Liquid-metal ion emitters

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This article describes and discusses the fundamental laws of ion emission from liquid-metal tips in a strong electric field. The widespread views of a liquid-metal emitter as being the smoothed tip of a Taylor cone are examined critically. The instability of a liquid metal in an electric field is discussed, and in line with this, an alternative concept is given of a sharp-tipped electrohydrodynamic emitter. The prospects for applying liquid-metal ion emitters are noted.

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1. INTRODUCTION

Noteworthy periods are known in the history of development of ion sources when fundamentally new ideas were advanced that stimulated a rapid improvement in the parameters of the ion beams being produced. One of these stages is the invention of ion emitters of a new type—liquid-metal ion emitters, whose application has facilitated the growth of important fields of contemporary technology. We should stress the difficulty of elucidating the nature of ion emission from a liquid metal, which is observed under extreme conditions characterized by the combination of high ion-current density, very small dimensions of the emission zone, the action of a strong electric field—as we know, analogous circumstances prolonged by many decades the studies of electron emission by the cathode spot of an arc.

Apart from technical details, the ion emitter being discussed is a small part of the surface of a liquid metal wetting a metal needle and covering it with a thin layer (Fig. 1). Facing the emitter is an electrode—the extractor, which gives rise near the tip to a strong electric field ~10^8 V/cm that accelerates the ions, and which has an aperture for extracting the ion beam that is formed. The intense emission of ions of the metal that is observed under these conditions cannot be explained in any way by a single known process, in particular, by the process of field evaporation.[1] In situations that reduce to this process, when, e.g., gallium exists on the surface of a tungsten tip in the solid phase, or arrives at the vertex of the tip from a reservoir containing liquid gallium but doesn't accumulate there, but gives rise to an autoionization image of the tungsten in leaving,[2,3] the emission current of Ga^+ does not exceed 10^{-9} A. Yet the specific regime of a liquid-metal ion emitter differs: in the large emission current 10^{-6}–10^{-3} A; in the existence on the surface of the needle of liquid metal that acquires a specific configuration in the strong electric field; in the action of a mechanism of self-heating of the emitting region—up to 500–1000 °C for gallium; in a characteristic glow near the tip; in an autoionization image in the form of a continuous spot, or sometimes in the form of bright concentric rings that appear and disappear.[4] One can arrive at this regime either by raising the temperature of the metal by external heating at a fixed, sufficiently high potential U (Fig. 2), or by increasing U to a cer-

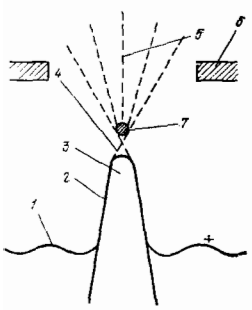

FIG. 1. Schematic diagram of a liquid-metal ion source. 1, 2—liquid metal; 3—metallic needle; 4—liquid-metal tip; 5—metal ions; 6—extractor; 7—glow region.

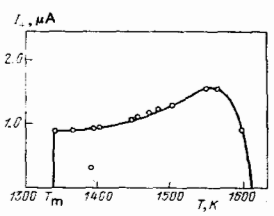

FIG. 2. Temperature-dependence of ion emission (gold emitter). T_m is the melting point of gold.

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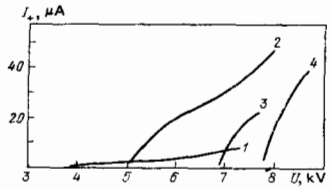

FIG. 3. Volt-ampere characteristics of gold ion sources for different needle radii (μm): 1 (1); 3 (2), 7 (3), and 9 (4).

tain threshold value U_{thr} (Fig. 3).[5,6] In both cases the establishment of the regime is accompanied by a jump in the ion current by several orders of magnitude.[5,7] After the threshold jump, the volt-ampere characteristic of the emitter has the average slope $dI/dU \approx 10-50$ μA/kV.

In order to draw a picture supported by facts of the complex phenomenon of ion emission from liquid metals in a strong electric field, after generally characterizing the phenomenon, we must proceed to analyzing it in detail.

2. COMPOSITION OF THE BEAM, ION-FORMATION ZONES

The composition of the beam drawn from a liquid-metal emitter is of undoubted interest. Mass-spectrometric analysis shows that the most widespread gallium emitter mainly emits[3] the atomic ions Ga^+ (99%) and a small amount of Ga^{++} ions, while a gold emitter differs in the high content of doubly-charged ions (65% Au^+ and 20% Au^{++}). As the total emission current I_+ increases upon varying the accelerating potential, in addition to increasing the number of atomic ions emitted, a component of molecular ions[2,8] such as Ga_n^+, where $n = 2-5$, appears and rapidly increases, or, e.g., Au_n^+ $(n = 2-7)$.

In addition to charged particles, the emitter also emits neutral particles. The existence in a flux of cesium ions of neutral atoms, with a ratio of these fluxes $\nu(Cs)/\nu(Cs^+) \leq 5\%$, could easily be established by using a detector whose action is based on surface ionization of atoms.[9] It proved possible to establish the existence of a neutral component near other liquid-metal emitters from the radiation detected here of neutral-atom radiation.[10,11] In the studied range of wavelengths 2000–8000 Å a set of lines was established, primarily the lines of excited atoms, in the case of gallium the lines are at 4172 and 4033 Å The lines of ionized gallium atoms are a hundredfold less intense, while the very weak molecular bands of Ga_2 could be detected only very recently. A quadratic dependence was established[10] of the radiation intensity of the 4172 Å line on the emission current. An number of conclusions was drawn from this: the free neutral atoms are formed by thermal desorption when accelerated electrons strike the emitter; these electrons arise in the beam because the predominant mechanism of ion formation is the field ionization of neutral particles; and the Ca I radiation is due to electron-atom collisions. However, further studies have shown that the radiation intensity increases with the current more rapidly than a quadratic law, and the authors of Ref. 11 arrived at the opposite conclusions: the source of neutral atoms, whose flux increases exponentially with the emission current, cannot be ordinary thermal desorption—there is not enough energy release for this[12]; at least up to a current of 30 μA the predominant mechanism of ion formation is not field ionization, but field evaporation; the Ga I radiation is due not to electron-atom, but to ion-atom $(Ga^+ - Ga)$ collisions.

Thus reliable data have been obtained by the mass-spectrometric method on the complex composition of the ion beams formed by different liquid-metal sources, while the specific radiation that has been found has made possible the quite evident detection of neutral atoms in the ion beam, although the mechanism of their intensive formation has not yet been elucidated.

Since liquid-metal emitters exist in a strong electric field, the study of the energy distribution of the ions is a rather sensitive method of determining their site of origin and serves as one of the fundamental sources of information to facilitate elucidating the mechanism of ion emission. The deficit of energy of an ion measured by retardation makes it possible to determine the difference in potentials between the formation zone and the liquid metal, and hence to determine also the location of this zone.

The energy deficit of an ion formed at the emitter by field evaporation is[13] $\Delta\mathscr{E} = I_n - n\phi + H_a - Q(E)$, where I_n is the ionization potential for an n-fold charged ion, ϕ is the work function of the retarding electrode-collector, and H_a is the evaporation energy. For Ga^+ with an activation energy $Q(E) \approx 0$, the calculated value is $\Delta\mathscr{E}$ = 4.9 eV. Figure 4 shows the energy distribution of gallium ions Ga^+ for different emitter temperatures.[13] At a low temperature at the maximum of the distribution the energy deficit amounts to $\Delta\mathscr{E} = 4 \pm 1$ eV. This is close to the value corresponding to the appearance of ions in the process of field evaporation. With increasing emitter temperature, the main peak shifts toward smaller $\Delta\mathscr{E}$. This situation is preferentially connected with the excitation of surface atoms of gallium by electrons that are produced in the process of field ionization and which approach the emitter. However, the extra broad peak with an energy deficit $\Delta\mathscr{E} \approx 10$ eV is explained by field ionization of the increasing flux of neutral atoms (the ions formed in free space do not pass through the entire accelerating potential difference).

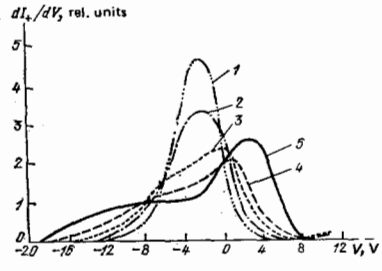

FIG. 4. Variation of the energy distribution of ions with temperature (gallium emitter). T(K) = 295 (1), 375 (2), 412 (3), 560 (4), and 900 (5). $I = 2$ μA.

An analogous shift of the maximum of the energy distribution of the ions toward a greater energy deficit of the gallium ions is observed with increasing emission current.[3,13] However, it is difficult to determine this shift exactly, in connection with the simultaneous increase in the half-width of the distribution. Figure 1 shows the relationship of the half-width of the energy distribution of the ions to the value of I_+. A similar relationship[14,15] is explained by the idea that a conversion of transverse momenta into longitudinal occurs in a dense, divergent ion beam owing to Coulomb collisions—the Boersch effect.[16] The solid angle Ω within which the emitter ions travel increases with I_+ from the same reason. In spite of this, the angular intensity I_+/Ω of the beam also increases[17] and amounts to ≈10–50 μA/steradian. The energy distribution of Au^+ ions is approximately Gaussian with a maximum at ≈100 eV, while the distribution of the Au_n^+ ions possesses a second peak corresponding to an energy up to 300 eV. This indicates that the site of formation of these ions is especially remote from the tip.[8]

Thus, the experimental data on the energy distribution of ions have proved to be a source of important information. There are three zones of formation of ions. The atomic ions at low currents are mainly formed directly at the emitter; with increasing current they are formed at the emitter and in part near it at distances ≲10–100 Å. The molecular ions are formed preferentially at distances ≈100–1000 Å from the emitter.

Finally, we should call attention to the actual lack of reliable data on the transverse dimension of the emission zone—another quantity that we must know for elucidating the physical nature of the phenomenon. Gomer,[18] in association with a number of authors, has estimated the radius of this zone as $\sim 10^{-7}$ cm. Yet it is difficult to agree with this estimate, which is not confirmed by any experiments, from the most general considerations. One cannot elucidate the true configuration of the emitter and the dimensions of the emission zone without studying the stability of the surface of a liquid metal in a strong electric field.

3. INSTABILITY OF THE SURFACE OF A LIQUID METAL IN AN ELECTRIC FIELD AND THE SHAPE OF THE EMITTER

Taylor's[19] study is usually taken as the basis for assuming that a liquid-metal emitter assumes the shape of a cone with a certain vertex angle $2\alpha_0$ (Fig. 6) at the critical potential difference U_{cr} between the extractor and the emitter. The condition of equilibrium of pressures at the surface of a conductive liquid with the surface tension γ and with the electric field E at this surface has the form

$$\gamma\left(\frac{1}{\rho_1}+\frac{1}{\rho_2}\right)=\frac{E^2}{8\pi}, \qquad (1)$$

Here ρ_1 and ρ_2 are the principal radii of curvature. Since the single radius of curvature of a conical surface is inversely proportional to the distance R from the vertex, Eq. (1) implies that the electric field at equilibrium declines proportionally to $R^{-1/2}$. Equilibrium is attained in a field having the potential

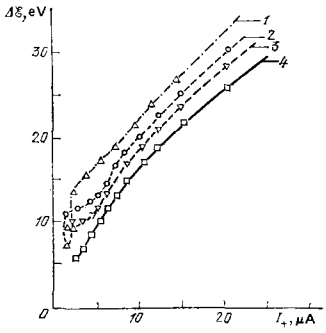

FIG. 5. Dependence of the half-width of the energy distribution of the ions on the total ion current (gallium emitter). T (K) = 552 (1), 455 (2), 390 (3), and 293 (4).

$$U = CR^{1/2}P_{1/2}(\cos\theta), \qquad (2)$$

Here C is a constant, and $P_{1/2}(\cos\theta)$ is the Legendre function. One can find from the condition for the cone to be an equipotential $P_{1/2}(\cos\theta) = 0$ that $\theta = \theta_0 = 130.7°$ and $\alpha = \alpha_0 = 49.3°$. An equation arising from (1),

$$\frac{\gamma\cot\alpha_0}{R}=\frac{1}{8\pi}\left(\frac{1}{R}\frac{dU}{d\theta}\right)^2$$

enables one to determine the field normal to the surface of the Taylor cone:

$$E=\frac{1}{R}\frac{dU}{d\theta}=1.4\cdot10^3\gamma^{1/2}R^{-1/2}\text{ V/cm}, \qquad (3)$$

and also to eliminate A and calculate the critical potential difference necessary for producing such a cone:

$$U_{cr}=1.4\cdot10^3\gamma^{1/2}R_0^{1/2}\text{ V}; \qquad (4)$$

Here R_0 is the value of R at $\theta = 0$—the distance from the vertex of the cone to the extractor, the shape of which is fixed by the relationship $R = R_0[P_{1/2}(\cos\theta)]^{-2}$.

A series of experiments confirms the transitions from a spheroidal to a conical surface at a certain critical field.[20] Liquid-metal emitters solidified in the form of Taylor cones have actually been observed,[8] although in a number of cases[6] the angle at the vertex substantially differs from $2\alpha_0$. Employment of an electron microscope with an electron energy of 200 keV has made it possible to study the shape of the point of the needle directly in the process of emission[21] and to establish a small conical projection at the tip with an angle near 90° (Fig. 7). At the same time, it proves difficult to fit the shape of the emitter in the form of an infinite Taylor cone with possible mechanisms of emission.

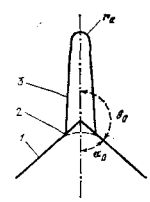

FIG. 6. Shapes of liquid-metal emitters. 1—Taylor cone; 2—Taylor cone with a smoothed vertex; 3—emitter with a hemispherical tip.

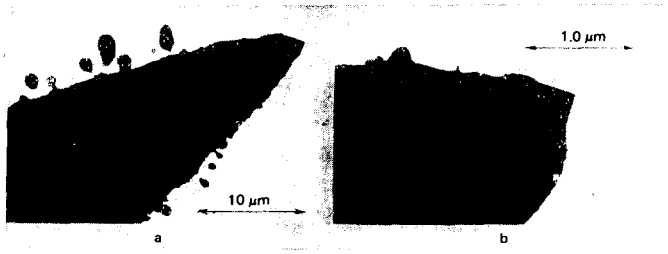

FIG. 7. Electron micrographs of operating indium emitters. a—drops being ejected by the field can be seen on the lateral surface; b—the tip can be approximated by a hemisphere of radius ~0.5 μm with a small conical projection.

As the calculations show,[1] a field $E \approx 10^8$ V/cm is required for obtaining ions by field evaporation or field ionization. According to (3), this is localized in a gallium (γ = 700 dyne-cm) Taylor cone in an emission zone of radius $\approx 10^{-7}$ cm. Such an improbably small radius of the emission zone is specific for an emitter of conical shape, for which the field declines sharply on moving away from the vertex. The need for a critical attitude toward the concept of this shape of the emitter primarily arises from Ref. 22. Attempts to determine numerically the self-consistent electric field with allowance for the space charge of the ions near the emitter suffered failure even at the minimal current $I_+ = 1$ μA. When the space charge was taken into account, the field changed sign in the region of the vertex. This ruled out the possibility of passage of current, and hence contradicted the original data. At the same time, the model of an emitter in the shape of the same Taylor cone, but with a projection at the vertex ending in a hemisphere (Fig. 6) enabled one to obtain the sought self-consistent field distribution with account taken of the space charge.[22] We should add that Taylor[19] did not consider the evolution of the surface of the liquid with increasing electric field. Hence the expressions (3) and (4) amount to a necessary, but insufficient, condition for formation of an equilibrium conical configuration (see also Ref. 23).

The need for eliminating the contradictions that arise impels one to turn to the problem of the stability of the surface of a liquid metal in an electric field. Tonks[24] and Frenkel'[25] have predicted an instability of gravitational-capillary waves at the surface of a liquid metal when a strong enough electric field exists there: $E > E_{cr}^{T-F}$. The instability arises because a small perturbation of the liquid $\xi = a\exp[i(kx - \omega t)]$ gives rise to the negative pressure $E_k^2\xi/4\pi$, in addition to the extra pressures $\rho g\xi$ and $\gamma\partial^2\xi/\partial x^2$. This negative pressure leads to an increase of the initial perturbation, to a new increase in the negative pressure, etc. One can obtain from the dispersion equation for gravitational-capillary waves in the presence of an electric field[26]

$$\omega^2 = \frac{k}{\rho}\left(\gamma k^2 + \rho g - \frac{E^2}{4\pi}k\right) \qquad (5)$$

the well-known equation for the critical field for aperiodic instability ($\omega^2 < 0$) of Tonks and Frenkel':

$$E_{cr}^{T\text{-}F} = \sqrt[4]{64\pi^2\gamma\rho g}. \qquad (6)$$

Under the conditions that we are treating, only the short-wavelength capillary waves ($\alpha k^2 \gg \rho g$) are important. These are unstable at the critical field

$$E_{cr}^{V} = \sqrt{4\pi\gamma k}. \qquad (7)$$

Interestingly, the Tonks-Frenkel' instability has attracted attention in recent years in connection with the instability that has been discovered in a charged surface of liquid helium in an electric field[27,28] and with the study of the behavior of a ferromagnetic liquid in a magnetic field.[29]

The expression (7) implies that liquid-metal emitters in a strong electric field that is characteristic of them are unstable, even with respect to very-short-wavelength perturbations. As we have pointed out above, Taylor[19] did not touch upon problems of stability in his theoretical analysis, but at the same time demonstrated experimentally the instability of the vertex of a cone that was manifested in the emission of a very thin flux of liquid. The hypothesis that the actual emitter is not the smoothed vertex of a cone, but the front of a flux flowing from the vertex had been advanced already in Ref. 30. This hypothesis agrees with the results of theoretical and experimental studies on the nonlinear stage of the Tonks-Frenkel' instability.

Upon summarizing the results of the theoretical studies[24,25,31] in this regard, one could predict two stages of instability. The first culminates in the formation of a spheroidal or conical projection on the surface of the liquid metal. In the second stage the projection keeps its volume and additional liquid does not enter it, but the concentration of electric charge leads to a rapid growth of a peak on the projection, with a decreasing radius of curvature. Near the leading front one can observe the appearance of a constriction—or precursor of detachment of a drop from the growing peak.[31] The picture that we have presented has been reproduced experimentally. The growth of microprojections on the surface of a liquid metal situated in a strong electric field has been established by shadow photography and with a fast-acting camera.[32] With all the attractiveness of this method, which allows one to trace directly the development of instability *in situ*, it has certain limitations, like any optical method, and cannot yield information on the very fine details of the developing peaks having a characteristic dimension of the order of a wavelength of light. In this sense, an advantageous method is one that is based on using the contact of a liquid metal with a plasma,[32] which enables one to record at a certain moment the developing nonlinear waves, and then to study all the details at high resolution with a scanning electron microscope. Figure 8 shows a panorama of these "frozen" waves, together with photographs of individual peaks. They confirm the two-stage character of the development of instability—formation of spheroidal or conical projections, with subsequent growth of peaks on them with a decreasing radius of curvature; on some of the peaks one can see drops "frozen" at the instant of detachment. The shape of the layer of metal covering the emitter needle (see Fig. 7) is directly associated with the instability that we have discussed.

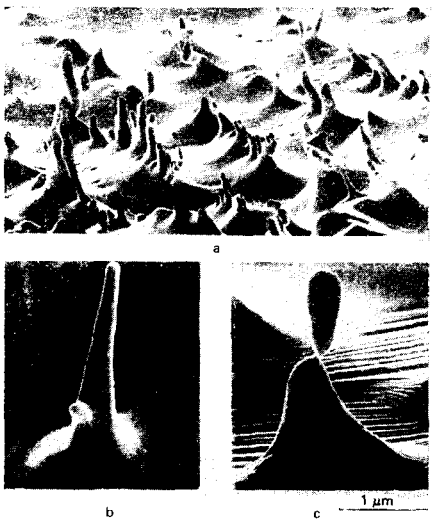

FIG. 8. Panorama of "frozen" nonlinear capillary waves produced by the Tonks-Frenkel' instability (a); magnified image of an individual peak (b); and a drop "frozen" at the instant of rupture (c).

Thus, it is highly probable that the ions are extracted not from the smoothed vertex of a Taylor cone, but rather from the front of a very thin flux of metal emitted by the vertex owing to instability. In this case, one can apply the formula well known in autoionization microscopy for calculating the field at the emitter: $E = U/\mu r_a$, where μ is a coefficient approximately equal to 5 over the broad range of values of R_0/r_a from 10^3 to 10^5, and r_a is the radius of curvature of the front of the flux.[35] Since under these conditions the field is relatively homogeneous, the value $r_a = 10^{-5}$ cm calculated for the typical values $U = 5$ kV, $E = 10^8$ V/cm must be close to the radius of the emission zone r_{zone}. It is comparable with the commonly adopted value of the maximum radius of this zone.[7,13,21,36] As is implied by the interesting electron micrographs of a working emitter[21] (see Fig. 7), in spite of the existence of a conical projection, the electric-field distribution cannot be described by Eq. (3), which pertains to a Taylor cone of infinite dimensions.

The current density $j_+ = 10^6$ A/cm^2 corresponding to the radius 10^{-5} cm (for $I_+ = 100$ μA) no longer arouses the objections presented above, which were based on taking account of the effect of the space charge. Of course, at such a relatively large value of r_a the equation $E^2/8\pi = 2\gamma/r_a$ may not be satisfied. That is, the pressure at the surface may prove unbalanced. We should conclude that the concept of a sharp-tipped emitter unavoidably leads to the idea that the configuration and arrangement of the emitting boundary of a liquid-metal ion source are not determined by the equation of balance (1) of the pressure, but merely by the equality of the number of atoms arriving at the emitter and the number of ions departing from it.[37]

If we assume that instability of the metal surface sets in at the threshold potential with a maximum wavelength equal to the radius of curvature of the needle, then, upon substituting $E = f(r)$, we obtain from Eq. (7) an expression for this potential

$$U_{thr} = \sqrt{8\pi^2\mu^2\gamma r_{needle}} \qquad (8)$$

This agrees qualitatively with the experimental data[5] for "sharp" needles (see Fig. 3). As we see by comparing (4) and (8), the threshold potential need not coincide with the critical potential at which a Taylor cone is formed.

4. ION EMISSION FROM A LIQUID-METAL TIP. LIMITATION OF THE ION-CURRENT DENSITY AND THE BEAM CURRENT

Figure 9 shows schematically the processes of formation of ions and of neutral particles accompanying them as discussed in connection with the problem of ion emission from liquid metals. Greatest attention is drawn to the process of field evaporation, as well as the process of field ionization of atoms in the region adjoining the emitter. Apparently the contribution from the latter process becomes appreciable with increasing total ion current. Molecular clusters are formed both at the surface of the liquid metal and in the flux of moving neutral particles.[8] The most probable cause of a cluster's acquiring a charge is the process of charge transfer: $Au^+ + Au_n \rightarrow Au_{n+1}^+$; $Au^+ + Au_n \rightarrow Au_n^+ + Au$. Especially at large currents, drops can be formed with diameters up to several micrometers. The metals are more prone to drop formation that have a high value of the "evaporating field",[1] i.e., usually the metals with high ionization potentials.[9] The fundamental cause of heating of the emitter is bombardment by electrons formed in the process of field ionization.

Above we have already called attention to two possible stationary states of an emitter. According to the widespread viewpoint, the shape and position of the emitter are determined by the condition (1). However, in the continuous-action sources that we have been discussing, the equality of the flux density $\rho v/M$ of atoms reaching the surface of the emitter to the flux density of the emitted ions j_+/e, with allowance for the neutral component, must be satisfied at the same time.

In addition, another state can also exist in which balance of pressures is not maintained, but namely we have $E^2/8\pi \gg 2\gamma/r_a$, and the location of the emitter is determined only by the stated dynamic equilibrium of

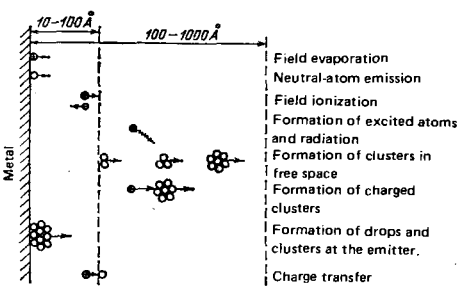

FIG. 9. Schematic diagram of some processes that occur near a liquid-metal emitter.

the flux of particles at its boundary with the vacuum.[37] The latter allows one to calculate the limiting current density of ions formed by field evaporation. When the metal atoms supplied to the surface are completely converted into free ions, the law of conservation of momentum implies that

$$\rho v v_1 \leqslant \frac{E^2}{8\pi}. \tag{9}$$

Here v_1 is the mean velocity of the ions being emitted and v is the velocity of movement of the metal. The inequality (9) imposes a limitation on the value of the ion current density:

$$j_+ = \frac{e\rho v}{M} \leqslant \frac{eE^2}{8\pi M v_1}. \tag{10}$$

The typical values $E = 10^8$ V/cm, $v_1 = 4\times10^5$ cm/s, and $M = 10^{-22}$ g correspond to the limiting ion current density $j_{+,\lim} = 10^7$ A/cm^2.

Thus, if we start with the picture of emission of ions from the equilibrium surface of a Taylor cone with action of the field described by Eq. (3), then the calculated emitted ion current proves smaller than that observed: $I_+ = j_{+,\lim}\pi r^2_{\text{zone}} < 10^{-7}$ A. However, if we start with the picture of an emitter with an unbalanced pressure with a radius of the emission zone of 10^{-5} cm, rather than 10^{-7} cm, then the ion current agrees with that observed experimentally: $I_+ = j_{+,\lim}\pi r^2_{\text{zone}} < 10^{-3}$ A. This is yet another argument favoring the concept of a sharp-tipped emitter with an unbalanced pressure at the surface.

Despite the fact that liquid-metal emitters are situated in a strong electric field, the space charge of the ions proves substantial, even at minimal current, owing to the large current density. By diminishing the electric field, the space charge creates a negative feedback, with which Gomer[18] explains the high stability of the ion current drawn from a liquid-metal emitter.[15] Actually the space charge should damp the current oscillations. However, evidently, one must agree with the opinion expressed in Ref. 38 that the noise in the ion current is limited by the fluctuations in the flux of metal to the emitter.

The space charge also determines the value of the maximum current of an ion beam whose source is a single tip with a fixed potential difference U_0 between it and the extractor. To calculate the field with allowance for the space charge, one resorts to the very simple model of two concentric spheres[18,23,37]—an inner emitter with the radius r_a and an outer extractor ($R_0/r_a = \xi$). Introduction of the dimensionless parameters of potential $y = U/U_0$ and of radius $x = r/r_a$ enables one to represent the Poisson equation in the form

$$\frac{d^2y}{dx^2} + \frac{2}{x}\frac{dy}{dx} = \frac{A}{x^2\sqrt{y+\alpha}} \tag{11}$$

with the boundary conditions $y(1) = 0$, $y(\xi) = 1$. Here we have $A = I_+(M/2e)^{1/2}U_0^{-3/2}$, and $\alpha = Mv_1^2/2eU_0$, where e and M are the charge and mass of the ions, and v_1 is their initial velocity. Figure 10 shows some relationships of the field at the emitter $dy/dx|_{x=1}$ to the parameter A. One of them has been obtained numerically with a computer, and the other is an approximate solution of

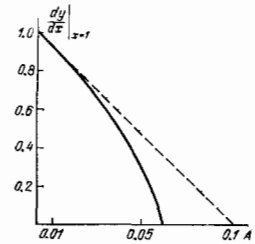

FIG. 10. Dependence of the field at the emitter on the parameter A. The approximate solution is shown by the dotted line.

(11) by replacing the function on the right-hand side with the corresponding solution of the Laplace equation. The approximate solution

$$\frac{dy}{dx}\Big|_{x=1} \approx [1 - A(\ln 4\xi - 1)] \tag{12}$$

implies, primarily, that the attenuation of the field owing to space charge in the studied system of electrodes depends weakly on the radius of curvature of the emitter (the quantity ξ). This situation justifies posing the problem with the fixed radius r_a. The evident limitation of the parameter A enables one to estimate the maximum ion current that one can extract from a single tip in an actual non-spherically-symmetric system. Thus, when $U_0 = 6$ kV and $M = 10^{-22}$ g, the maximum current does not exceed 1 mA, which agrees with the experimental data.[20]

The volt-ampere characteristic of a liquid-metal emitter is determined in a complicated way by the restriction of the field by the space charge (Fig. 10), by the dependence of the emission current on the field, by the interrelation of the shape of the tip and the electric field at its surface, and by the impedance of the flux of viscous liquid along the base of the emitter—the lateral surface of the needle.[38-40]

The solution of Eq. (11) points out another interesting feature of the manifestation of the space charge—at relatively large currents the field maximum does not lie at the surface of the emitter, but at some distance from it.[18,23]

5. APPLICATIONS OF LIQUID-METAL EMITTERS

The application of liquid-metal emitters[41-44] primarily involves the unique potentialities that are opened up by using dense, well-focused ion beams—ion probes, which enable:

1. Rapid analysis of a given microvolume of a solid object, including isotopic analysis and obtaining of an image of the surface "in rays of atoms of a given mass".

2. Ablation of the solid with the ion probe, and hence, layer-by-layer analysis, and various forms of dimensional processing of a solid object.

3. Control of the composition of a given microvolume by dosed implantation of the required atoms transported by the ion probe.

The transverse resolution of an ion probe is determined by the diameter of the region of development of the cascade of collisions caused by the primary ion in a plane at a depth ≈10 Å, from which the secondary particles can then emerge; under typical conditions this value amounts to ≈100 Å, and it determines the optimal dimensions of the ion probe. The potentiality that has been established of forming submicron ion probes by using liquid-metal ion emitters has attracted the attention of physicists, technologists, geologists, archeologists, etc., and has opened up broad prospects for applying these probes for scientific and applied purposes.

The most important element of a system for forming an ion probe is the ion source. The essential point is that the defects of the latter unavoidably affect such fundamental properties of the beam as its phase volume or stability. In turn the latter often fatally limit the potentialities of using this beam. The minimum diameter of the beam formed by aberration-free optics is

$$d_{\min} = \frac{d_0}{\sqrt{\Phi}} \sqrt{\frac{kT_1}{eU_0}} \frac{1}{\sin\theta}. \qquad (13)$$

Here d_0 is the initial diameter of the beam, eU_0 is the energy of the ions, T_1 is their effective temperature, θ is the half-angle of the cone in the region of the focus, and Φ is the ratio of the actual current density at the focus to the limiting current density according to Langmuir. This well-known expression implies that one primarily must take care that the quantities d_0 and T_1 are minimal (minimal emittance of the beam) in order to form a thin probe. We should add that in order to decrease the spherical aberration of an actual ion-optic system—this is quite important—the ion beam having the required current must be paraxial. Thus, a very important parameter of an ion source proves to be its brightness—the current density of the beam being formed per unit of the corresponding solid angle. In connection with the extremely high initial current density of liquid-metal ion emitters, their brightness is estimated to be ~10^6 A/cm^2 steradian.[17,45] This exceeds by four orders of magnitude the brightness of the best classical plasma sources of the type of the duoplasmatron. The relatively large scatter in the energy of the ions obtained by using the discussed emitters can also be attributed to a substantial chromatic aberration. In this regard it proves important to adopt the correct ion-beam current.[14]

As an example, Fig. 11 shows a diagram of the formation of a submicrometer ion probe using a gallium liquid-metal ion source.[46] This system enables one to form an ion probe of diameter 10^{-5} cm with a current density of 1.5 A/cm^2, energy 57 kV, and beam brightness at the target of 3.3×10^6 A/cm^2 steradian. In an apparatus for studying the condensation of gallium ions[47] transported by an ion beam, the liquid-metal emitter was employed only at small currents ~μA, at which one could reliably avoid the formation of clusters or drops, which was inadmissible in this case. It has recently been possible to achieve the formation of an ion probe of diameter 0.1–5 μm with an energy regulated over the broad range 20–2 kV by using an asymmetric three-electrode lens.[48]

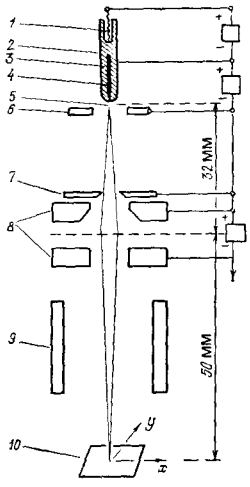

FIG. 11. Schematic diagram of a system for producing a submicrometer beam of gallium ions. 1—heater; 2—reservoir; 3—liquid gallium; 4—tungsten needle; 5—emitting tip; 6—extractor; 7—aperture; 8—electrodes of the accelerating lens; 9—deflection plates; 10—target.

We can arbitrarily classify liquid-metal ion sources into three groups in terms of features of construction. In the sources belonging to one of them, the liquid metal is supplied through a capillary,[20,30] at the exit of which the ion-emitting surface of this metal is formed by an external electric field. The sources of the second group are distinguished in having the liquid metal in a special reservoir from which a metallic needle emerges, usually made of tungsten; the surface of the latter is wetted by the metal to become convered with a thin layer of it, which is the ion emitter proper.[49] Finally, the sources of the third group amount to a metallic needle welded to a metal loop (Fig. 12); the temperature of the needle is regulated by passing a current through the loop. The needle and the loop simultaneously also play the role of reservoir of liquid metal, which covers their surfaces more or less uniformly, or which exists preferentially in one site in the form of a drop.[4,15] Just as in the sources of the second group, the emitter is the surface of the liquid metal wetting the end of the needle and covering it with a thin layer. In every apparatus an extractor lies in front of the emitter, and sometimes another control electrode in front of the former.[48]

FIG. 12. Photograph of a liquid-metal gallium-ion emitter.

The operation of the source is preceded by treatment of the needle aimed at cleaning the surface until it is well wetted by the liquid metal.[5,15] Naturally, the material of the needle must not dissolve in the liquid metal. Special difficulties, which have been encountered, e.g., in developing aluminum ion sources,[50] have consisted in the active interaction of molten aluminum with most metals, including tungsten. In this case a graphite needle treated with titanium has been used successfully (see also Ref. 34).

Above we have presented as typical data those on gallium and gold emitters. However, at present the properties have been studied of liquid-metal emitters of the ions of Ga, In, Au, Cs, Al, Bi, Li, Sn, Hg, Ge, U, Pt, Fe, and other metals. In cases in which the obstacle to designing an emitter is the high melting point of the metal or its high vapor pressure, one can employ alloys with a melting point below that of the initial components. For example, emitters have been described based on the alloys Bi-Sn-Cd, Au-Si, Au-Ge,[51] and Sb-Au-Pb.[52]

In closing, we must also touch on liquid-metal emitters of intense ion beams intended for, e.g., application in accelerator technology or in the field of controlled thermonuclear fusion.[36,53,54] One of the methods of obtaining intense ion beams consists of using a block of liquid-metal tips,[36,53,54] whose surfaces serve as the source of a dense plasma during a discharge pulse.

Fluxes of clusters and drops from the surface of a liquid metal situated in a strong electric field can be employed, in particular, for obtaining thin films and rapidly solidifying alloys and production of fine particles for powder metallurgy.[21]

6. CONCLUSION

Owing to the record-setting values of such parameters as the ion current density, brightness, stability, and other valuable properties, e.g., the self-renewal of the material, liquid-metal ion emitters possess broad prospects, not yet fully evaluated, of application in various fields of science and technology. Yet we must assume that the study of the mechanism of ion emission by liquid metals in a strong electric field will discover new interesting features of the complex phenomenon, which develops, as in the case of the cathode spot of an arc, in a region of very small volume $\approx 10^{-17}$ cm^3 with a very high energy density.

Serious arguments contradict the widespread view[18] that the liquid-metal ion emitter proper is a zone of radius $\sim 10^{-7}$ at the smoothed vertex of a Taylor cone—the surface of a liquid metal with the action of the electric field and the surface tension balanced at every point. In particular, such arguments include the above-discussed suppression of the field near the stated zone by the space charge of the ions being emitted, and also the restriction that we have pointed out on the flux of the latter that is imposed by the law of conservation of momentum.

The alternate concept of a sharp-tipped emitter formed by the instability of the surface of the liquid-metal tip, and characterized by a radius of the emission zone of $\sim 10^{-5}$ cm and an ion-current density of 10^4–10^6 A/cm^2, merits attention. The configuration and position of the front of such an electrohydrodynamic emitter having an unbalanced pressure at the surface is determined by the equality of the number of atoms arriving there and of emitted ions.

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