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FOCUSED DROPLET BEAM FROM A GOLD LIQUID METAL ION SOURCE

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ABSTRACT

A gold LMIS has been used in the charged droplet emission mode with a single lens focusing column operating at 20 kV beam voltage to achieve a deposit size at the target of 2.3 to 6.0 μm depending on the exposure time. The net mass deposit rate maximized at 1.5 $\mu\text{m}^3/\text{s}$. From the focusing characteristics an energy spread of 50 V and an energy deficit of 350 to 500 V could be inferred for the charged droplet beam.

I INTRODUCTION

Liquid metal ion sources (LMIS) provide a high brightness ion beam which is ideally suited for fine focus applications such as scanning microscopy, ion microlithography, micromachining and high resolution SIMS. Singly and doubly-charged ions, charged molecular clusters and droplets are present in the beam to varying degrees as a function of total emission current.

Originally, the charged droplet emission mode was studied as a possible source for electric space propulsion [1], however, the lower current ion emission mode soon dominated the research interests. Nevertheless possible applications for large area [2] and localized [3] metal film deposition has caused a small but continued interest in the droplet emission mode. Mahony and Prewett [3] used a focused gold droplet beam at an emission current $I=600\text{ }\mu\text{A}$ to obtain a 300 μm wide focused beam of droplets. Recently D'Cruz et. al.[4] investigated various properties of the emission characteristics of the droplet emission mode of a gold LMIS. They gave an estimation of the droplet virtual source size of 8 μm along with other emission characteristics and obtained gold films with fine texture morphology. An important conclusion of their work was that the droplet emission occurred from the apex of the elongated Taylor cone.

In this work, we examine the properties of both unfocused and focused beams of charged droplets from a Gold LMIS. The current dependence of the droplet size and the corresponding droplet emission rate were determined. From the focused beam results the energy deficit and broadening of the droplet beam could be estimated.

II EXPERIMENTAL METHOD

The gold LMIS used was a truncated, tungsten needle type with a $25\mu\text{m}$ radius flat ground on top of a 90° cone. The needle was spot welded to a tungsten wire spiral containing the gold reservoir which allowed flow onto the shank of the needle. When a high positive voltage, greater than the threshold value, is applied to the needle the liquid meniscus forms a cone-like shape and the well known ion emission occurs at a few μA of current. This current can be varied widely from 1 to several hundred μA . At low current the beam consists mainly of Au^+ and Au^{++} , but with increasing current other species such as neutral atoms, charged molecular clusters and droplets appear in the beam [5].

The droplet emission becomes important only for large current and a deposit occurs when the deposit rate exceeds the ion sputtering rate. Because the LMIS current is usually limited by the liquid metal supply, the use of a large needle radius to enhance the flow rate to the emitter apex has been suggested [3]. The particular source shape used in this investigation provided a stable and long life droplet source.

A single, electrostatic lens column, shown in Fig. 1, was used to focus the droplets. During operation the vacuum level of the column was maintained at 1×10^{-7} torr using 200 l/s vac-ion pumping system. Combined beam deflection and astigmatism correction was carried out with an octupole stigmator/deflector. The optical properties of the asymmetric lens have been determined elsewhere [6]. For this study a beam acceptance half angle of 2.0 mrad ($140\text{ }\mu\text{A}$ diameter aperture) was used. The large emitter radius required a relatively high extraction voltage of 15 to 20 kV for the indicated geometry; for this study the first element of the lens was grounded, thereby allowing the more convenient einzel operating mode to be used.

In addition to the focused beam investigation, a simple diode deposition study similar to D'Cruz, et. al. [4] was carried out. In this study the gold LMIS was positioned 1 mm from a flat silicon (111) wafer and the gold deposit examined by SEM.

III EXPERIMENTAL RESULTS

A. Diode Results

The first studies of the droplet emission were performed in the close-spaced diode system. After operating the gold LMIS for 120 s at a current of $150\text{ }\mu\text{A}$ the silicon target was examined in an SEM. Fig. 2 shows an SEM photo of the gold deposit on the silicon target which exhibits a $60\text{ }\mu\text{m}$ diameter bright-centered gold deposit surrounded by a larger dark circle of sputtered silicon surface. Beyond this circle the brightness is due to the thermally deposited gold. In agreement with D'Cruz, et. al, we found a 2° droplet beam divergence in contrast to a 40° divergence for the monomer ion beam.

Using a Sloan Dektak profilometer the volume deposit rate and angular flux were calculated to be $610\text{ }\mu\text{m}^3/\text{s}$ and $6.7 \times 10^5\text{ }\mu\text{m}^3/\text{s/sr}$ respectively. The latter is in excellent agreement with the value of $5 \times 10^5\text{ }\mu\text{m}^3/\text{s/sr}$ reported by D'Cruz, et. al. for a gold LMIS operating at $135\text{ }\mu\text{A}$. As discussed below, assuming an average droplet size of $0.5\text{ }\mu\text{m}$ one can calculate an average droplet emission rate of $\sim 1\text{ kHz}$, also in agreement with D'Cruz et. al. If one further assumes that on the average each particle carries a single charge, then one can calculate that within the 1° acceptance half angle there is $\sim 10^{-2}$ charged particles per atom deposited in droplet form; thus, one would expect to eventually reach an equilibrium size of deposit where removal by sputtering and deposition are equal.

B. Focused Droplet Results

In a preliminary experiment with the focusing column, the emitter was operated without any focusing in order to study the current dependence of the droplet size and flux. The beam acceptance was about 2 mr and the source to silicon target distance was 18 cm. The ion source was operated at different current levels from 100 to 300 μA for 120 s. From the SEM photographs of the droplet deposit on the target, we obtained a size distribution of the droplets as shown in Fig. 3. These results show that the droplet size is relatively uniform throughout the field of view and increases from a mean value of 0.25 to 1.75 μm as the current increases from 100 μA to 300 μA . The average number of droplets per unit area was surprisingly constant at a rate of ~ 0.056 droplets/ $\mu\text{m}^2/\text{s}$ and independent of emission current in the range investigated.

In the focusing experiments the scanning ion microscope image (formed by the secondary electrons) was used to focus the ion beam as perfectly as possible; next, droplet deposition on various target positions was performed as the beam was slowly underfocused and then overfocused. After examination of the target in the SEM we found that when the ion beam was focused the droplet beam was overfocused and that the best droplet focus for 150 μA emission current was about 500 V (in terms of lens excitation) below the best ion focus. This result agrees with studies of the energy properties of gold clusters (up to Au^{5+}) emitted from LMIS, which have an energy deficit, depending on the cluster size and charge, up to several hundred volts [5].

Next an attempt was made to measure the focused droplet deposition rate and deposit shape. SEM photographs in Fig. 4 show a typical gold deposit and its corresponding profile for a 120 s exposure time. The deposit shape had a roughly Gaussian profile and therefore could be characterized by a full width at half maximum (FWHM) and deposit thickness height (h). Table I summarizes the deposit characteristics as the exposure time was varied from 10 to 120 s.

Fig. 5 gives a graphical representation of the variation of deposit volume and ratio of h/FWHM with deposit time. Interestingly, the deposit volume does not increase linearly with time, but shows a short induction period, followed by a linear increase with time and then a sharp leveling off for $t > 60$ s. Similarly, the droplet shape, after initially exhibiting a shallow deposit shape with a small value of h/FWHM , became more peaked as h/FWHM increased to a value of 9 and then remained constant with further exposure. The deposit rate maximized at $\sim 1.5 \mu\text{m}^3/\text{s}$ at a 60 s exposure. As expected this value of deposit rate is smaller than the $610 \mu\text{m}^3/\text{s}$ rate for the unfocused case by the ratio of the solid angle of the apertured to unapertured beam (i.e. $(.002/.017)^2$).

In another experiment we attempted to characterize the effect of the droplet energy spread on the deflected beam due to the transverse chromatic aberration effect. Fig. 6 shows a gold deposit on axis and at 0.5 mm distance from this axis. The off-axis deposit is clearly distorted along the deflection direction as expected. The deposit elongation (Δd) was obtained from profile SEM analysis of the Fig 6 deposits to roughly estimate the beam energy broadening (ΔV) by

$$\Delta V/V = \Delta d/d$$

where d is the on axis beam size. A value of $\Delta V = 50$ V was obtained for the energy spread of the charged droplet beam.

We were also able to make an estimate of the energy deficit of the droplet beam relative to the ion beam by noting the difference in deflection amplitude between the ion and droplet beams for fixed deflection

and beam voltages. Accordingly, the relative energy deficit was estimated to be 350 V for the droplet beam at 150 μ A emission current. This is in reasonable agreement with the less accurate method of noting the change in focus voltage between the droplet and ion beams which gave a value of $\sim$ 500 V as mentioned earlier.

Fig. 7 demonstrates the ability to deflect the focused beam to form 100 μ m long deposited gold lines. The two lines of equal length were obtained by using sweep times of 70 and 140 s and an emission current of 150 μ A. The width of the lines are 4 and 6.5 μ m respectively. Their morphology is very rough but a profile view indicates that there is no discontinuity.

IV DISCUSSION

At 150 μ A of emission current droplet emission was of the order of 1 to 10 kHz in agreement with the relaxation times associated with the formation of the Taylor cone as pointed out earlier by D'Cruz, et. al. The droplet emission occurs with a very high flux due to its small angular confinement. Although there is a range of droplet sizes, the average size increases from 0.5 to 2.0 μ m with increasing current. Thus, a relatively large portion of the few nanometer sized Taylor cone apex is removed during droplet formation leading to total mass flow rates of $\sim$ 600 to 1000 μ m³/s. The details of the droplet formation mechanism are not completely clear at this time, but undoubtedly involve hydrodynamic instabilities in the liquid cone as the mass flow rate increases above a critical value. The somewhat larger energy deficits associated with the charged droplets suggest its formation occurs further out from the position where ions are formed. However, in view of the fact that the energy spread is quite narrow, of the same order as the monomer ions (50 to 100 V) [7], the droplet formation occurs in a relatively small increment of distance from the emitter surface. For example, if the electric field is $\sim$ 2 V/ $\text{\AA}$ the droplet formation occurs within a 100 $\text{\AA}$ region positioned 600 to 1000 $\text{\AA}$ from the reference surface.

In view of the concomitant emission of both charged droplets and charged monomer ions simultaneous deposition and sputtering are occurring for both focused and non-focused beams. In fact because the ion flux is several orders of magnitude greater than the droplet flux and traveling at a much higher velocity, the droplets will be sputtered to some extent during flight. The results suggest that for both focused and nonfocused modes the deposit gradually forms into a particular equilibrium shape such that the sputtering and deposition rates are equal. The dependence of sputtering on angle of incidence probably determines the slope of the peaked deposit shape when steady state is achieved.

The focused beam size d is limited by the chromatic aberration coefficient C_{c_0} of the lens and the energy spread of the droplets as follows:

$$d_c = MC_{c_0} \alpha \Delta V / V$$

where M is the overall magnification of the column. The values of M and C_{c_0} are 2.67 and 187 mm respectively. Thus, for $\Delta V = 50$ V and $V = 20$ kV one calculates a value of $d_c = 2.5$ μ m - a value that is in good agreement with the Table I results for the 10 s exposure deposit. From their studies, D'Cruz et. al. were able to conclude that the virtual source size (d_v) of the droplet source was < 8 μ m; from this investigation we can conclude that $d_v \ll 2.5$ μ m. Because of the surprisingly small energy spread of the droplet beam one can readily deflect the beam over small distances (< 0.5 mm) without appreciable beam broadening due to transverse chromatic aberration. With reduction of C_c further reduction of the on axis beam size can be realized.

V CONCLUSION

The ability to focus the charged droplets emitted from a gold LMIS has been demonstrated. Despite the limit of chromatic aberration a focused beam size of $\sim 2.5 \mu\text{m}$ with a mass flow rate of 1 to $1.5 \mu\text{m}^3/\text{s}$ has been demonstrated by a simple single lens column. The use of this phenomena to provide means to pattern small areas with metal films is obvious. For example, a combination of droplet mode emission for mass deposition and submicron, focused monomer ion beam mode emission for micromachining and trimming of the deposit can be envisioned.

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TABLE I

Deposit dimensions for focused beam results at 150 μA emission current and 20.2 kV beam voltage.

Exposure time (s)	FWHM (μm)	Thickness (μm)	Maximum Volume (μm^3)
10	2.3	0.27	1.62
30	4.5	1.6	25.9
60	5.8	2.4	92
120	5.8	2.4	92

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1. Diagram of the single lens focusing column. The suppressor electrode was removed for this study.

2. Gold LMIS deposit on a Si target placed 1 mm from the emitter. Emission current and exposure time were 150 μA and 120 s respectively.

3. Distribution of droplet size for the indicated emission currents and 120 s exposure time. A target area of $520 \mu\text{m}^2$ was examined and the droplet size increment was $0.5 \mu\text{m}$.

4. Typical shape of the focused beam deposit showing top (a) and side (b) views for a 60 s exposure at 150 μA emission current and 20.2 kV beam voltage.

5. Plots of the ratio of the deposit height to width and deposit volume vs. time for a focused droplet beam at an emission current of $150 \mu\text{A}$ and 20.2 kV beam voltage.

(a)

(b)

6. SEM photos of the focused beam deposit on axis (a) and deflected 0.5 mm off axis (b) for a 20.2 kV beam voltage. The exposure time was 120 s and beam current was $150 \mu\text{A}$.

7. SEM photos of a line scan of the focused droplet beam for scan rates of 1.4 (a) and 0.71 (b) $\mu\text{m/s}$ at an emission current of 150 μA and 20.2 kV beam voltage.