STUDY of MICRO-and NANO-SCALE TRANSPORT of LIQUID METAL on THIN SOLID FILMS

In recent times, researchers have found potential in using liquid metals in broad-spectrum applications such as processing ceramics, stretchable electronics, 3D printing, coolant technology, energy storage, and many others. Gallium and its alloys are preferred over conventional mercury in all these applications because of its low melting point, high boiling point, and non-toxicity. Given these exciting applications, there is a need to understand the flow behavior of low melting metals in the liquid state, especially at small length scales. The wetting of gallium on metal thin solid films is not well understood. Wetting of liquid metals on thin metal films is complex due to the nature of the interaction between the liquid and the substrate. This interaction involves solidification and chemical reactions, including oxidation of the liquid metal itself.

The central theme of this thesis is to understand the liquid metal flow on thin solid films due to various driving forces, such as wetting, chemical reaction, and electromigration. This quest was pursued because of the versatile nature of the phenomenon, which produces patterns ranging from distinct islands to periodic patterns at microscale and nanoscale (see Figure 1a). Given its interesting and technologically important features, we pursued our research to understand the fundamentals of this phenomenon.

In the first part of the work, we investigate the formation of patterns of liquid metals with the rippled surfaces on the metal thin solid films. Flow is observed *in situ* using a scanning electron microscope (SEM) at high magnifications to gain fundamental insights into the phenomenon of the spontaneous flow of the liquid metal and the formation of the unique surface features (see Figure 1b). SEM characterization indicates that the liquid-solid reaction and wetting front drives the entire flow and thereby pattern formation. To further understand the type of materials interaction during the liquid metal flow, characterization of the features formed in multiple systems, such as Ga-Pt and Sn-Pt, is performed using the energy dispersive spectroscopy (EDS) inside a scanning transmission electron microscope (STEM). All material systems demonstrated similar features of chemical reaction taking place between the liquid metal and solid thin film similar observations and enrichment of the top layer of the solidified flow pattern by the substrate metal film. Based on the observations, a finite element (FE) model is developed, whose predictions are compared with the experimental observations (see Figure 1c). The predictions match qualitatively well with the experimental findings, thereby confirming the important role of wetting, chemical reaction, and formation of semi-solidus membrane on top of the liquid film on the flow as well as ripple-pattern formation. Further, we performed an optimization study for controlling the features of surface ripples and implemented the inferred techniques for creating Ga ripple patterns with gaps as small as 100 nm.

In the second part of the work, the electric current-induced liquid metals flow or liquid electromigration is studied. Liquid electromigration is a special case of liquid metal flow on thin solid films, wherein the applied electric field provides an additional force for flow, along with the liquid-solid wetting and chemical reaction at the flow front. Liquid electromigration is considered to be a diffusion-controlled phenomenon, and this work is conducted with an objective to improve the understanding of the phenomena by examining it from a continuum-based perspective. Herein, experiments are designed to observe the behaviors similar to pressure-driven liquid flow in the open channel, with the pressure replaced by the electric current (or electromigration "force"). The observations, with a custom-made experimental setup, are carried out *in situ* inside an SEM as well as an optical microscope. *In situ* studies indicate the formation of an immovable layer on the top of the flow (see Figure 2a). Flow splitting and selective wetting of the thin film are demonstrated via modulation of the current density (see Figure 2b). Liquid metal flow front velocity is characterized for different geometries of the metal track, and the obtained profile is correlated with the current density value. Although liquid electromigration has some remarkable similarities with the pressure-driven open-channel flow and intuition about electric-field induced liquid metal flow can be built using the continuum ideas, the order of magnitude calculation suggests that the driving force for liquid electromigration cannot be converted into an equivalent pressure or body force term that can be used in the continuum fluid mechanics equations (e.g., Navier-Stokes equation) to quantify liquid electromigration. Therefore, the diffusion treatment remains the most accurate way for quantifying the liquid electromigration.

In the last segment of the work, we develop a new technique based on the principle of liquid electromigration and liquid metal wetting of thin solid films to transport liquid metal from a liquid metal pool over a metal-coated needle and then "back" to a location of choice by application of an electric current (see Figure 3a and 3b). The method is helpful in conformally coating the metal films, establishing electrical connections, and bridging a gap at the microscale and, potentially, at the nanoscale. The mechanism is analogous to a "liquid" dropper which uses suction to collect liquid and "release" to dispense a liquid, with, herein, the suction replaced by the liquid electromigration and wetting. The “sucked” liquid metal can be transported for the bridging process to form different electrical connections as well as standalone metallic structures (see Figure 3c and 3d). An experimental setup is developed, and the proof of concept for a method of bridging gaps at two different length scales is given.

In summary, the work contributes to both the fundamental understanding of liquid metal flow on thin solid films and taking advantage of the flow physics for technology development at micro-and nano-scale.

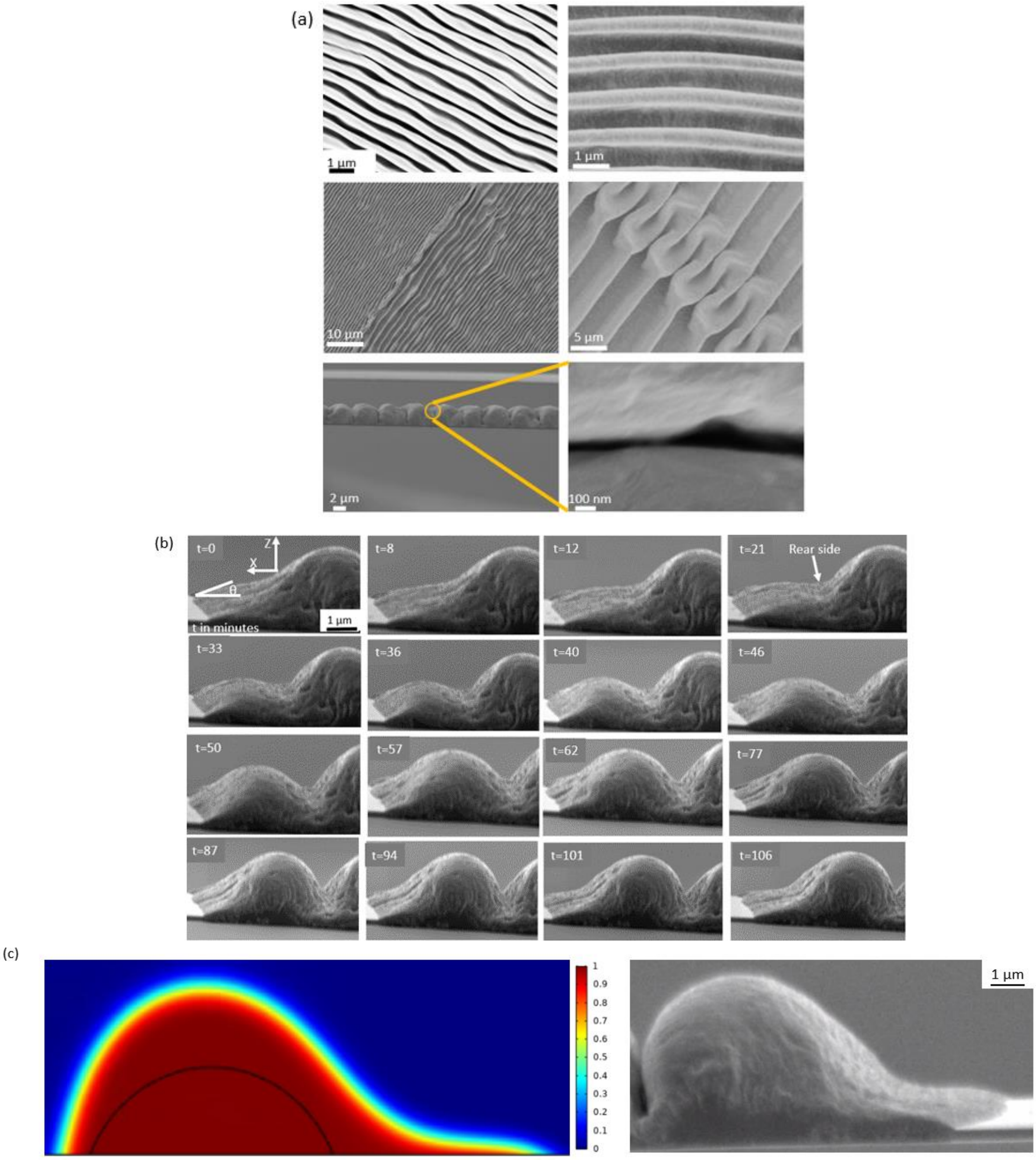

Figure 1 Few important results on the liquid metal ripple patterns: (a) SEM micrographs showing the various possibilities of ripple patterns when Ga flows on Au or Pt thin films (b) Time-lapse in situ SEM micrographs capturing the formation of a single ripple of Ga flowing on Au thin film. (c) Comparison of the FEM simulation carried out in COMSOL and the experimental result. The asymmetric shape predicted from the simulation is in good agreement with the actual scenario.

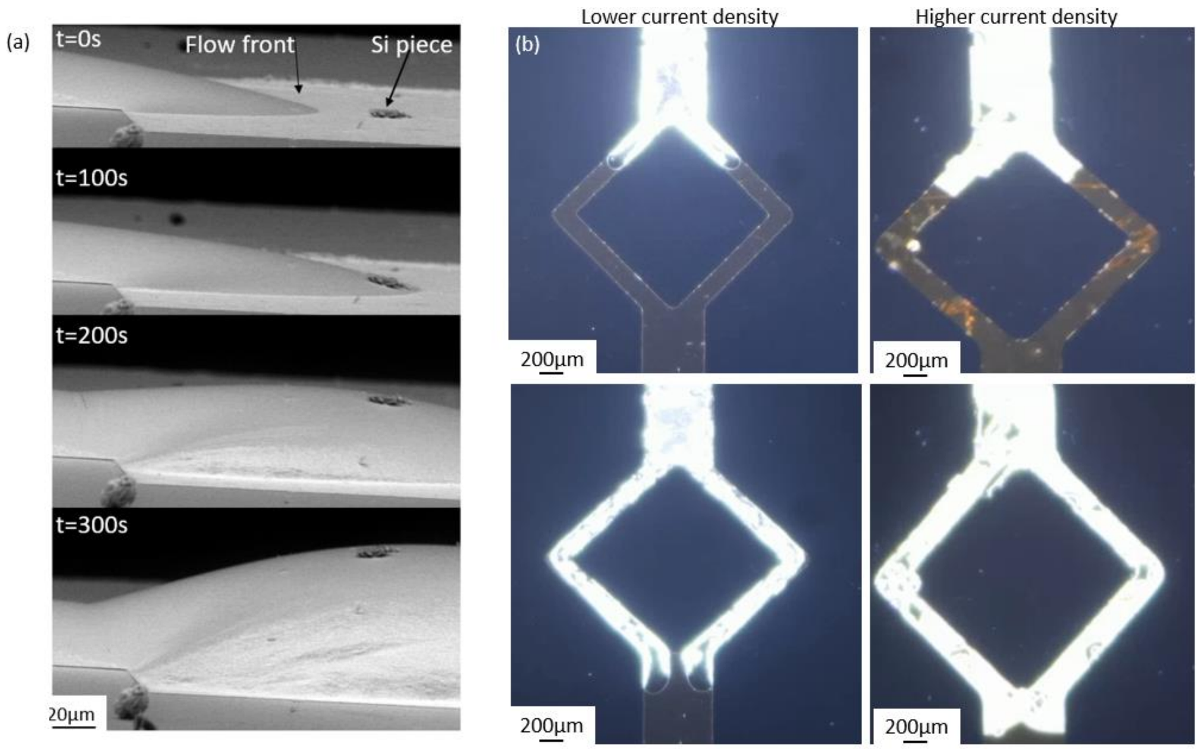

Figure 2 Few important results from the studies on liquid electromigration:(a) In situ SEM micrographs of Ga flowing on Au thin film, formation of an immovable layer at the top of Ga is observed. (b) Optical micrographs of liquid Ga flowing on Au track under electric field on splitting and merging tracks. Distinct behavior of flow front due to the change in applied current density is observed.

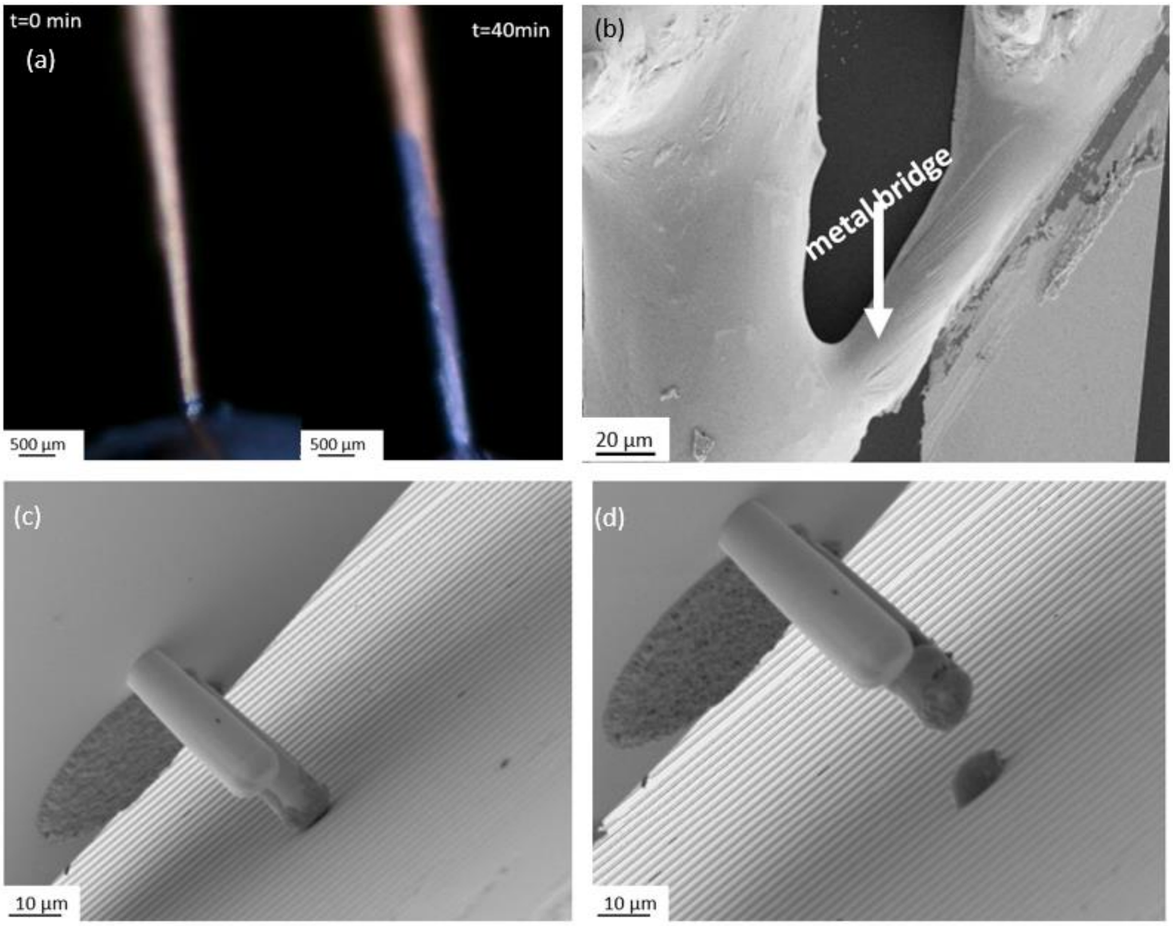

Figure 3 Few important results of Electrosoldering: (a) Optical micrographs of W needle at two different instances of suction of Liquid Ga. At t=40 min Ga is collected on the W needle. (b) SEM micrograph of metal (Ga) bridge between 2 Au lines. The bridge is established by flowing back of Ga from the W needle. (c) SEM micrograph of Si microneedle in contact with Au thin film. (d) SEM micrograph of transferred liquid Ga (an approximate volume of 1 pL) on Au thin film.