**, 383 (1964).
  - [3] G. I. Taylor and A. D. McEwan, *J. Fluid Mech.* **22**, 1 (1965).
  - [4] G. I. Taylor, *Proc. R. Soc. London A* **313**, 453 (1969).
  - [5] J. R. Melcher, *Continuum Electromechanics* (MIT Press, Cambridge, MA, 1981).
  - [6] See, for example, R. Clappitt and D. K. Jefferies, in *Low Energy Ion Beams I*, Institute of Physics Conference Series Vol. 38 (Institute of Physics, Bristol, England, 1978), Chap. 1.
  - [7] (a) R. Gomer, *Appl. Phys.* **19**, 365 (1979); (b) L. W. Swanson and D. R. Kingham, *Appl. Phys. A* **41**, 223 (1986).
  - [8] A. L. Pregonzer, *J. Appl. Phys.* **58**, 4509 (1985); A. L. Pregonzer and B. M. Marder, *J. Appl. Phys.* **60**, 3821 (1986).
  - [9] (a) J. P. Vandevender and D. L. Cook, *Science* **232**, 801 (1986); (b) R. A. Gerber, in *The Physics and Technology of Ion Sources*, edited by I. Brown (Wiley, New York, 1989), Chap. 18.
  - [10] J. A. Panitz, *Prog. Surf. Sci.* **8**, 219 (1978).
  - [11] SAES Getters, Milano, Italy.
  - [12] R. L. Park *et al.*, *J. Vac. Sci. Technol.* **9**, 1023 (1971).
  - [13] J. A. Panitz and P. R. Schwoebel (to be published).
  - [14] See, for example, S. P. Thompson and P. D. Prewett, *J. Phys. D* **17**, 2305 (1984).
  - [15] R. Gomer, *Field Emission and Field Ionization* (Harvard Univ. Press, Cambridge, MA, 1961).



FIG. 1. A photograph of the pulsed ion emission distribution as it appears on the phosphor screen ( $V=17$  kV,  $E\sim 0.15$  V/Å). The magnification indicated on the micrograph was calculated as discussed in the text. The intensity variation of the emission sites is consistent with ion emission from cusps having slightly nonuniform aspect ratios.