Mechanistic insights into nanobubble dynamics

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出版情報：Kyushu University, 2021, 博士（工学）, 課程博士
バージョン：
権利関係：

（様式２）

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論 文 名　：Mechanistic insights into nanobubble dynamics
　　　　　　(ナノバブルの動的挙動に関する研究)

区　　分　：甲

論　文　内　容　の　要　旨

Nanobubbles are the nanoscale gaseous or vapor domains enclosed by liquid interface or liquid-solid interfaces. Recent developments in nanofabrication and instrumentation capabilities have revealed several behavioral oddities for nanobubbles and gas cavities at the nanoscale. Therefore, the present course of the research related to nanobubbles is to understand the fundamentals of their existence and physical characteristics, so that their application can be expanded to domains related to thermal management of electronics, efficient phase change, medical healthcare, and various others. Previously, several researchers have devoted efforts towards understanding the counterintuitive stability and longevity of surface nanobubbles. However, most of the existing studies do not reveal the dynamic behavior of the surface nanobubbles, and the gas transport at nanoscale.

Therefore, this thesis is focused on studying the dynamic behavior of surface nanobubbles by directly visualizing these nanoscopic species using in-situ liquid-phase electron microscopy. Liquid phase electron microscopy is a direct observation technique which allows the observation of the fluids with high spatial and temporal resolution. Thus far, the major understanding about surface nanobubbles and their behavior have been revealed using atomic force microscopy, which inherently has a low temporal resolution. The ability to directly visualize surface nanobubbles at real-time scales has the potential to reveal their dynamics and provides the opportunity to comprehend their oddities and other physical characteristics. Therefore, this thesis aims at augmenting the understanding of surface nanobubbles by adding the dynamic component. Overall, the thesis is composed of 5 chapters.

In **Chapter 1**, the background and important literature on surface nanobubbles is showcased, and existing knowledge on their characteristic is discussed briefly. In addition, various surface nanobubble visualization techniques are compared, and their important findings are discussed. Several advantages of the surface nanobubbles are also discussed. The chapter concludes by framing the research objectives and the scope of the work presented in this thesis.

In **Chapter 2**, the focus of the study is on gaining insights into the nanobubbles' interfacial dynamics. The experiments are conducted using a commercially available silicon nitride liquid cell electron microscopy system, and the surface nanobubbles are nucleated "in situ" at the silicon nitride-water interface via radiolysis of water due to the TEM's electron beam. Further, addressing the probabilistic nature of surface nanobubble nucleation in previously reported liquid phase electron microscopy experiments, a new methodology is devised which allows to nucleate nanobubbles in user defined size and number density bands. Next, an interacting nanobubble pair having radii less than 50 nm is studied which reveals the unique pull-push dynamics of the nanobubbles. Interestingly, freely growing-shrinking nanobubble and pinned nanobubble are observed at identical experimental conditions, suggesting the possibility of multiple nanobubble stabilization theories and pathways. The study also reveals that a freely growing-shrinking nanobubble induces anisotropic depinning in the three-phase contact line of a strongly pinned neighboring nanobubble.

In **Chapter 3**, the focus is on the coalescence phenomenon in surface nanobubbles. Two closely spaced surface nanobubbles were considered for the coalescence and the changes in the region between the nanobubbles' adjacent interfaces were imaged. The results reveal that the surface nanobubbles do not merge via direct contact of their contact line or interface, rather, merging is initiated by gradual localized changes in the physical properties of the region between the adjoining nanobubbles' interfaces. More precisely, the merging of the stable nanobubbles is initiated by the deposition of gas molecules and the formation of a thin gas layer. In addition, the merging of multiple surface nanobubbles and gas layer formation dynamics is also discussed. Further, the kinetics of deposition and formation of the thin film is found to be majorly governed by the degree of gas oversaturation in the liquid.

In **Chapter 4**, as a demonstration of surface nanobubbles' functionality, a novel methodology to fabricate soft line features from surface nanobubbles is illustrated. The highly focused electron beam from the TEM is used to generate highly localized high gas oversaturation in the liquid film whereas the TEM's trackball is used to facilitate sample-electron beam relative motion. This methodology nucleates the surface nanobubbles along the path traversed by electron beam in high number density. Features made of nanobubbles using this method are less than 300 nm in width, which are proposed to have applications in bubble-assisted nanofabrication techniques. Further, the study also reveals the stability of nanobubble patterns, both temporally and in flow.

In **Chapter 5**, the major results and conclusions of the thesis are put forward. The future scope and recommendations are also presented in this chapter.

Overall, the studies included in this thesis will potentially pave the way for a better understanding of nanobubbles and gas transport phenomenon at nanoscale.