

Laurence D Marks

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Professor Laurence Marks was born in Barnet, London in 1954, and after attending Trinity School of John Whitgift in Croydon was awarded a scholarship to Kings College Cambridge in 1973. After completing a BS in Natural Sciences (Chemistry) he went on to do a PhD in the Department of Physics, Cavendish Laboratory with Professor A. Howie, followed by postdoctoral work with Professor A. Howie and Dr. D. J. Smith in Cambridge, moving to Arizona State University in 1983 to work with Professor J. M. Cowley. He joined the Department of Materials Science and Engineering at Northwestern University in March 1985, being promoted to Associate Professor in June 1986 and full Professor in June 1992.

He has a long history of pioneering new methods, ranging from technical approaches to imaging materials to new numerical algorithms for solving surface structures as well as algorithms for density functional fixed-point methods. While the largest number of his publications have involved transmission electron microscopy, both experimental and theoretical, he has also worked extensively in other areas. Most often cited of these is his work in nanoparticles, where a class of particles he discovered have become known as the “Marks Decahedron”. Other areas include combining experimental and theoretical approaches to understand surface structures for which he was awarded the 2017 ICSOS Surface Structure Prize, as well as many other contributions to understanding structures for which he received the 2016 Warren Award of the American Crystallography Association. He has also worked extensively in nanoscale tribology, including pioneering ways to image samples during sliding and dislocation based models of sliding and wear. More recently he has ventured into aqueous corrosion and high temperature oxidation, initially for hip implants but more recently the nonequilibrium development of protective oxide films at the nanoscale.

Significant Science, in approximately chronological order

1. Discovered what has now become known as the “Marks Decahedron”. This is the equilibrium shape of five-fold multiply twinned particles (1-3), and is one of the three stable structures of fcc materials at the nanoscale, the other two being icosahedra and single crystals. The term has now become a generic, and is used without citation.
2. The first atomic resolution imaging of nanoparticles in 1981 (4-6), joint with D. J. Smith. This work was the first detailed study of the structure of nanoparticles using HREM, and showed details such as internal structure and dislocations. The early work on experimental imaging was reviewed in 1994 (7), a widely cited paper.
3. The first profile imaging of surfaces in the early 1980’s (8, 9), joint with D. J. Smith, as well as the conditions needed (10-14) for direct interpretation of images of surfaces, taking into account both channeling and localization. This method was called “Profile Imaging”, and is

now widely used in the study of heterogeneous catalysis and nanoparticle research, the term having become a generic frequently used without citation.

4. The first experimental observation of surface plasmons in a STEM in 1982 (15), what has now become known as Aloof Scattering. These experiments showed that there were energy losses when the electron beam was outside of MgO cubes, and demonstrated that they could be explained in terms of the appropriate dielectric loss function.
5. Demonstration that structural fluctuations in small particles observed in the electron microscopes was not simply a consequence of the electron beam, but rather was due to the fact that the energy barriers between the different structures was relatively small (16-22). This work also was the first to introduce the concept of a phase map for small particles as a function of size and temperature (17, 20), a concept that still appears to be valid.
6. Developed the first fully UHV-HREM transmission electron microscope with 0.2nm resolution joint with Hitachi (23, 24), later extended with a multichamber surface science system (25). While there had previously been UHV electron microscopes constructed using cryocooling of the sample they were not fully UHV and rather limited. There had also been some other attempts to build UHV-HREM instruments, but they were not successful. To date it is the only instrument that has demonstrated room-temperature operation with sensitive surfaces such as the Si (001) 2x1 surface.
7. A body of work of various aspects of plan-view imaging and diffraction of surfaces as well as experimental studies of semiconductor surfaces both when clean and with metals on them (23, 24, 26-49).
8. A body of work on electron radiation damage phenomena at the surface of oxides (50-62). This work includes both experimental work on the atomistic structure of the surface and seldge regions, as well as work understanding the symmetry conditions for the phase transitions and kinetic modelling of the moving boundary problem.
9. A body of work on local structure in high-temperature superconductors as well as the growth and properties of Josephson junctions (63-88). Almost all of this work was highly collaborative, with groups from other fields as well as other Universities.
10. Work on in-situ growth in the UHV-HREM system with its coupled multichamber science chamber (48, 89-104). In addition to some work for metals on semiconductor surfaces, this involved two projects. The first was to grow cubic boron nitride (BN), the second quasicrystalline thin films. The work on BN produced atomic sheets of hexagaonal BN where it was demonstrated that the local bonding only involved even numbers of rings, work which has been well cited.
11. Pioneered the use of directly methods with electrons or x-rays for surfaces to solve unknown structures (92, 105-112). While others had tried to use conventional direct methods for surfaces before, they were not successful. This is because these methods could not handle

cases where very strong beams are not measurable because they overlap with bulk diffraction spots that are many orders of magnitude stronger. He developed methods that could handle this.

12. Exploiting the direct methods, solution of a number of complicated metal-semiconductor surface structures that could not be solved by other approaches (92, 94, 113-116). These papers are moderately well cited.
13. Work on the connection between mathematical methods such as non-convex sets and crystallographic phase problems (117), as well as the connection between dynamical diffraction and crystallographic phase invariants and the conditions when direct methods will work with electrons (117-128). This work laid the foundation for direct methods with Precession Electron Diffraction that were developed later.
14. The first precession electron diffraction (PED) camera (student Chris Own) in the US (122), as well as a large body of work including the first dynamical analysis of PED (123), and many other contributions to the theory behind and experimental application of the method (117, 119-121, 124-132).
15. Pioneering work on the surface structure of strontium titanate. He determined the structure of what has become the standard model for the titanium-rich surface, a double layer (133). Following on from the original work there is a relatively large body of work on other structures in the system as well as other oxides analyzed by a range of methods including electron diffraction, profile imaging, scanning probe microscopy and density functional theory calculations (134-162), which has recently been reviewed (163). This work is well cited.
16. Pioneering work to develop a fast method of determining the Local Surface Plasmon Resonances (LSPR) of individual nanoparticles and image exactly the same nanoparticle by electron microscopy (164-166). The methods developed are now widely used around the world for correlated measurements. They have been exploited to obtain statistically significant measurements of the size, shape and LSPR of thousands of nanoparticles to determine accurate relationships between them (167-173), work frequently cited by others.
17. Pioneered the use of in-situ measurements of tribological processes (174-181). This work involved sliding a scanning probe (STM or AFM) along a surface inside a transmission electron microscope and directly imaging processes taking place at the surface. The work has been relatively widely cited in the (smaller) nanoscale tribology community with frequent invited presentations. Several other groups around the world have adopted the method with varying degrees of success.
18. Complementing the in-situ tribological measurements, the development of material science based dislocation models to understand nanoscale tribology processes (182-186). This includes modelling friction via the motion of interfacial dislocations and Bowden-Tabor ploughing as creep.

19. Pioneered the use of spatially resolved nanoindentation of cement to understand the properties (187-189), work which is well cited in the smaller field of scientists active in cement research.
20. Developed a robust code for solving the self-consistent (fixed-point) problem in density functional theory (190), and a more recent form which simultaneously solves the fixed-point problem for the density and atomic positions (191). The robust code involved analysis of the best approach using established Broyden methods, the second went beyond the established mathematical literature. Both these algorithms are the recommended ones used in the Wien2k code by about two thousand groups around the world. He is now officially one of the authors of the Wien2k code (192).
21. Identification of a graphitic layer *in-vivo* in metal-on-metal hip implants (193), as well as a body of work on hard phases in these materials as well as corrosion of them (194-203). There remain serious issues with hip and other implant materials, but the current political and legal is currently preventing further research to develop better materials. This work laid the foundations for a larger effort on corrosion, funded by ONR as a larger Multi-Investigator University Research Initiative, Corrosion in 4D: <http://MURI4D.numis.northwestern.edu>.
22. Returning relatively recently to the topic of nanoparticles, a body of work combining analysis of nanoparticle structure (both metals and oxides) with the thermodynamics and kinetics of growth and their catalytic performance (154-156, 158, 160, 204-228). Highlights of this work include a new model for the Wulff shape of nanoalloys showing size-dependent segregation and shape effects and the first model to correctly predict the growth shape of multiply twinned particles (as against the thermodynamic shape).
23. Moving in to a new area, aqueous corrosion and high temperature oxidation a number of recent publications have investigated both experimental structures at the nanoscale (229-231) as well as density functional models (232, 233). A recent significant paper describes the formation of Nonequilibrium Solute Capture in oxides, a new way of understanding protective oxide films which is disruptive new science (234).
24. Last, but certainly not least, by combining expertise in a new research area, flexoelectricity (235-237) with prior expertise in tribology, combined the two to explain triboelectricity (238).

Publications can be found at www.numis.northwestern.edu/Research/Articles/articles.shtml

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