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Ultramicroscopy 57 (1995) 45–58

ultramicroscopy

HV TEM in situ investigations of the tip shape of a gallium liquid-metal ion/electron emitter

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Received 23 March 1994; in final form 28 August 1994

Abstract

The tip shape of a gallium liquid-metal ion/electron emitter has been observed in situ in the Halle 1 MeV electron microscope in both the ion and electron emission modes. In the ion emission mode deviations of the liquid-metal cone shape from the Taylor cone are found. Starting at low emission currents ($I_e \approx 2$ μA) a jet-like protrusion of the Taylor cone vertex was observed. There was a linear dependence of the decrease of the cone half-angle α and of the increase of the length l of the jet-like protrusion on the increased emission current. The experimental results are in good agreement with the calculations of the jet parameters. Also dynamical effects as e.g. droplet emission and spatial shifts of the Taylor cone are registrated during the experiments. Emission of "Faraday droplets" was observed in the Taylor cone surroundings. The radius of the droplets varied between 0.04 and 0.5 μm.

In the electron emission mode, an emitter of a tip radius of 20 μm shows behaviour different from that of a tip radius of 0.5 μm. In the first case pulse-like electron emission was observed, which starts at the onset voltage of the formation of the Taylor cone. Processes of the formation and disappearance of the Taylor cone had not been observed during these in situ experiments in the HV TEM. In the second case of a sufficiently sharp emitter, electron emission starts below the onset voltage of the formation of the Taylor cone. No stable liquid micro-protuberances were observed in the voltage range between the onset voltages of electron emission and ion emission. If the extraction voltage is increased to attain the value of the onset voltage of the formation of the Taylor cone, an "explosive" emission process is observed, which also results in an alteration of the underlying tungsten tip. If the voltage is further increased, pulse-like electron emission without any indication of the formation of a Taylor cone is observed analogously to the first case. Our investigations verify the model established by Rao et al. [J. Vac. Sci. Technol. B 7 (1989) 1793] on the microscopic scale.

1. Introduction

Liquid-metal ion sources (LMIS) have found important application to microelectronics, for example to the nanometer-scale device fabrication [1], the preparation of structures of low dimensions [2], the field of micro-machining and device transplantation [3] and special analytical techniques as e.g. in situ cross-sectioning of a TEM specimen from an area selected during SIM inspection [4] or submicrometer-resolved secondary ion mass spectroscopy [5]. This is due to the high brightness up to 10^6 A cm^{-2} sr^{-1} of the LMIS.

A lot of theoretical work has been done to calculate the shape of the liquid cone [6–9] to better understand the mechanism of ion emission

SSDI 0304-3991(94)00165-0

[10], to calculate the jet length at the Taylor cone vertex [8,9,11] and to understand the processes of microdroplet emission [12,13]. But only in two cases has an operating liquid metal ion source been observed in situ in an 100 keV TEM [14] and a 3 MeV TEM [15], respectively.

Investigations applying rapid cooling of the Taylor cone below the solidification temperature during ion emission, to freeze in the liquid cone for subsequent TEM investigations, have the disadvantage that the jet-like protrusion rounds off and therefore are not suitable to investigate the shape of the liquid cone in more detail.

The in situ observation of an operating Ga LMIS by Benassayag et al. [15] served as basis for many theoretical considerations. But up to now there is not sufficient coherence between experiment and theory.

It is interesting, too, to investigate liquid-metal emitters in situ in a HV TEM with reversed polarity of the extraction voltage, since the static forces involved in the formation of the Taylor cone are independent of the polarity of the extraction voltage so that one might speculate whether reversed polarity might yield stable DC field electron emission from a stable Taylor cone, or not. Swanson et al. [16,17] and Hata et al. [18–21] have carried out experiments to measure the electron emission characteristics of liquid-metal electron sources (LMES). With respect to emitters with small tip radii ($r < 10$ μm) the two groups differently interpret their experimental results to obtain a stable DC field electron emission from a Taylor cone, or not.

Nevertheless, in situ experiments at LMIS in a HV TEM are suitable to illustrate various electrohydrodynamic phenomena under the influence of a strong electric field. As detailed information about the shape of the liquid cone in dependence on the working conditions is important for theoretical treatments as well as determining optimum working conditions, in the above-mentioned applications, in situ investigations of gallium liquid-metal ion/electron emitters of different tip radii were carried out in both the ion and electron emission modes in the Halle HV TEM. In situ investigations of indium liquid-metal emitters were published elsewhere [22,23].

2. Experimental

The liquid-metal ion/electron emitter and the extraction electrode are fitted into the object plane of the HV TEM (acceleration voltage 1 MV) by means of a special specimen holder (Fig. 1), which enables one to install the LMIS/LMES and the extraction electrode in object level, to connect the LMIS/LMES with a high-voltage and a heating-power supply, and to bias and measure the current of the extraction electrode during ion or electron emission experiments. The distance between emitter tip and extraction electrode is adjustable in the experimental arrangement. In the experiments a small distance of about 0.5 mm was used to decrease the extraction voltage, which is required to extract ions or electrons from the emitter tip. Gallium liquid-metal ion/electron emitters used for these experiments consist of a hairpin-like heating filament with a tungsten tip point welded to the hairpin. The reservoir of the liquid gallium is between the legs of the hairpin heating filament. The most important part of the emitter, the tungsten tip, was prepared according to the instruction given in Ref. [24]. Tungsten emitter tips of radii between 0.5 and 20 μm were investigated. As an example, Fig. 2 shows an emitter with a sharp tip ($r = 0.5$ μm). Emitters with larger tip radii which are also used, are rounded off electrolytically. For the experiments in the HV TEM only those emitters

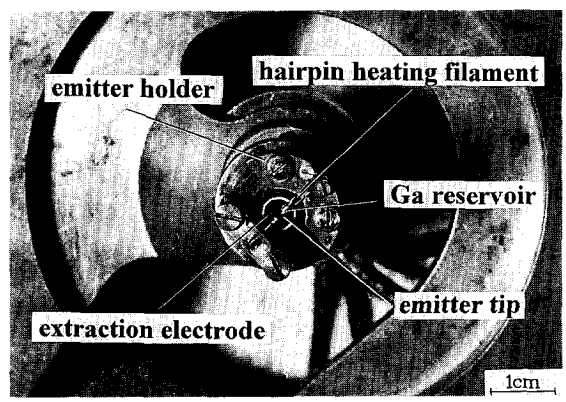

Fig. 1. Specimen holder used for in situ observation of gallium liquid-metal emitters.

are used which are prepared and tested for their emission capability in another vacuum chamber. To evaporate the gallium oxide, which is formed by exposing the liquid-metal ion/electron emitter to air, when moving the emitter from the preparation chamber to the HV TEM, the emitter is heated up to 850°C prior to emission experiments at room temperature. During the in situ experiments in the HV TEM, the surroundings of the special specimen holder was liquid-nitrogen-cooled, to improve the vacuum conditions near the emitter tip ($p \approx 5 \times 10^{-5}$ Pa) yielding a higher stability of the emission current. The images of the shadow projection of the emitter tip were taken by a photo- or video camera.

Because the strong electric extraction field between emitter tip and extraction electrode causes deviations of the imaging electron trajectories, a very high acceleration voltage of 1 MV for the in situ experiments in a TEM was necessary, since the imaging error decreases with increasing acceleration voltage. Our estimations show, that this error is approximately 20 nm [22] roughly corresponding to the value given by Benassayag et al. [15] for their experimental arrangement.

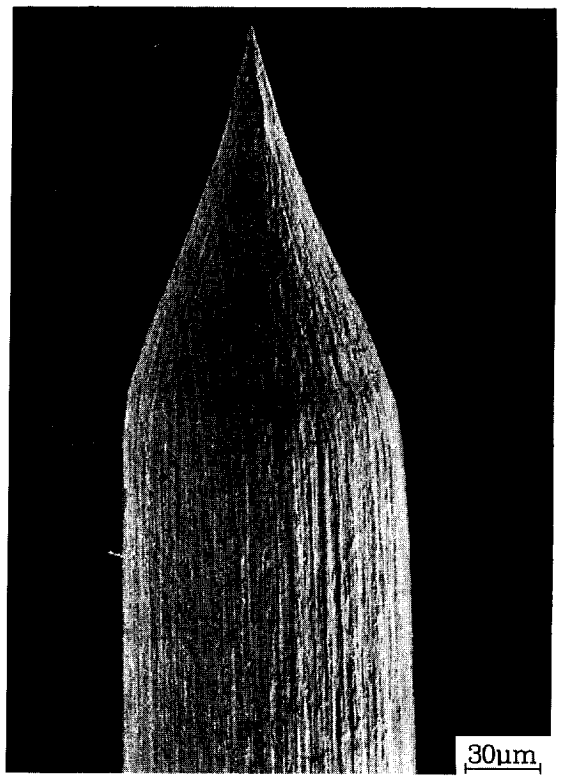

Fig. 2. Tungsten needle electrolytically etched and roughened (tip radius $r = 0.5$ µm).

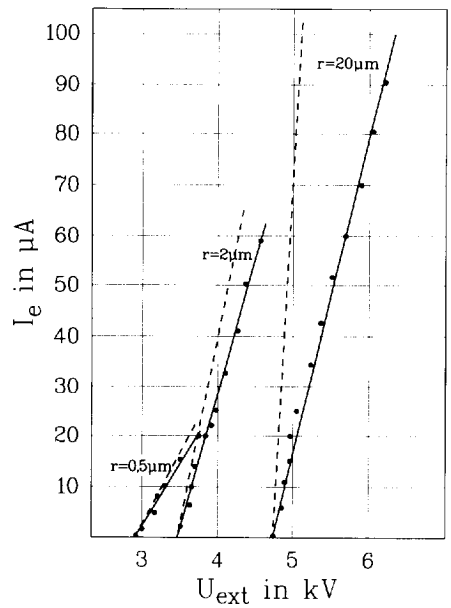

Fig. 3. Total ion emission current versus emitter/extractor potential difference: measured curves (solid lines), analytical ones (broken lines; parameter $\alpha = 1$).

3. Ion emission mode

3.1. Formation and shape of the Taylor cone

Most of the in situ experiments in the ion emission mode are carried out using emitters with tip radii of $r = 2$ µm and $r = 20$ µm, but one emitter with a tip radius of $r = 0.5$ µm (Fig. 2) is also used, particularly for the ion/electron emission experiments mentioned in Section 4.

To characterize the emitters with respect to the liquid-metal flow along the supporting needle, current/voltage curves are measured (Fig. 3). In this figure experimental curves are compared with analytical ones calculated with Mair's formula [30]. The slope of the measured current/

voltage characteristics of the emitters with a tip radius of $r = 2$ μm and $r = 20$ μm are somewhat less than Mair's theory [30] predicts. Viscous drag effects should be taken into account, as the "jet length versus current" relationship (see Fig. 7b) could depend on whether the flow along the

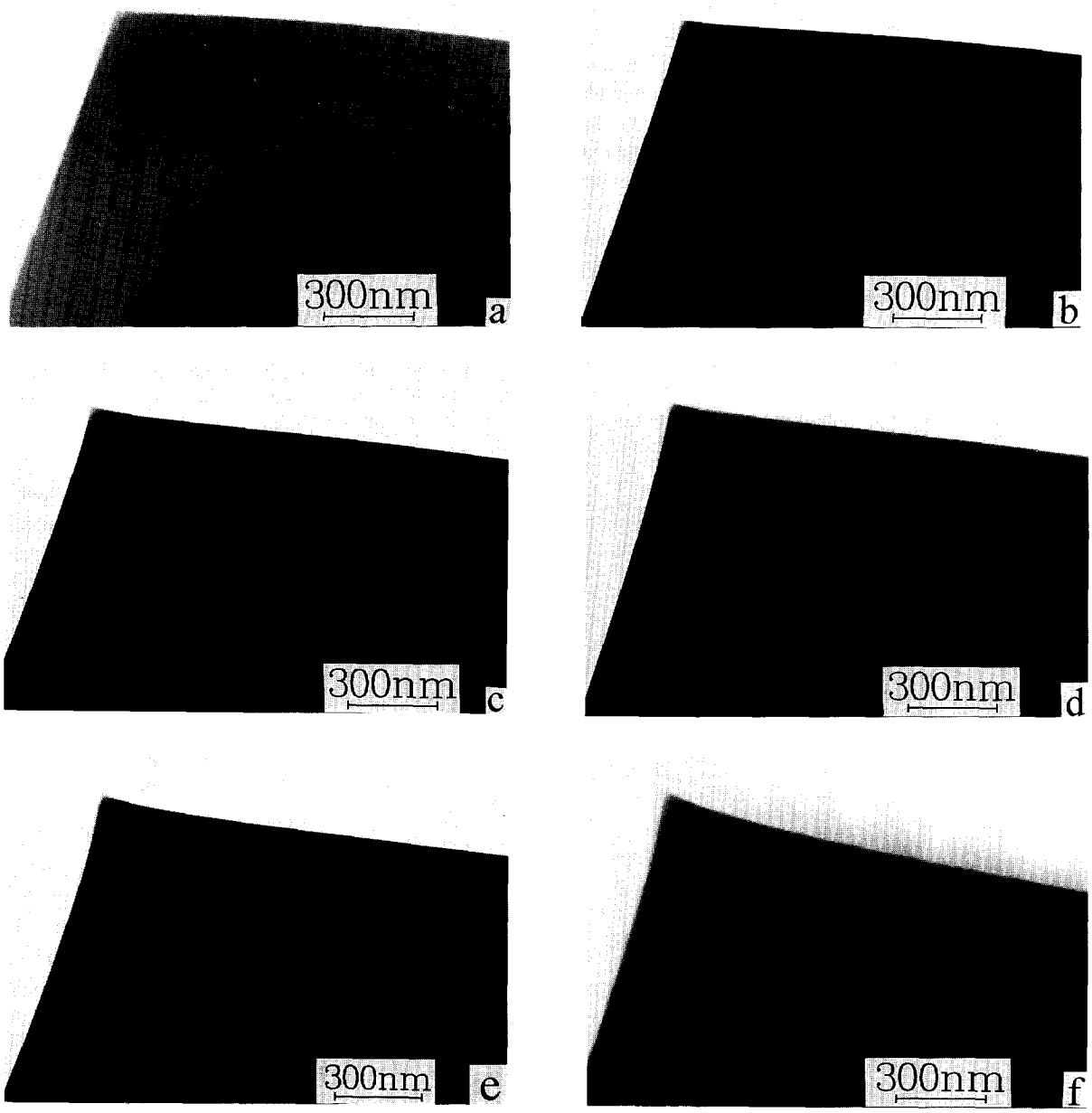

Fig. 4. Shape of the Taylor cone at different ion emission currents (tip radius 2 μm): (a) formation of the Taylor cone (height ≈ 1.4 μm), (b) $I_e = 2.2$ μA, (c) $I_e = 10.2$ μA, (d) $I_e = 15.4$ μA, (e) $I_e = 20.4$ μA, (f) $I_e = 32$ μA.

supporting needle involves significant viscous drag [8].

First of all, results are given that are obtained from in situ ion emission experiments using an emitter with a tip radius of $r = 2$ μm. To observe changes in the shape of the liquid-metal surface on the tungsten tip, the extraction voltage between emitter tip and extraction electrode was

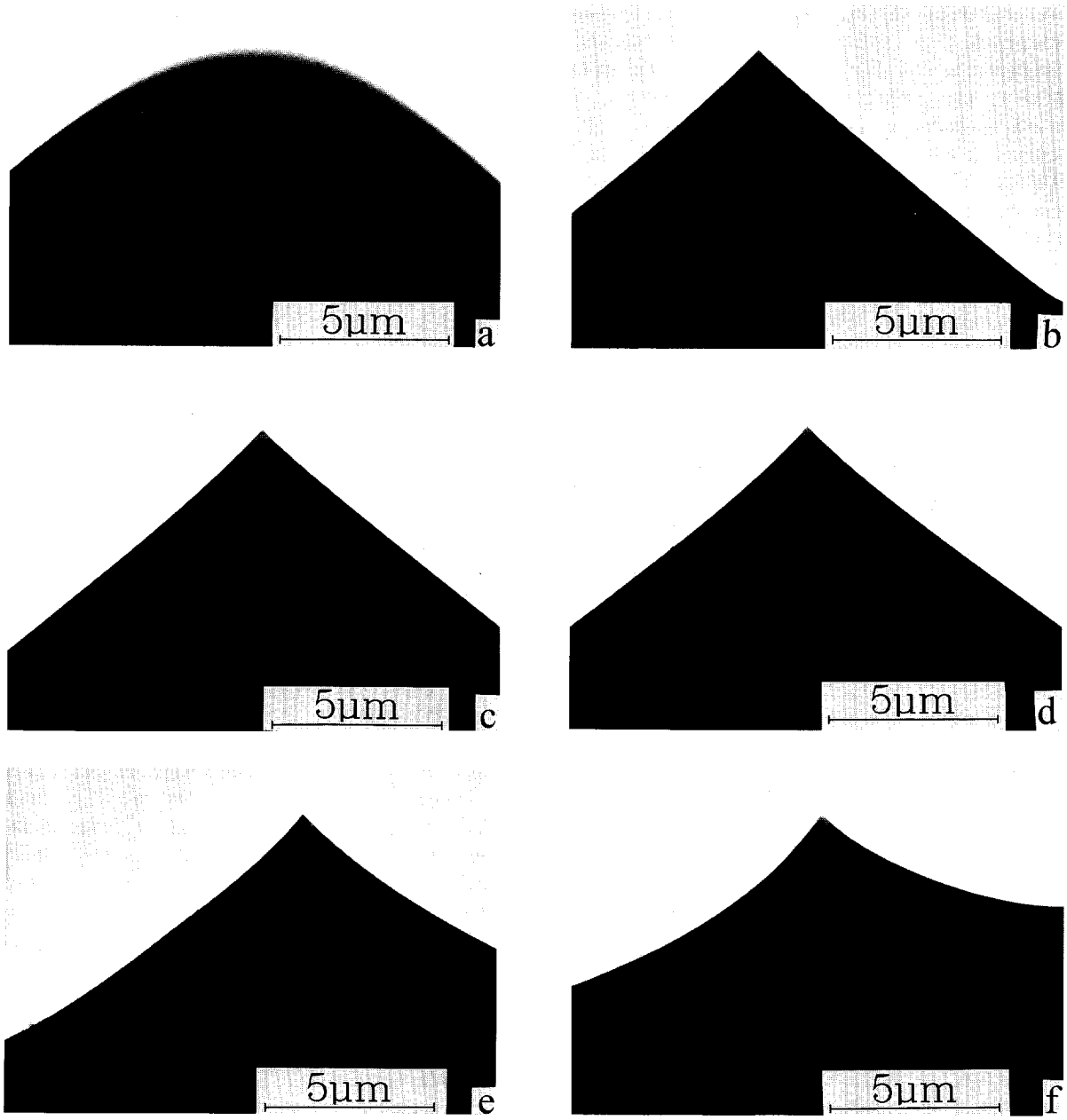

Fig. 5. Shape of the Taylor cone at different ion emission currents (tip radius 20 μm): (a) formation of the Taylor cone, (b) $I_e = 25$ μA, (c) $I_e = 33$ μA, (d) $I_e = 45$ μA, (e) $I_e = 60$ μA, (f) $I_e = 80$ μA.

increased in small steps from zero up to the onset voltage U_c of forming a Taylor cone. In this voltage range the emitter tip, which is coated with the liquid gallium, has a spherical shape, which does not change until U_c is reached. As the onset voltage is applied between emitter tip and extraction electrode the liquid gallium is pulled out (shape like a spheroid), which immediately changes into the Taylor cone [6]. The formation steps of the Taylor cone cannot temporally be resolved, because the formation process of the cone is much faster than the picture frequency (TV standard) used during video recording. But it was possible to measure the height of the cone from a micrograph taken at the onset voltage (Fig. 4a), where the Taylor cone is periodically formed and disappeared during the exposure time of the micrograph. This leads to differences in the brightness between coated tungsten tip and cone. The height of the Taylor cone amounts to 1.4 ± 0.3 μm. But the height of the Taylor cone versus the emission current could not be determined, because the image of the cone shifts on the fluorescent screen, if the emission current is being changed. Figs. 4b to 4f show the shape of the Taylor cone versus the emission current. Table 1 lists all the values measured of both the cone half-angle α and the jet length l versus the emission current I_e in small steps of ΔI_e. The cone half-angle α of the liquid cone decreases from $\alpha = 53.8°$ to 49.3° with increasing emission current I_e. As Fig. 4a shows in the original at the Taylor cone vertex a jet-like protrusion was visible, but its contrast remained weak, making photographic enlargements difficult. The length l of the jet-like protrusion, which is measured from the apex of the underlying Taylor cone (Fig. 6), increases with increasing emission current I_e. The experiments show that an increase in the extraction voltage not only causes an increase of the jet length, but also a decrease of the cone half-angle α.

Table 1
Cone half-angle α and length l of the jet-like protrusion versus the ion emission current I_e (tip radius 2 μm)

I_e (μA) $\Delta I_e = \pm 0.3$ μA	α (deg) $\Delta\alpha = \pm 0.5°$	l (nm) $\Delta l = \pm 20$ nm
on/off	53.8	–
1.2	53.9	–
2.2	53.0	~ 10
3.6	53.5	~ 10
4.1	53.2	~ 10
5	53.0	~ 10
9	51.9	15
10.2	52.0	25
11.0	52.5	30
15.4	51.4	45
18.0	51.5	65
20.4	50.9	75
21.6	50.5	85
25.4	49.9	115
32	49.3	165

Table 2
Cone half-angle α and length l of the jet-like protrusion versus the ion emission current I_e (tip radius 20 μm)

I_e (μA) $\Delta I_e = \pm 0.3$ μA	α (deg) $\Delta\alpha = \pm 0.5°$	l (nm) $\Delta l = \pm 20$ nm
10	52.5	40
12	51.8	50
15	51.4	65
19	51.0	70
20	50.9	75
25	50.0	110
30	49.8	130
33	49.6	150

The results regarding α and l obtained for an emitter with a tip radius of $r = 20$ μm in the emission current range from the onset current up to 33 μA (Figs. 5a to 5c, Table 2) agree with those obtained for the above-mentioned emitter (tip radius $r = 2$ μm). At higher emission currents ($I_e \geq 32$ μA) the cone walls of the Taylor cone become more and more concave (Figs. 5d to 5f) so that a linear approximation defining the cone half-angle α becomes less correct. Therefore only the ion current range between onset current and 32 μA (Tables 1 and 2) is used for fitting the dependence between the cone half-angle α, the jet length l and the current I_e, respectively.

Fig. 6 illustrates the dependence of the cone half-angle α and the jet length l on the ion emission current I_e. The graph in Fig. 7a shows the asymptotic cone half-angle α, whereas that in Fig. 7b illustrates the measured length l of the jet-like protrusion versus the emission current I_e.

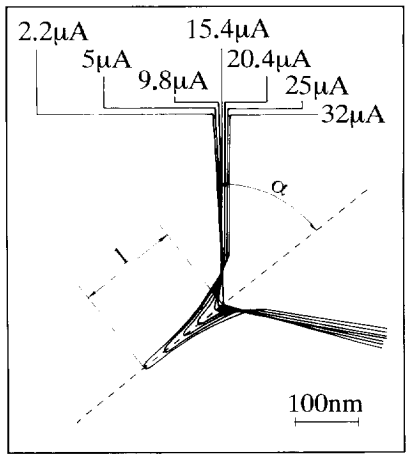

Fig. 6. Shape of the Taylor cone as function of the ion emission current: α, l are the cone half-angle and the length of the jet, respectively.

Note that for gallium the measured jet length variation with the emission current seems to be not entirely linear, which is in agreement with numerical calculations [29]. The fitted curves are calculated by using the following expressions:

$$\alpha = \alpha_0 - (\mathrm{d}\alpha/\mathrm{d}I_e), \tag{1}$$

$$l = (\mathrm{d}l/\mathrm{d}I_e)I_e, \tag{2}$$

with $\alpha_0 = 53.5°$, $\mathrm{d}\alpha/\mathrm{d}I_e = 0.129°/\mu A$, $\mathrm{d}l/\mathrm{d}I_e = 0.0044\ \mu m/\mu A$. The measured values of the asymptotic cone half-angle α versus the emission current I_e (Fig. 7a) agree qualitatively with computer simulations of the ion emission process from a gallium liquid-metal ion source [9]. According to these calculations α changes from 60° to 45° with increasing emission current (I_e varies from 1.5 to 57 μA). The experimental results concerning the jet length versus the emission current (Fig. 7b) are compared with those of Benassayag et al. [15] and with theoretical results [7–9]. We observed a jet-like protrusion near the onset voltage (Fig. 4b). Our straight line fitted to the measured values is in good agreement with theoretical results given by Zheng et al. [9].

Similar dependences were revealed by in situ HV TEM investigations of an indium liquid-metal ion source [22]:

$$\alpha = \alpha_0 - (\mathrm{d}\alpha/\mathrm{d}I_e)I_e, \tag{3}$$

$$l = l_0 + (\mathrm{d}l/\mathrm{d}I_e)I_e, \tag{4}$$

with $\alpha_0 = 51.1°$, $\mathrm{d}\alpha/\mathrm{d}I_e = 0.298°/\mu A$, $l_0 = -0.23\ \mu m$, $\mathrm{d}l/\mathrm{d}I_e = 0.039\ \mu m/\mu A$. For the indium emitter the cone half-angle α decreases by a factor of two relative to that of a gallium emitter and the increase of length l of the jet-like protru-

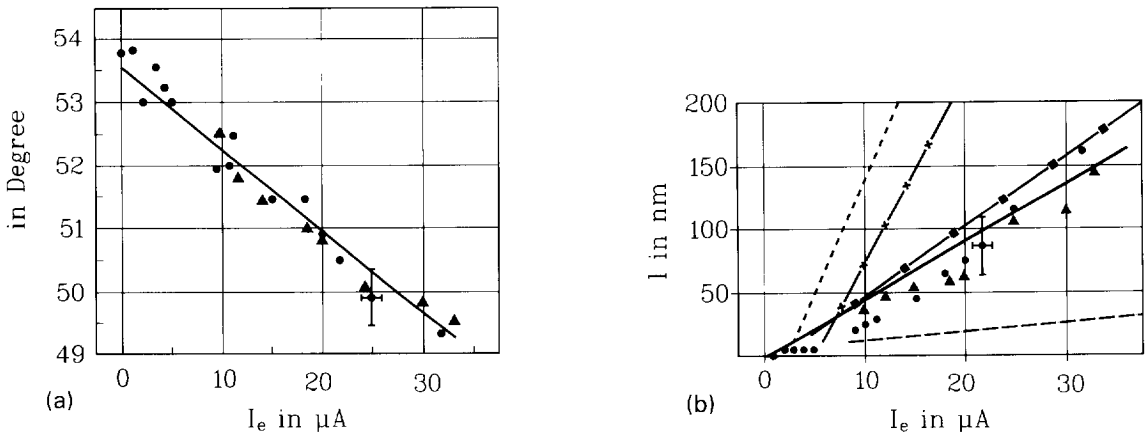

Fig. 7. (a) Cone half-angle α versus the ion emission current I_e: (solid line) curve fitted to the measured values (tip radius: 2 μm/points; 20 μm/triangles). (b) Jet length l versus the ion emission current I_e: (solid line) curve fitted to the measured values (tip radius: 2 μm/points; 20 μm/triangles), (dash-dotted line) experimental results taken from Benassayag et al. [15], (dashed line) theoretical results taken from Kingham et al. [7], (line with crosses) theoretical results taken from Forbes et al. [8], (line with squares) theoretical results taken from Zheng et al. [9].

sion is also about one order of magnitude larger than for the gallium emitter. These differences between Ga and In emitters give experimental evidence of a material dependence of the Taylor cone shape. One reason for this material dependence may be that the In ions are heavier than the Ga ones and therefore move more slowly from the tip and thus cause a stronger space-

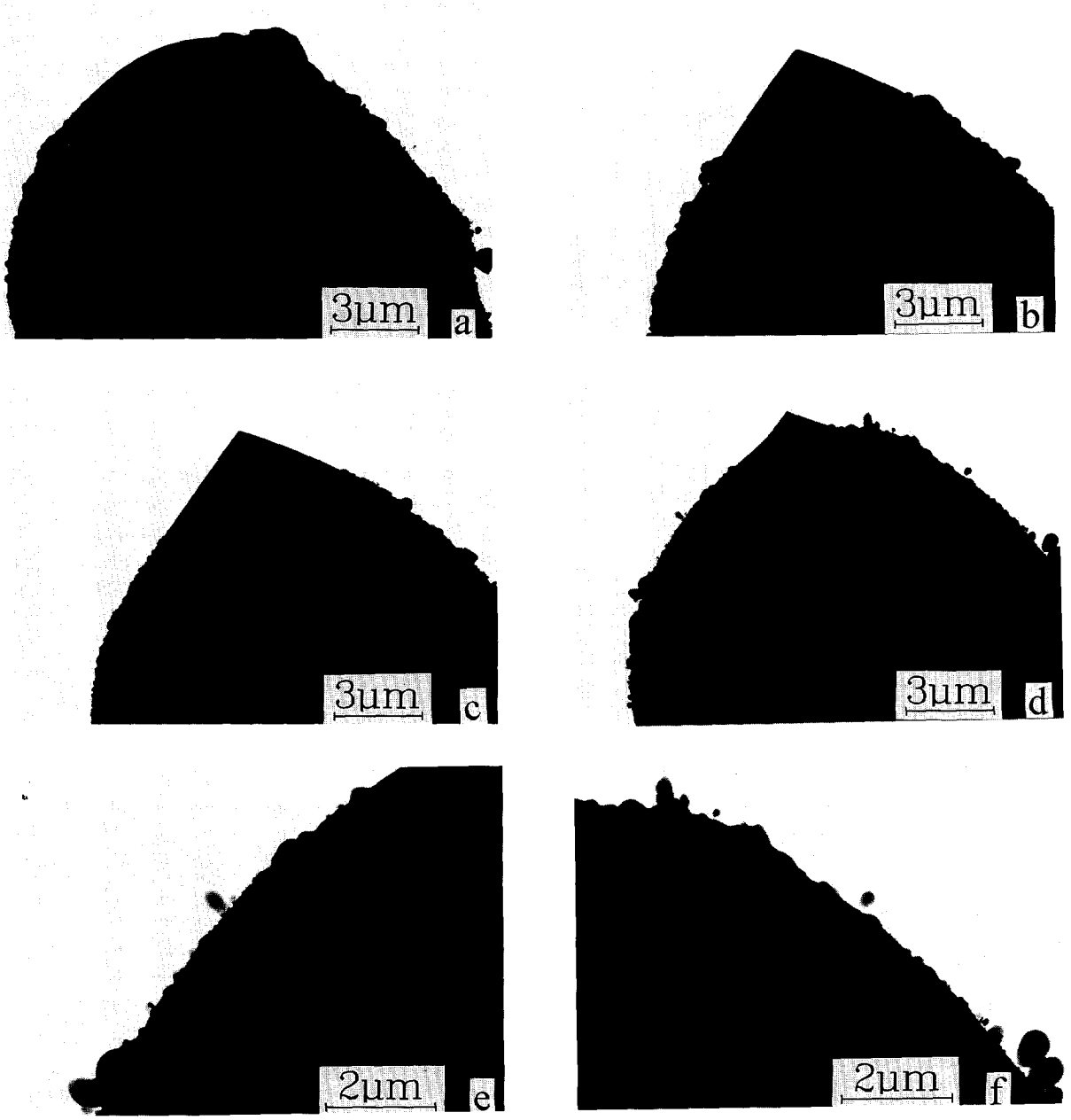

Fig. 8. Microdroplet emission from the surrounding of the Taylor cone at different emission currents (tip radius 2 μm): (a) no emission, (b) $I_e = 1.1$ μA, (c) $I_e = 5$ μA, (d) $I_e = 25.4$ μA, (e) $I_e = 25.4$ μA (left side) (f) $I_e = 25.4$ μA (right side).

charge-induced depression of the surface field because of the greater space-charge effect. A second reason is the difference in the liquid surface free energy between the two elements which too may influence the cone shape and the jet length [11].

3.2. Emission of microdroplets

It is well known that an intensive emission of charged microdroplets from a liquid-metal ion emitter is registrated at high ion emission currents. According to Vladimirov et al. [25] two distinct groups of droplets have been observed, small ones ($r = 1$–10 nm) and larger ones ($r > 100$ nm). At high ion emission currents the jet-like protrusion extends over quite a distance (Fig. 7b) and the jet becomes unstable (Rayleigh instability [26]) according to theories regarding microdroplet emission, see e.g. Ref. [13]. This is one process which may result in the generation of microdroplets ("Rayleigh droplets") having a radius close to that of the jet-like protrusion. Periodic processes of jet decay and re-establishment furthermore cause a pressure modulation in the liquid cone that leads to the parametrical excitation of the capillary waves (Faraday effect [27]) on the surface of the Taylor cone accompanied with large droplets ("Faraday droplets") tearing off the peaks of capillary waves. The size of these droplets is expected to have a value of up to 200 nm [25].

In our TEM the emission of "Rayleigh droplets" created at the jet-like protrusion at the Taylor cone vertex could not be noticed because of the limited lateral resolution (about 20 nm), whereas large droplets were observed to be emitted from the surrounding of the Taylor cone. The process of microdroplet emission is better illustrated by video recording, but the quality of photos made from the video film is not suitable for printing. We therefore used micrographs to prove this emission process (Figs. 8a–8d). We think that the process of exciting the emission of these large droplets is the same as briefly described above for the "Faraday droplets". The radius of these "Faraday droplets" observed varies between $r =$ 40 and 500 nm (Figs. 8e and 8f). A displacement of microdroplets on the emitter surface to the apex of the tip was not observed during the experiments. As video recording shows, trajectories and stability of these droplets have not been observed after their emission because of their high velocity in relation to the picture frequency used for video recording. The video recording technique also shows that the emission of these droplets is correlated with the ion emission process. That means: if the extraction voltage is lower than the onset voltage of forming the Taylor cone, there is no emission of "Faraday droplets". When the onset voltage U_c is reached, the liquid cone is forming, ion emission starts at Taylor cone and "Faraday droplets" are emitted from the surroundings of the cone. Below U_c the arrangement of the "Faraday droplets" at the emitter tip is stable. According to Vladimirov et al. [13] the emission process of "Rayleigh droplets" at the jet causes the modulation of the liquid pressure, and furthermore the generation of large "Faraday droplets". As the observed "Faraday droplet" emission starts with the formation of the cone and ends with its disappearance, we can assume that the emission of "Rayleigh droplets" (not visible in our HV TEM) also starts with the formation of the Taylor cone. This is in agreement with our observation of the formation of the jet-like protrusion at the Taylor cone vertex somewhat above the onset voltage of ion emission.

As video recording shows, with increasing ion emission current the liquid surface near the cone becomes smooth with the emission areas of "Faraday droplets" moving further away from the front side of the tip. It should be pointed out, that during electron emission (Section 4) no emission of "Faraday droplets" was noticed. It should also be pointed out that all these processes of "Faraday droplet" emission observed on gallium emitters are more pronounced for an indium LMIS [22].

3.3. Instabilities of the Taylor cone

During our ion emission experiments we also investigated oscillations of the Taylor cone at the onset voltage, the spatial shifting of the Taylor

cone between two positions on the tungsten tip, and the formation of a double cone. Video recording of these instabilities better illustrates these processes, but the situation is the same as mentioned above. Therefore we used micrographs to illustrate these instabilities. Fig. 4a was

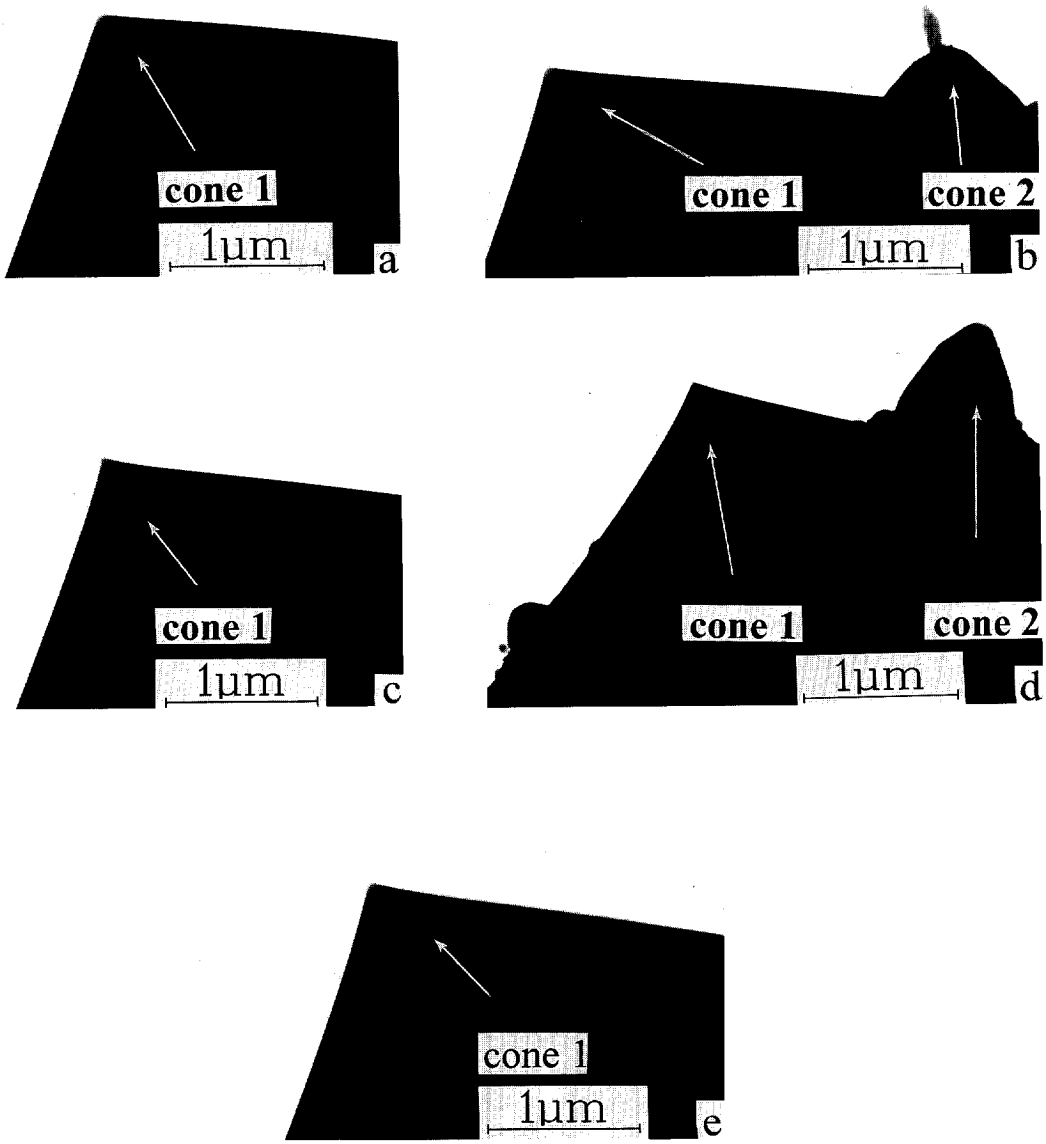

Fig. 9. Formation of a double cone at different emission currents (tip radius 2 μm): (a) $I_e = 5\ \mu A$, (b) $I_e = 9\ \mu A$, (c) $I_e = 10.5\ \mu A$, (d) $I_e = 20.2\ \mu A$, (e) $I_e = 25.4\ \mu A$.

taken near the onset voltage for ion emission, where oscillations of the Taylor cone have been observed. As mentioned above the two states are visible, first with cone and second without cone. Fig. 9a now shows the emitter tip with a stable liquid cone at an emission current of 5 μA. When

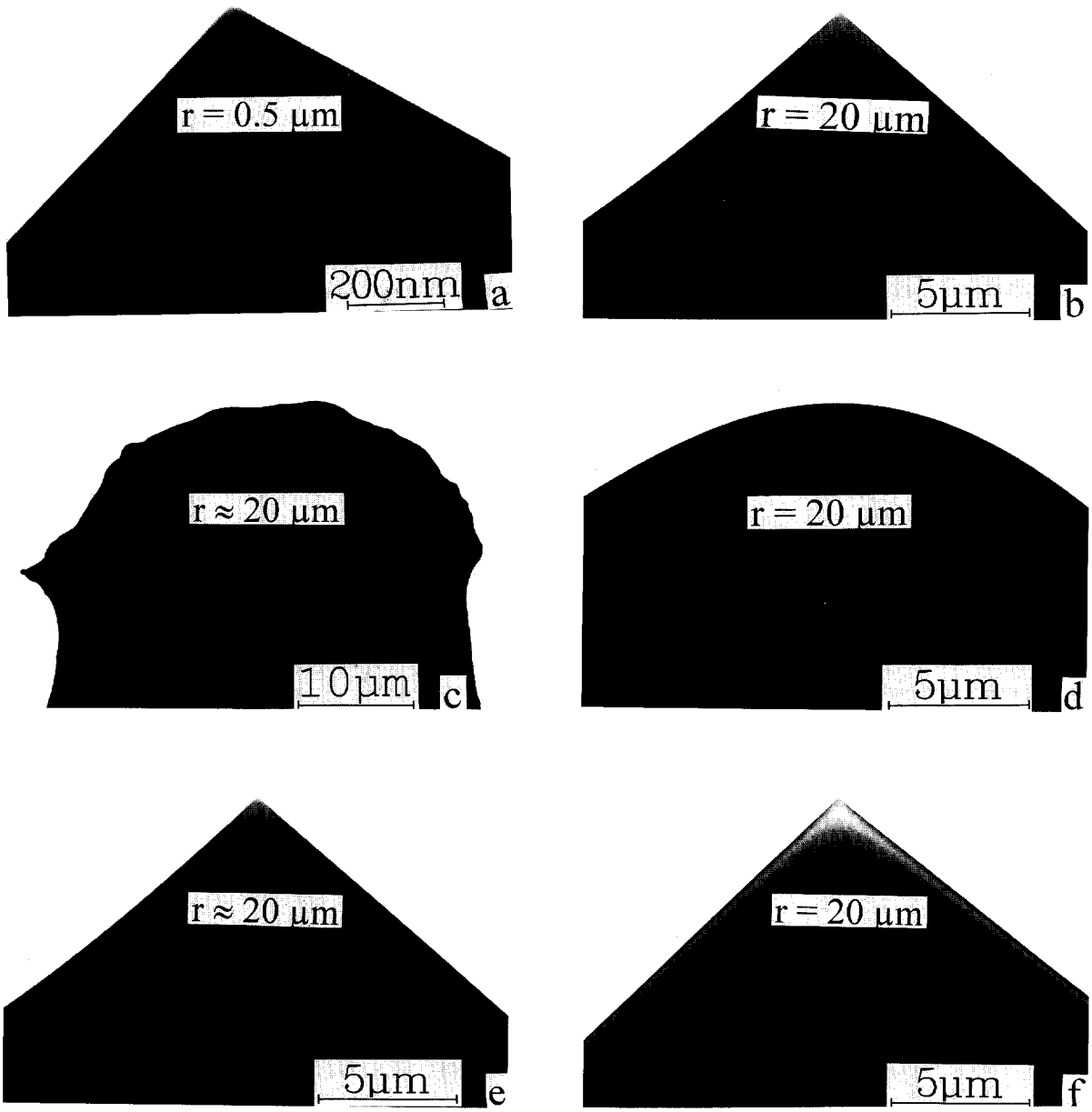

Fig. 10. Shape of the emitter tip during ion/electron emission: (a) $I_e = 5$ μA, (b) $I_e = 12$ μA, (c) $I_e = 35$ μA, (d) $I_e = 42$ μA, (e) $I_e = 25$ μA, (f) $I_e = 20$ μA.

the emission current is increased up to 9 μA a second cone is formed abruptly (Fig. 9b). This second cone disappears at an emission current of 10.5 μA (Fig. 9c), re-establishes a new one at an emission current of 20.2 μA (Fig. 9d), before the second cone disappears again at an emission current of 25.4 μA (Fig. 9e). The double cone is stable over only a short period, then one cone of the double cone configuration disappears. Spatial shifting of the cone between two positions at the tip has also been observed by video recording. These spatial vibrations of the Taylor cone, which are not always associated with current fluctuations, can destroy the brightness properties of the liquid-metal ion source.

4. Electron emission mode

As the process of the Taylor cone formation is independent of the polarity of the extraction voltage, one might speculate whether with reversed polarity on a LMIS one could obtain stable DC field electron emission under moderate vacuum conditions, or not. This case of electron emission has therefore been investigated by different groups, because the idea of having a high-brightness source for both electrons and ions, which may be switched between electron and ion emission, is important. Swanson and Schwind [16] investigated a Ga–In emitter with a tip radius of 20 μm in the electron emission mode. They found only high-current (up to 250 A/pulse) nanosecond length pulsed electron emission which they thought is due to a periodically occurring explosive destruction and subsequent formation of the Taylor cone. Hata et al. [18–21] have reported DC electron emission from wetted needle tips having small tip radii ($r < 10$ μm). They found a stable DC electron emission at low currents (I_e = 200 μA, $U_{ext} = -2$ kV, tip radius $r = 1$ μm) [19]. At higher emission currents (extraction voltage -4 kV) they registrated an explosive emission according to Ref. [16] with the tungsten tip also exploding (tip radius afterwards was $r = 20$ μm). Before destroying the emitter tip they measured an onset voltage for electron emission much lower than that for ion emission. They explained their experimental results regarding sufficiently sharp emitters, that the electron emission is from "micro-protrusions" at the tip. The existence and shape of these "micro-protrusions" is not yet clear. Recently, Rao et al. [17] investigated a gallium and an indium LMIS in both the electron and ion emission modes. In the case of an indium emitter it was possible to freeze the field-stabilized Taylor cone formed during ion emission by rapidly reducing the emitter temperature below the solidification point where the extraction voltage is somewhat higher than the onset value. Field electron emission from the freezed Taylor cone was then realized by reversing the extraction voltage polarity. If the tip, coated with indium, is heated up to the melting point of indium pulse-like electron emission occurred. For sufficiently sharp emitter tips the authors suggested that the electron emission is normal field electron emission from a solid emitter, coated with a liquid-metal layer, rather than from a field-stabilized Taylor cone. They explained the lack of stable DC electron emission from liquid-metal electron sources to be due to the absence of the stabilizing factors of the liquid flow and space charge, which are present in the LMIS but not in the LMES.

To distinguish between emission from "stable micro-protrusions", or a field-stabilized Taylor cone, respectively, and that from a solid emitter tip, coated only with a liquid film, the tip shape of gallium liquid-metal emitters with tip radii of $r = 0.5$ and 20 μm has also been observed in situ in the HV TEM as function of the extraction voltage. First we tested an emitter with a tip radius of $r = 0.5$ μm with respect to its ion emission capability. Fig. 10a shows the shape of the Taylor cone at an ion emission current of 5 μA. Then the extraction voltage was decreased to zero. Thereafter the polarity was reversed and the extraction voltage was increased again in small steps. The field electron emission starts far below that voltage required for the Taylor cone formation. In the voltage range between the onset voltage for electron emission ($U_e \approx 2.3$ kV) and that for ion emission ($U_e = 2.9$ kV) no distortion of the liquid surface has been observed. We therefore conclude that in this case the electron emission is normal field emission from an emitter

tip coated with a liquid gallium layer, which is in agreement with Rao et al. [17]. Applying the onset voltage of the formation of the Taylor cone between emitter and extraction electrode then causes an "explosive electron emission process" like that described by Hata et al. [19]. Only after this "explosive electron emission process" it was possible to observe the shape of the tip, as the imaging process in the electron microscope was not stable during this "explosive electron emission process". As Fig. 10c shows, this process destroyed not only the "liquid cone" but also the tungsten emitter tip below. The tip radius now was $r \approx 20$ μm and a pulse-like electron emission was registrated (no formation of a Taylor cone has been revealed). Fig. 10e shows the shape of the Taylor cone in the ion emission mode, after reversing the extraction voltage.

Fig. 10b displays the shape of the Taylor cone observed on the emitter having a tip radius of $r = 20$ μm in the ion emission mode. In the next experiment applying reversed polarity of extraction voltage the formation of a "bump" was investigated at the onset voltage for electron emission, similar to that during ion emission, if the extraction voltage is increased in small steps over the onset voltage. If the extraction voltage is higher than the onset voltage for ion emission (Fig. 10d) pulse-like electron emission occurs. Here too, no processes have been detected of formation and subsequent degeneration of the Taylor cone, because of the short intervals ($t \approx 5$ ns), during which the cone could be formed. In the following experiment the extraction voltage was reversed and the formation of the Taylor cone is observed in the ion emission mode (Fig. 10f). Our investigations verify the model given by Rao et al. [17].

5. Conclusions

The formation of the gallium liquid-metal cone and the emission of microdroplets outside the cone have been observed in situ in the Halle HV TEM. The cone half-angle decreases linearly with increasing emission current. At an emission current $I_e \approx 2$ μA an additional jet-like protrusion was observed at the Taylor cone vertex, which increases in length with increasing emission current. At emission currents greater than 32 μA further changes in the shape of the liquid cone are observed, making the measurement of the asymptotic cone half-angle difficult. The experimental results are in good agreement with calculations of the jet parameters [9]. Also dynamical effects such as droplet emission and spatial shifts of the Taylor cone are registrated during the experiments. The emission of "Faraday droplets" has been observed in the surroundings of the Taylor cone. The radius of the droplets varied between 0.04 and 0.5 μm. As the observed "Faraday droplet" emission starts with cone formation and ends with the disappearance of the cone, it can be concluded, that the emission of "Rayleigh droplets", too, starts with the formation of the Taylor cone.

Applying gallium liquid-metal emitters with tip radii of $r = 0.5$ and 20 μm in the electron emission mode in no case proved the formation of a stable liquid cone. The onset voltage for the electron emission from an emitter of a radius of 20 μm is roughly equal to that for ion emission leading to the assumption by Rao et al. [17] that in this case the electron emission starts from a liquid cone. This cone is being formed in a short time interval ($t \approx 5$ ns), then it is destroyed, newly formed, and so on. This may be the reason why we cannot observe the cone during our in situ experiments. Using an emitter with a tip radius of $r = 0.5$ μm has not revealed any changes in the shape of the tip (no stable micro-protrusions, no cone) between the onset voltage for electron emission and that for ion emission. This suggests, that in this case the electron emission starts from a tungsten tip, which is modified by a thin layer of gallium, which changes the work function of the tip. If the onset voltage for the formation of a Taylor cone is applied between emitter and extraction electrode, an "explosive electron emission process" follows, which not only destroys the "liquid cone" but also the tungsten emitter tip below. The remaining tip radius is $r \approx 20$ μm and a pulse-like electron emission is registrated. No formation of a Taylor cone is revealed. Our inves-

tigations verify the model concluded by Rao et al. [17] on the microscopic scale.

Acknowledgements

The authors would like to thank Prof. J. Heydenreich (Max-Planck-Institut für Mikrostrukturphysik Halle), Dr. R.G. Forbes (University of Surrey) and Prof. T. Mulvey (Aston University Birmingham) for stimulating discussions. We thank the Deutsche Forschungsgemeinschaft for the support of these investigations. Moreover, we thank Mrs. F. Pabisch (Max-Planck-Institut für Mikrostrukturphysik Halle) for her careful technical assistance in the experiments and for preparing and testing the LMIS.

Note added in proof

Recently, for Ga–In–Sn liquid alloy emitters of a tip radius of 0.2 μm and of a very small amount of the liquid alloy at the tungsten tip, Hata et al. [31] observed the formation of a field-stabilized cone having a base radius much smaller than that of the Taylor cone for ion emission. After having incresed the thickness of the liquid film at the tungsten tip, they observed pulse-like electron emission, which is in agreement with our observations on Ga emitters.

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