Simulation of Ozone Nozzle for Oxide Molecular-Beam Epitaxy

Joseph Dill
University of Minnesota, Twin Cities

ABSTRACT:

The Platform for the Accelerated Realization, Analysis, and Discovery of Interface Materials (PARADIM) operates a molecular-beam epitaxy system for the growth of oxide thin films. The nozzle that supplies ozone gas in the existing system emits a beam focused at the center of the film plane, inadequately oxidizing the material at its edges. In this study, we develop a computer simulation of gas emission from a nozzle in a high-vacuum environment and use it to find a nozzle design that generates a more even distribution of ozone across a 3-inch diameter substrate.

INTRODUCTION

Molecular-beam epitaxy (MBE) is a thin film growth method in which separate molecular beams of reactants are deposited on a substrate surface in an ultra-high vacuum environment, providing atomic-level control of the film contents. The growth of an oxide such as $YBa_2Cu_3O_7$ by MBE involves separate molecular beams of Y, Ba, Cu, and ozone. Molecular beams of the metals (Y, Ba, and Cu) are created by heating crucibles containing each element. The ozone gas, however, is emitted towards the substrate surface from a nozzle. The activity of ozone is roughly a billion times that of molecular oxygen [1], which allows it to form $YBa_2Cu_3O_7$ without destroying the long mean-free-path needed for MBE. Thus, ozone is commonly used as the oxidant in oxide MBE.

At PARADIM, thin films can be grown by MBE on one of two substrate geometries: 1) 1x1 cm square or 2) 3-inch diameter circle. The existing nozzle emits a beam of ozone from a single long pipe aimed at the center of the substrate. This design does not provide enough ozone at the outer edge of the 3-inch diameter substrate to fully oxidize the material being grown.

This study's objective is to develop a computer simulation of gas emission from a pipe in high-vacuum and design a nozzle that emits an even flux across a 3-inch wafer. This work builds on previous simulations by Kraisinger et al. [2], who also designed an ozone nozzle for an oxide MBE system.

METHODS

In a high-vacuum environment, the angular distribution of gas emitted from a circular pipe orifice is a cosine distribution [3] that becomes more focused as the pipe's length-to-diameter ratio increases [4]. The equations used to calculate this collimating effect are given in [5]. Mathematica was used to simulate the flux pattern generated by inputted nozzle specifications. The simulation discretizes the pipe orifice into a grid of smaller pipe elements. Each element acts as a flow area emitting the collimated distribution, as is done in integral form in [6]. The substrate plane is also discretized. The fluxes from all nozzle elements are then summed to compute the total flux at each substrate position.

The substrate platform rotates during growth, making approximately one revolution in the time required to deposit an atomic monolayer. This rotation is simulated by interpolating the flux pattern, and then numerically integrating around a circle concentric with the substrate to compute the average flux through all substrate positions at a given radius from the center of the substrate.

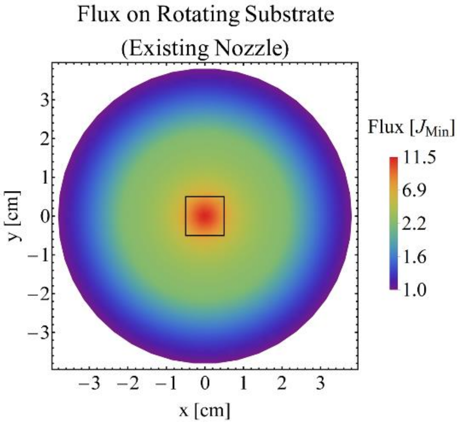

Figure 1: Flux pattern generated by the existing nozzle. The minimum flux in the 1x1 cm substrate region, seen in the middle, is six times that at the outer edge of the 3 in substrate. Flux is plotted in units of the minimum flux, $J_{min.}$

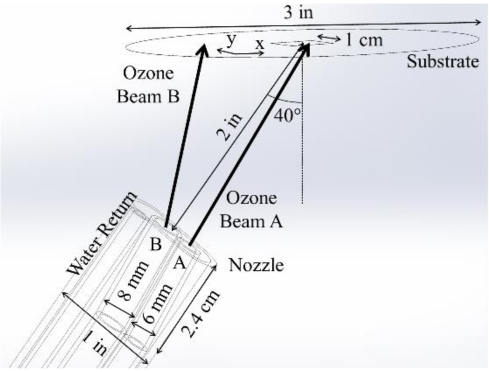

Figure 2: 3D Schematic of the new nozzle.

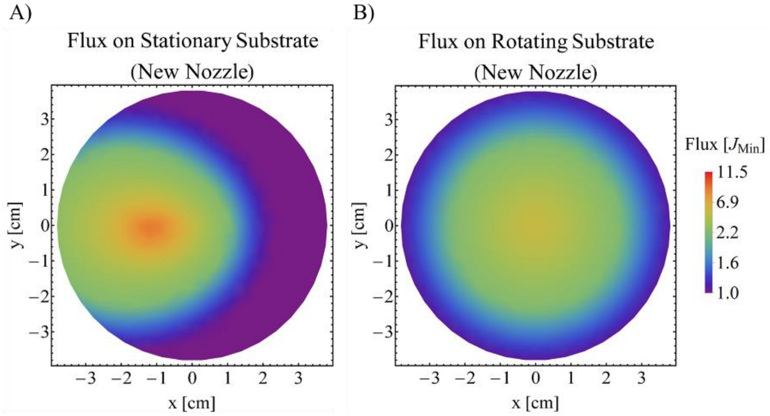

Figure 3: The flux pattern created by the new nozzle design A) before and B) after rotation-averaging.

The referenced emission equations are only valid for gas in molecular flow. Under typical growth conditions, ozone enters the nozzle in intermediate flow but exits in molecular flow. To determine the length of the portion of the nozzle in which the gas is in molecular flow, we treat the nozzle as two pipes in series where gas is in intermediate flow in the first pipe and molecular flow in the second. Equations for the conductance of pipes in each regime are provided in [7]. By equating the throughput of both pipes, one can solve for the length of the molecular regime pipe.

RESULTS AND CONCLUSIONS

Figure 1 shows the simulated flux pattern generated by the existing nozzle. The highest flux, located at the center of the 1x1 cm substrate, is eleven times that at the edge of the 3-inch diameter substrate.

New nozzle designs were constrained to the size and position of the existing nozzle to avoid interference with other MBE system components. Additionally, the ozone must be water-cooled until it is emitted. Thus, all nozzles were water-tight and included a water return pipe. The available degrees-of-freedom were the orifice diameter and the location and exit angle of the pipe with respect to the nozzle face.

The best-performing design, drawn in Figure 2, consisted of two pipes, labeled A and B. Both pipes are bent, which introduces an additional source of conductance [8]. Pipe A is aimed at the center of the growth surface and operates alone when growing on a 1x1 cm substrate. Pipe B is aimed as far off-center as possible within the confines of the nozzle walls and the other pipes. When growing on a 3-inch substrate, both pipes are open. Roughly 85% of the nozzle throughput comes from pipe B because it has a larger diameter, and thus a smaller flow resistance.

Figure 3 shows the flux pattern generated by the new nozzle design. The sizeable off-center flux created by pipe B is distributed across the substrate by rotation. The center of the 3-inch substrate receives only five times the flux as the edge, compared to eleven with the existing nozzle. Pipe A also matches performance with the existing nozzle on the 1x1 cm substrate.

FUTURE WORK

The results of this simulation will inform the construction of a new ozone nozzle. Other nozzle designs utilizing non-circular orifices may be analyzed with a Monte-Carlo simulation before constructing the design proposed in this report.

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