ELECTROHYDRODYNAMIC ION EMITTERS

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The anomalies in the behavior of liquid films on electrodes in a strong electric field determine the main characteristics of many physical processes and have recently been the object of much study. Liquid films, which play an important role in electrohydrodynamic ion emitters, make it possible by very simple means to ensure generation of stationary ion beams with an ultra high current density. When the electric field strength at the positive electrode is raised above 10^8 V/cm intensive field evaporation of material in the form of ions begins from the condensed phase. Fields with such a high strength are obtained on needles with very small radii of curvature ($\sim 10^{-6}$ cm) [1]. When the electric field strength is raised above the evaporating level, however, the needle becomes blunt and the emission ceases. Steady-state emission can be achieved by depositing onto the point a film of melt which can flow to the emission zone during emission.

In recent years such electrohydrodynamic (EHD) emitters have found important application in the generation of ion beams with a high emission current density $j_0 \sim 10^8$ A/cm^2, which is 10^8-10^{11} times the typical value for ion sources of other types. The brightness of the generated beams increases to an even greater extent, viz., $B \sim j_0/T$, where T is the ion temperature at the emission surface. The emitter brightness determines the possibilities of forming submicron ion beams with a high current density, which are used for precision micromachining in microelectronics and for fabricating microproducts with a high level of complexity. Leading scientific centers and industrial firms of many countries have been working vigorously to develop and study EHD emitters.

The main features of the processes that determine the generation of ion beams in EHD emitters have been explained. A number of problems, however, still await resolution. The surveys in [2-4] give an idea of the principles underlying the operation of EHD emitters and their technological applications.

One of the objectives here is to draw the attention of readers of this journal to the physics of EHD emitters. The focus here is mainly on experimental studies of electrohydrodynamic effects that manifest themselves in the ion emission.

The processes studied occur in regions with dimensions in angstroms in ultrastrong fields, which virtually precludes their direct observation. It is thus necessary to use indirect methods for recording an long-range extrapolation, which enhances the role of physical models and imposes special requirements for their substantiation.

Figure 1 shows a scheme illustrating the operating principle of EHD ion emitters. When a voltage U relative to the extractor 5 is applied to the needle 1 wetted with a film of liquid metal 2, the electric field tends to separate the liquid which is held on the needle tip by capillary forces. As the voltage increases a time is reached when the pressure of the electric field becomes equal to, and then begins to exceed, the pressure of the capillary forces, i.e.,

$$E^2/8\pi \geqslant 2\gamma/R \tag{1}$$

where γ is the surface tension and R is the radius of curvature of the needle tip. At that time the liquid film on the tip is deformed, the liquid takes on a near-conical shape with a very sharp apex (Taylor cone), from which ions begin to emit. In this case a characteristic cross section 4 in the form of a point under 1 μm in size appears at the cone apex 3. The loss of material in the form of ions is balanced by an influx from the lateral surfaces of the needle. Another variant of EHD emitter is in the form of a liquid-filled capillary; a Taylor cone is also formed at its open end.

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The emitter operating voltage usually ranges from 5 to 10 kV at a needle tip radius of 2-20 μm and emission current I ~ 1-100 μA. Emission proceeds into a cone with a half-angle of ~ 20-30° at the apex. A typical current-voltage characteristic of an EHD emitter is shown in Fig. 2. Emission arises at the voltage U_2 and disappears at U_1, i.e., there is a segment with hysteresis. Intensive low-frequency oscillations of the emission current are excited at the voltage U_3.

Gallium, as well as other metals or their alloys, are most often used as the working substance in EHD emitters. In recent years methods have been mastered for obtaining ion beams from melts of nonmetallic materials, which in the molten state exhibit conductivity and a rather low vapor pressure, e.g., melts of $NaBO_2$, $LiBO_2$, and NaOH [5], $LiNO_3$ [6, 7].

Let us consider the operation of EHD emitters in greater detail.

Flow of Liquid to the Point. The needles of EHD emitters are usually made of wires sharpened by electrochemical etching. The needle is mirror-smooth after dc etching while after ac etching it is rough, with grooves about 1 μm deep in the direction in which the wire had been drawn [8]. In these two cases liquid flows to the point in different ways.

If the needle is rough, the liquid flows to the point along the grooves. We estimate the maximum "carrying capacity" of the grooves, assuming them to be semicylindrical grooves of radius r ~ 1 μm and length ℓ ~ 3 mm, the number of grooves being N ~ R/r, where R ~ 0.2 mm is the radius of the wire from which the needle was made. The grooves near the reservoir are filled completely and the liquid pressure in them is $p_1 = 0$. The grooves near the point may be almost empty and the pressure in them is $p_2 = -\gamma/r$ because of the capillary forces. The flow q of the liquid through half of a capillary with radius r is

$$q = \frac{1}{2}\frac{\pi r^4}{8l\eta}(p_1 - p_2) = \frac{\pi\gamma r^3}{16l\eta}$$

The total flow of the liquid is

$$Q = Nq = \frac{\pi\gamma r^2 R}{16l\eta} \qquad (2)$$

On substituting the typical values η = 1 cP and γ = 700 erg/cm^2 into (2), we have Q ~ 10^{-5} cm^3/sec, which corresponds to a current I = 10 mA, i.e., the grooves on the needle deliver (with a large surplus) material from the reservoir to the Taylor cone.

If the needle is smooth, the liquid will flow to the point in the form of a thin film, which evidently flows because of the "disjoining pressure" [9]. Disjoining pressure arises because, as a rule, it is energetically disadvantageous to form films thinner than several tens of angstroms. Accordingly, if the thickness of the film in an EHD emitter decreases because of the flow of liquid near the point, liquid begins to flow to that spot. The flow is difficult to calculate exactly in as much as no data are available on the disjoining pressure for liquid metals on metal substrates, but since we are dealing with the flow of thin (~100 Å) films, the hydrodynamic impedance of smooth needles should be much larger than that of needles with grooves.

Many authors [8, 10] associate the slope of the current-voltage characteristic of an EHD emitter with the hydrodynamic impedance of the needle. Indeed, the I—V characteristic of an EHD emitter with a smooth needle is much flatter than that of an EHD emitter with a rough needle [8]. Attempts to explain the flatter I—V characteristics of pointed needles by their higher impedance are inadmissible, it seems to us, since the contribution of impedance for rough needles is apparently small in all cases.

Liquid in an Electric Field. The shape assumed by a drop of a conducting liquid in an electric field was studied by Taylor [11]. He found an equilibrium configuration in the form of a cone with apex half-angle α = 49.3°. The electric field strength on the surface of a cone with this angle depends on the distance to the apex as $r^{-1/2}$ and a balance of the electric and capillary pressures persists along the entire surface at a particular voltage U_*. It is important to note that Taylor did not investigate the stability of this equilibrium configuration. Nor is it known whether other equilibrium configurations different from the Taylor cone exist. And although a number of studies [12, 13] question the existence of the Taylor cone, this cone is clearly visible in photographs of an operating EHD emitter [4, 14], even though the emitter voltage $U \neq U^*$ and the shape of the electrodes differ from the ideal.

Ion Emission and Size of Emission Zone. The Taylor solution was obtained for the static case while in an EHD emitter the liquid flows to the cone apex, from which it leaves in the form of ions. It is easily shown, however, that everywhere apart from the emission zone the velocity term $\rho v^2/2$ of the Bernouilli equation is substantially smaller than the pressure of the capillary forces and taking the flow into account virtually does not change the shape of the cone.

The Taylor model is limited in that it presumes the existence of an infinitely sharp cone with an infinitely strong field at the apex. In EHD emitters ions are emitted from the cone apex and the Taylor solution obviously becomes invalid in the region of the emission zone. It has now become generally accepted that the main emission mechanism is that of field evaporation of ions from the surface of the liquid in the strong electric field.

Field evaporation is a thermally activated process, whose current density is described by the equation [15]

$$j = e\sigma\nu \exp(-Q/T), \tag{3}$$

where $\sigma \sim 2 \cdot 10^{15}$ cm^{-2} is the surface atomic density, $\nu \sim 5 \cdot 10^{12}$ sec^{-1} is the characteristic frequency of the oscillations, Q is the activation energy, and T is the temperature in energy units. The activation energy is calculated from the formula

$$Q = H_0 + I - e\varphi - (e^3F)^{1/2} \tag{4}$$

where H_0 is the atomic sublimation energy, I is the ionization energy, and φ is the work function.

From (3) and (4) we see that the ionic current density increases exponentially with the electric field strength E on the surface of the liquid. It is often convenient to assume in calculations, therefore, that as the current density varies over wide limits the evaporating field E_0 remains constant and in the case of gallium emitter lies in the range E_0 = 1.5-1.6 V/Å [15] and no evaporation occurs at lower fields.

The characteristic flow that appears at the apex of the Taylor cone is a cloud of neutral atoms of the working material which are excited by the ion beam. Evidently, this glow is a "side" effect and does not have any appreciable impact on the emission process.

The radius of curvature of the spherical emission zone r_0 for different materials was calculated in [16]. It was assumed that the electric field strength is constant at the surface of the emission zone and that at low current the pressure of the electric field is balanced by the capillary pressure. The results of the calculations for different elements are given in Table 1.

Instability of EHD Emission at Low Currents. Many studies have noted the existence of a minimum EHD emission current ($I_* \sim 0.1-1$ μA), below which the emitter switches off [8, 17]. An explanation for this effect was proposed in [18]. It turns out that the flow of liquid to the emission zone and its departure in the form of ions may be unstable. If this process is to be stable it is necessary that under a random increase in the electric field the escape of material in the form of ions grow more than its flow in the form of a liquid. Then a pit where the electric field decreases forms at this point. For stability it is necessary that the emission current density be above some threshold j_*. We can integrate j_* over the surface of the emission zone to calculate the minimum emission current. Since the exact shape of the emission zone is unknown, it is difficult to expect a high degree of accuracy but the predicted temperature dependence of the minimum current ($I_* \sim T^{1/2}$) can easily be checked. The results of the calculations are given in Table 1, where an asterisk denotes the results of our measurements.

As shown by experiments, the minimum emission current I_* does not depend on the rate of decrease of the voltage, the resistance in the circuit of the needle, and the radius of the needle point. Bell et al. [19] report, however, that they managed to obtain stable emission with a current of 3 nA, using sharp needles. We repeated the experiments with sharp needles, measuring the ionic current with a high time resolution of ~10 nsec. To obtain such a high resolution the ions knock secondary electrons out of a converter and these electrons impinge on a fast phosphor. The flow is detected with a photomultiplier. In these experiments we were unable to obtain low-current stable emission. At an average current below the threshold $I < I_*$ emission occurred in pulses with a length of ~100 nsec and the current in each pulse was

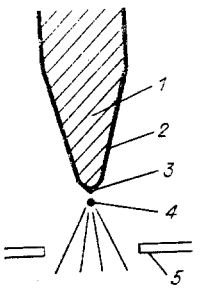

Fig. 1

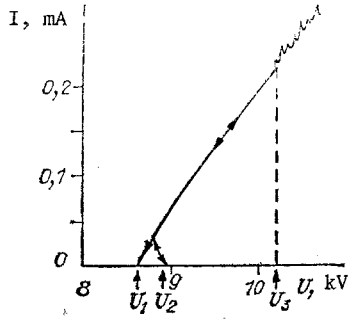

Fig. 2

I = I*, the intervals between pulses being of the order of 100 nsec as well. Pulses in the circuit of the needle were not resolved on the oscilloscope trace. It may be that in [19] the emission was transient but the emission pulses were averaged during the recording of the current. This assumption is supported by the results of measurements of the ion energy spread at low emission currents [19]. It cannot be ruled out, however, that we were unable to obtain a regime of a stationary low current while in [19] the flow of liquid to the emission zone was stabilized by the high impedance of a very sharp needle.

Emission Instability at High Currents. An investigation of the noise of the ion beam revealed spontaneous excitation of emission oscillations in the frequency range f = 0.1-100 MHz [20]. The spectrum of the current oscillations of a gallium EHD emitter is shown in Fig. 3 [1) 220 μA, 2) 80 μA, 3) 20 μA] and Fig. 4 shows the intensity of the oscillations with frequencies 7.8 and 55 MHz versus the emission current. The development of emission oscillations can be associated with the development of flow instability in the Taylor cone at a current above the threshold. The frequency f = 10-100 MHz corresponds to capillary waves on the surface of the liquid gallium with a scale of 1/k ~ 0.07-0.3 μm [20, 21].

Similar oscillations, but with a slightly different spectrum, are also observed with other working materials (e.g., the threshold of oscillation development is I = 44 μA with gold and about 80 μA with $LiBO_2$ melt). In work with a B—Ni—Si melt very weak oscillations were observed at an emission current I ~ 100 μA, which may be due to the high viscosity of this alloy. The current oscillations of a tin EHD ion emitter were studied in [22].

The oscillations of the current emission correspond to the development of asymmetrical modes at which the cone apex moves right-left. While not modulating the emission current, the asymmetrical modes nevertheless lead to an increase in the virtual size, which may prove to be undesirable when forming submicron ion probes.

The excitation of capillary waves on the surface of the Taylor cone in a natural way explains the generation of clusters in EHD ion emitters [20, 23]. Under intensive oscillation the apex of the Taylor cone can break away from time to time, forming a cluster (clusters) of radius $r \sim 1/k\left(k \sim \left(\frac{\rho}{\gamma}\omega^2\right)^{1/3}\right.$ and ρ is the liquid density). As the current increase more low-frequency modes develop (see Fig. 4), which results in the generation of large clusters. The frequency of the breakaway of clusters in this case may be much lower than the frequency of the oscillations, since the amplitude of oscillations in a high-Q oscillating system increases slowly until a cluster breaks away and as a result the amplitude decreases sharply and then again slowly increases. The frequency of cluster breakaway was not measured directly but if the magnitude of the mass transfer is known it can be easily estimated at several kilohertz. Since the current dependence of the intensity of the oscillations has a threshold, the graph of the mass transfer versus the emission current exhibits a kink [10].

The mechanism proposed above, of course, explains only the generation of large clusters (drops) with a size r > 10 Å. As for cluster ions of the form M_n^+ (n < 100), their mechanism of formation is still not clear. We should also note that complex compounds (of the H_2LiBO_3 type) emit large numbers of large molecules and molecular aggregates, which can then disintegrate into charged and neutral fragments. Such "soft" ionization can be useful in the mass spectroscopy of complex chemical compounds, including bioorganic compounds.

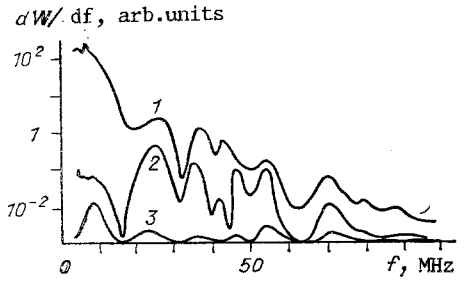

Fig. 3

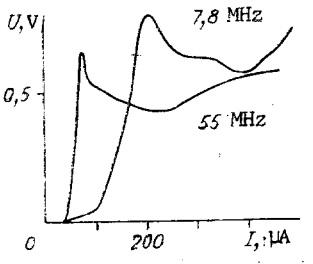

Fig. 4

Transient Processes. Transient processes in EHD emitters were studied in [20, 24, 25].

The liquid film on the point is deformed when the emitter voltage is raised above the critical value from condition (1). The Taylor cone does not deform immediately, however, since the volume of the liquid at the end of the needle is very small. The liquid begins to flow to the point, where it accumulates as a drop. As soon as the drop becomes large enough, conditions arise for the development of a smaller-scale mode and a "tongue" stretches and protrudes from the drop. A smaller "tongue" protrudes from the first "tongue", etc. Finally, the Taylor cone is formed. This mechanism, proposed by Grigor'ev [26], for the development of instability of a liquid drop in an electric field provides a correct qualitative description of this process, in our opinion.

Zheng and Linsu [27] suggested a different mechanism for switching on emission. They assumed that when the voltage is increased the liquid breaks away from the needle and rises higher and higher. At some voltage the field at the surface of the liquid increases to the value of the evaporating field and ion emission begins. We think, however, that stable configurations of the liquid, besides the liquid film and the Taylor cone, do not exist at the point. As soon as the film becomes unstable (at the voltage U_2; see Fig. 2), the Taylor cone is formed immediately. At a voltage below U_1 the cone is also unstable and a thin film forms on the needle point. Such "hard" formation and disappearance of the Taylor cone can be observed visually in a microscope in work with capillary EHD emitters and in emitters with blunt needles.

An oscilloscope trace of the current when the EHD emitter is switched on is shown in Fig. 5. The leading edge of the current is steep, even when the emitter voltage is raised slowly, and the duration of the current is about 100 nsec (gallium emitter). The burst of current is evidently due to the formation of a jet of liquid when the Taylor cone is completed. During the formation of the Taylor cone many modes of hydrodynamic oscillations are excited and the emission current oscillates a few times before reaching a steady-state value.

The delay with which the current is switched on was studied in [24, 25] as a function of the overvoltage. Two models were presented in [24] for explaining the delay: inflow of liquid with constant acceleration without allowance for viscosity (inertial model) and inflow of liquid with constant velocity (viscosity model). The first model gives the dependence of the delay τ on the applied voltage U in the form

$$\tau = \tau_0[(U/U_2)^2 - 1]^{-1/2}, \quad (5)$$

where U_2 is the threshold voltage (see Fig. 2) and τ_0 is a constant. The value of $[(U/U_2)^2 - 1]$ is in proportion to full intensity (electrical minus capillary), active at the liquid point. The second model gives the dependence in the form

$$\tau = \tau_0[(U/U_2)^2 - 1]^{-1}. \quad (6)$$

In [25], however, we showed that the experimental results, including those of Thompson and Prewett [24], are described much better by a dependence of the form

$$\tau = \tau_0[(U/U_0)^2 - 1]^{-2}, \quad (7)$$

where U_0 is slightly lower than U_2. The stronger dependence (7) of τ on U can be attributed to the fact that at a higher applied voltage the conditions for the development of a small-scale instability, which is responsible for the formation of the emitting point, arise with a smaller amount of liquid at the needle tip.

TABLE 1

Element	T, °C	E_0, V/Å	r_0, Å	j_*, $\frac{A}{cm^2}$	I_*, μA	
					calc.	exp.
Al	800	1,9	13	$2{,}0 \cdot 10^8$	1,5	1,4*
Ga	30	1,5	12,5	$6{,}6 \cdot 10^7$	0,47	0,26 [17] 0,28*
Ga	800	1,5	13,2	$1{,}3 \cdot 10^8$	1,0	0,7*
In	330	1,3	15	$4{,}3 \cdot 10^7$	0,44	0,5 [10]
Cs	30	0,5	17	$5{,}9 \cdot 10^6$	0,08	0,1 [28]

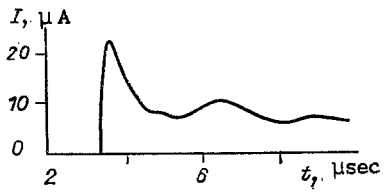

Fig. 5

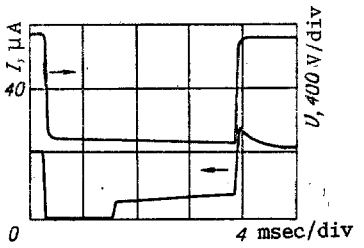

Fig. 6

In another series of experiments the voltage of an operating emitter was changed abruptly by a square-wave generator, but remained above U_2. The behavior of the emission current is illustrated in Fig. 6. In steady-state operation the liquid flows along the lateral surface of the needle in a viscous regime and the pressure in the Taylor cone should be negative in order to ensure this flow. Negative pressure is reached because the cone walls are "concave" and when the emission current is higher the negative pressure is larger (in absolute value), the walls of the Taylor cone are more concave, and its volume is smaller. The current, therefore, disappears as soon as the voltage decreases since there is insufficient liquid on the point for small-scale instability to develop at the lower voltage. When additional liquid flows to the point emission resumes, but with a lower current, naturally. When the voltage rises to its previous level the current corresponding to the static current-voltage characteristic is established in about 2 msec, after the surplus liquid at the point has been consumed.

In a capillary emitter the liquid flows in a comparatively thick layer inside a capillary and, hence, the viscosity has little influence and the effects due to the bending of the Taylor cone become negligible. Effects associated with the inertia of the column of liquid in the capillary, however, manifest themselves. If the voltage of the working capillary of the emitter rapidly decreases to a value slightly smaller than U_1, the emission current at first drops to ~ 0.5 μA and only after a comparatively long time τ ~ 0.2-0.8 sec does it fall off to zero. This effect is due to the fact that after the decrease in voltage the liquid in the capillary continued to flow by inertia and a drop forms at the end of the capillary and the development of small-scale instability, which initiates emission, is facilitated on this drop. When the velocity of the liquid in the capillary decreases, the electric field cannot elongate the drop and the emission disappears.

The experiments considered here revealed only some peculiarities of the functioning of EHD emitters, which manifest themselves in ion emission with a pronounced relief. A complete theory of melts in EHD emitters has not yet been developed, but the available experimental laws will serve as important reference points constructing it. The extremely high emission current density and the rapid cooling of ions in the expanding flow introduce specific anomalies into the evolution of the momentum distribution function of the ions.

Until recently, the development of practical applications of EHD emitters had been based mainly on empirical results and in many ways had been determined by the art of the experimenters. There is reason to hope that a more profound understanding of the processes in EHD emitters will help to determine their limiting characteristics more reliably and to realize these potentialities in practical applications.

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