

On the Early History of ALD: Molecular Layering

J. Aarik,¹ A.R. Akbashev,² M. Bechelany,³ J.S. Becker,⁴ M. Berdova,⁵ D. Cameron,⁶ V. E. Drozd,⁷ C. Dubourdieu,⁸ J. Elam,⁹ S. Elliott,¹⁰ G. Gottardi,¹¹ K. Grigoras,¹² J. M. Kanervo,⁵ Yu. Koshtyal,¹³ M.-L. Kääriäinen,¹⁴ T. Kääriäinen,¹⁴ L. Lamagna,¹⁵ A. Malkov,¹⁶ A. Malygin,¹⁶ J. Molarius,¹² J. Nikkola,¹² C. Ozgit-Akgun,¹⁷ H. Pedersen,¹⁸ R.L. Puurunen,¹² A. Pyymäki Perros,⁵ R.H.A. Ras,⁵ F. Roozeboom,¹⁹ T. Sajavaara,²⁰ H. Savin,⁵ T.E. Seidel,²¹ P. Sundberg,⁵ J. Sundqvist,²² M. Tallarida,²³ J.R. van Ommen,²⁴ C. Wiemer,²⁵ O.M.E. Ylivaara¹²

¹ University of Tartu, Institute of Physics, Estonia; ² Drexel University, Philadelphia, Pennsylvania, USA; ³ CNRS University of Montpellier, Montpellier, France; ⁴ Cambridge, Massachusetts, USA; ⁵ Aalto University, Espoo, Finland; ⁶ Miktech Oy, Mikkeli, Finland; ⁷ St. Petersburg State University, St. Petersburg, Russia; ⁸ Institut des Nanotechnologies de Lyon & Joint Laboratory InCVD, Lyon, France; ⁹ Argonne National Laboratory, Illinois, USA; ¹⁰ Tyndall National Institute, University College Cork, Lee Maltings, Cork, Ireland; ¹¹ Fondazione Bruno Kessler, Center for Materials and Microsystems, Trento, Italy; ¹² VTT Technical Research Centre of Finland, Espoo, Finland; ¹³ Ioffe Physical Technical Institute, St. Petersburg, Russia; ¹⁴ University of Colorado Boulder, Colorado, USA; ¹⁵ STMicroelectronics, Agrate Brianza, Italy; ¹⁶ St. Petersburg State Technological Institute (Technical University), St. Petersburg, Russia; ¹⁷ Bilkent University, Ankara, Turkey; ¹⁸ Linköping University, Linköping, Sweden; ¹⁹ Eindhoven University of Technology, Eindhoven, and TNO, Eindhoven, The Netherlands; ²⁰ University of Jyväskylä, Jyväskylä, Finland; ²¹ Seitek50, Palm Coast, Florida, USA; ²² Fraunhofer IPMS-CNT, Dresden, Germany; ²³ Brandenburg University of Technology Cottbus-Senftenberg, Cottbus, Germany; ²⁴ Delft University of Technology, Delft, the Netherlands; ²⁵ Laboratorio MDM, IMM-CNR, Agrate Brianza, Italy
riikka.puurunen@vtt.fi

Atomic layer deposition (ALD) is a technique that has been instrumental in enabling the semiconductor industry to maintain its adherence to Moore's Law, and is becoming a game-changer in several other fields. A worldwide open collaborative initiative called the "Virtual Project on the History of ALD" (VPHA) was launched in summer 2013 to explore how the ALD concept was developed; which were the first ALD experiments; when, where, why and by whom they were made. ALD was invented independently under the name atomic layer epitaxy in Finland in 1974. In this presentation, we expose another major branch of ALD development made under the name molecular layering (ML, молекулярное наслаивание), which has remained poorly known outside the USSR/Russia. For example, the first ML (ALD) investigations were carried out in the 1960s on silica particles, using metal halides (e.g., TiCl_4) and water as reactants; the first Soviet ALD patents ("author's certificates") date from 1972 and are related to catalysis; growth of thin films of TiO_2 and SiO_2 on planar semiconductor substrates such as Si and Ge were published in the 1970s; ALD films were grown on carbon in 1974 and on polymers in 1984; in situ gravimetry was used to follow ALD in 1982; and amine-catalyzed SiO_2 was deposited in 1982. The information presented will help today's growing global ALD community to see their research in perspective.

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