Gert Ehrlich

1926–2012

BIOGRAPHICAL *Memoirs*

A Biographical Memoir by
Grażyna Antczak
and Robert S. Chambers

NATIONAL ACADEMY OF SCIENCES

GERT **EHRLICH**

June 22, 1926–August 8, 2012

Elected to the NAS, 1986

By Grażyna Antczak and Robert S. Chambers

Gert Ehrlich was noted for his ingenious and highly definitive studies of atoms and molecules interacting with clean and structurally well-characterized metal surfaces. His research, including the work he inspired in a small group of students at the University of Illinois, has contributed significantly to the foundations of nanoscience and nanotechnology. He was an extraordinarily careful and thorough scientist and an excellent teacher. Ever modest, he was quick to give generous credit to others.

The early years

Gert was born and raised in Vienna, Austria, during the tumultuous years prior to the country's annexation by Germany in 1938. Although his mother Paula Maria (Kucera) was Catholic, his father Leopold was Jewish. Consequently, he and his father were detained for questioning by the police in early 1939; luckily, they were released. Responding to this "wake-up call," Leopold departed immediately on an existing work visa to the United States and emergency children's visas were obtained for Gert and his sister Dorothy. Because Gert's mother was unable to also procure the necessary documents for herself, the pre-teenagers were sent unescorted to the United States. Although the actual ocean crossing was something of a grand adventure for Gert, the forlorn siblings arrived in New York on the T. S. S. Veendam in May 1939 and were reunited with their father. Paula Maria managed to immigrate several months later.

In 1944 Gert enrolled at Columbia University and—although interrupted by two years of military service in the U.S. Army (he served in Austria after the end of the war)—received an A.B. degree with honors in chemistry in 1948. Gert continued his education at Harvard University, earning an A.M. in 1950 and a Ph.D. in 1952 in physical chemistry. His research, culminating in a thesis titled "Studies on Synthetic Polyampholytes,"

involved the synthesis of polyampholyte and the characterization of its solution properties by light scattering. His advisor was Professor Paul Doty.

In 1951–52, Gert did postdoctoral study at Harvard as a National Institutes of Health Fellow. He expanded his doctoral research to more biologically oriented studies by examining nucleoproteins. The following year he worked with Gordon Sutherland at the University of Michigan's physics department, where Gert carried out studies on the infrared spectroscopy of macromolecules.

General Electric, 1953–1968

In 1953, Gert accepted a job as an industry-based scientist. He later wrote:

> *Although strongly inclined toward academic pursuits, I received a tempting offer to undertake studies of solid surfaces at the General Electric Research Laboratory in Schenectady, NY. This proved [to be] a marvelous opportunity. Immersed in a truly stimulating environment, I was able to devote myself wholly to becoming acquainted with an area until then unknown to me, and to develop a research program aimed at a better understanding of the kinetics of gas-solid interactions.*

For the next several years, Gert developed the equipment and techniques necessary for quantitative work on solid surfaces. He built various contraptions—mostly glass systems with triple-stage mercury-diffusion pumps—to achieve the highest possible vacuums of the day; and he used liquid hydrogen or helium cryogenics for low-temperature studies. His temperature-programmed desorption, or flash-desorption, studies demonstrated the importance of distinct states of binding in chemisorption phenomena.

Although he carried out this work on well-characterized crystal surfaces, the macroscopic nature of the results were insufficient for Gert; he became curious about the underlying atomic details of chemisorption. Taking advantage of the field ion microscope recently invented by Erwin Müller, Gert was able for the first time to observe atomic motion on precisely characterized atomic surfaces. With Frank Hudda he made the first quantitative observations of individual atoms diffusing on a metal surface. The corresponding paper, "Atomic View of Surface Self-Diffusion: Tungsten on Tungsten" [1], has since been cited over 1,000 times. This article showed real-space images of individual atoms moving on a surface almost 30 years before such studies became common in nanotechnology.

During his service at General Electric, Gert met and married Anne V. Alger, who also worked at the GE research facility.

University of Illinois, 1968–2012

In 1968, after 15 years of research in Schenectady, Gert received an offer that would allow him to continue his studies in an academic environment. He accepted the offer—a professorship in the Department of Metallurgical Engineering, subsequently renamed the Department of Materials Science, at the University of Illinois at Urbana-Champaign—and established himself as a superb teacher as well. He developed popular courses in thermodynamics, statistical mechanics, and surface science.

Gert wryly commented that he and his research colleagues "formulated a satisfying and fairly simple picture of how atoms behave on metal surfaces when we made an important mistake: we did one too many experiments."

For his continuing research efforts, Gert assembled a small number of students who learned and often enhanced his exacting methods. In his early work at Illinois, he emphasized the scattering of molecular beams from surfaces [2], and his group was among the first to recognize the phenomenon of rotationally inelastic diffraction [3]. His research then proceeded in two areas: identification of molecular processes in activated chemisorption [4]; and examination of the atomic events in layer growth [5], using the field ion microscope (FIM).

What follows in this section is a description of the second—in Gert's words, "particularly exciting"—area.

Regarding the early years of layer-growth studies, Gert wryly commented that he and his research colleagues "formulated a satisfying and fairly simple picture of how atoms behave on metal surfaces when we made an important mistake: we did one too many experiments." Thereafter, the preconceived simple notions went out the window, to be followed by over 43 years of careful systematic research. The fruits of this work are over 200 coauthored publications that describe a rather surprising array of surface phenomena. For Gert, the complexity of atomic-surface phenomena became his "puzzle box," and he spent much of his time and energy working on its various aspects. A "short list" of the phenomena to which he directed so much effort is given below. (It is important to note that although we are presenting only the results of Gert's group, several other surface-science groups around the world made equally important contributions, during roughly the same period, to the understanding of surface diffusion.)

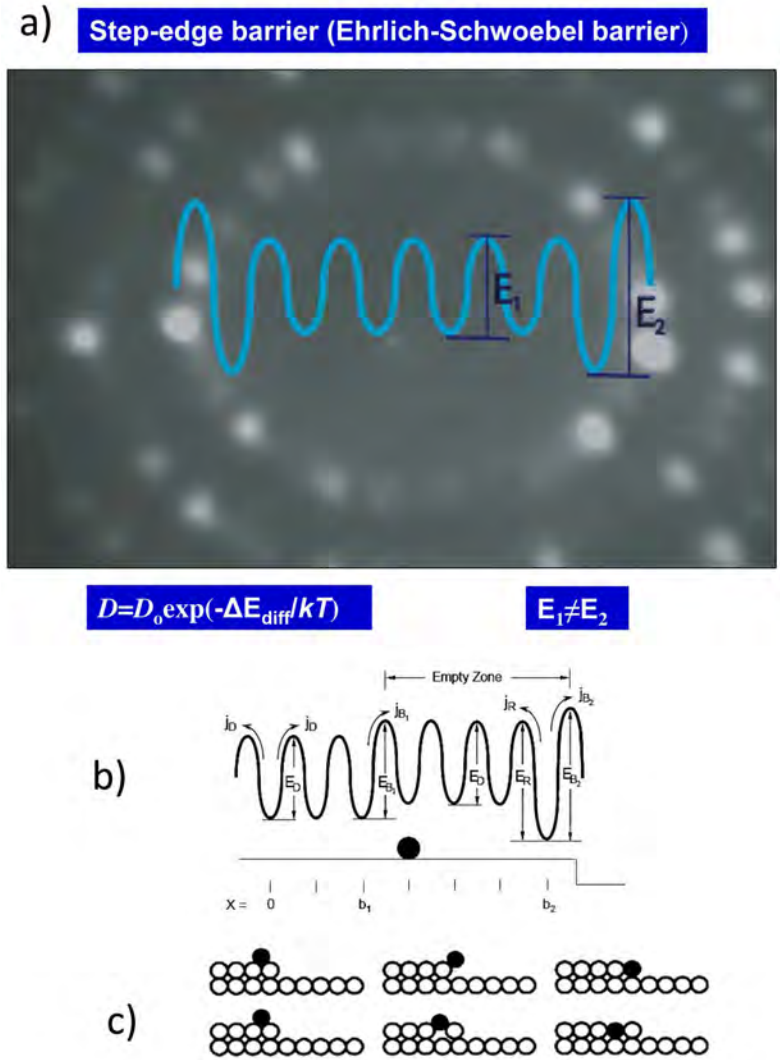

Figure 1.
a) The shape of potential observed on the W(110) surface;
b) the interior barrier observed for the Ir(111) and Pt(111) surfaces with FIM; [11];
c) if an adatom escapes a plane it can do so in two different ways—by hopping over the edge or by exchanging with an edge atom of the plane.[12]

In the early models of surface diffusion proposed by Gert's group (and others), the atomic surface plays a more-or-less static role, as in an exercise obstacle course, in which the various barriers and obstructions are fixed. In this simple model the atomic surface provides barriers in one dimension for some (channeled) planes and in two dimensions for others. The task is first to characterize the atomic structure of a perfectly clean substrate and then to deposit an adatom. If the temperature is raised from cryogenic values and is controlled precisely enough—and if enough measurements are taken—an Arrhenius plot will yield the activation energy (and prefactor) for a specific adatom on a specific surface. Of course, each type of surface plane of each element, and with a varied choice of adatom, yields a huge number of experiments to be performed.

In Gert's lab, numerous examples were published that reasonably confirmed this general model [6].

Although the atomic arrangement of some surface planes is easily understood, others are puzzling and must be characterized in atomic detail before meaningful diffusion studies are possible. For example, some planes—including Ir(111), Pt(111) and Re(001)—display two different adsorption sites, fcc and hcp, and an adatom therefore encounters a more complex diffusion barrier [7–9].

But surface complexity is only a small part of the overall problem. The behavior of the adatom at the edge of a plane—or at the end of a row on one-dimensional diffusion surfaces, such as the bcc(211) plane—requires much more scrutiny. Does the adatom fall off the edge to be incorporated with the substrate at a kink site, as predicted by the simple model? Surprisingly, on some planes there is instead a reflective barrier, which is now called the Ehrlich-Schwoebel barrier [Figure 1a] [1, 10]. Equally surprising, sometimes this barrier is only at the edge while sometimes the barrier extends inward from the edge, yielding a so-called empty zone [Figure 1b] [11].

An additional complication occurs if an adatom escapes the plane: it may simply jump over the edge to become part of the plane; or it may exchange with an edge atom, as shown in Figure 1c [12]. Such ambiguity in the incorporation of an adatom with the lattice may be significant to our understanding of nucleation and crystal-growth processes and may be useful in the manufacture of various nanomaterials.

As more precision is established, a number of curious discrepancies begin to appear. One of them is the existence of long jumps at higher diffusion temperatures. It appears that the adatom does not always equilibrate with the surface after an individual jump

and continues diffusing to another site. The occurrences of long jumps increase with increasing temperature and can be the dominant mechanism for high diffusion temperatures. Vital for such an investigation to have any validity is to develop a supporting statistical theory to distinguish data pertaining to rapid single jumps from data related to long jumps. The often-cited "lattice walks by long jumps" [13] combines Monte Carlo simulations with data analyses to distinguish the various diffusion possibilities. This procedure has also been utilized by other researchers [14]. The Ehrlich group has reported proof for long jumps in diffusion of palladium and tungsten on a one-dimensional W(211) surface [15], a two-dimensional long-jump diffusion of palladium and tungsten on W(110) [16], and the temperature dependence of long jumps and its correlation with single jumps [17]. Long-jump phenomena are yet another key to our eventual understanding and possible improved atomic control of crystal growth.

What happens if two or more adatoms are simultaneously deposited on a surface? Do the adatoms ignore each other and diffuse independently of one another or, if they interact, in what way do they interact, and how are diffusion and crystal growth affected? The Ehrlich group has reported several studies on clustering and cluster diffusion—for example, interactions of two identical atoms as well as two atoms of different chemical identity [18]; and three-body interactions [19]. In addition to multiple-adatom studies, the group has also investigated adatom/vacancy, adatom/step, and adatom/dislocation interactions [20]. In another study, the interaction of an adatom with a "nanowire" (a chain of atoms) was explored [21]. Supporting such diffusion studies, detailed mechanisms of small-cluster motion [22] and larger-cluster motion [23] have been reported. Also, molecular dynamic investigations have been made of "gliding phenomena of magic clusters"[24]. The molecular dynamic analysis, combined with FIM investigation, has also been used to explore the energy exchange in the collision of atoms from the gas phase with the surface, and its influence on the transient motion of an adatom [25]. More recently, the group has utilized a scanning tunneling microscope (or STM) to investigate surface self-diffusion of gold adatoms and the nucleation and growth of gold clusters on Au(100) [26].

Often the surface is not sufficiently static to ignore, and unexpected diffusion events occur. What if the surface atoms, in addition to the adatom, are moving causing shifts in the barriers to diffusion? A perhaps more realistic view of atomic diffusion is to consider the combination of random temperature-dependent changes in the adatom energy with the random variations of the barriers it encounters. (This brings to mind a cartoon in which our hapless adatom encounters a wild variety of randomly moving barriers in

an obstacle course.) In the extreme case, there is the possibility that the adatom will exchange with a substrate atom. The motion of the substrate atoms may be sufficient at a selected diffusion temperature for the adatom to steal the position of one of the substrate atoms; the substrate atom no longer has a spot in the substrate and therefore becomes an adatom.

This is unequivocally demonstrated in an atom-probe FIM experiment [27] in which a tungsten adatom is deposited onto an iridium substrate. The diffusion process proceeds as expected for several intervals, but then the kinetics (and the adatom image) mysteriously change significantly. It "seems" that the tungsten atom has been incorporated into the surface layer and that the adatom is no longer tungsten, but chemical identification is critical to such a conclusion. The atom-probe experiment first identifies using time-of-flight mass spectrometry that the adatom has changed from tungsten to iridium. Then the surface layer is peeled off atom-by atom until the tungsten atom is detected (where expected) within the top iridium layer. The concept and data are shown in Figure 2. If a fourth-ball model were added to the figure, showing the surface after the displaced atom has been field evaporated, a possible single-atom-doping technique in the manufacture of nanomaterials would be demonstrated. This mechanism has recently been used for incorporating a phosphorus atom into a silicon matrix to create a single-atom transistor [28].

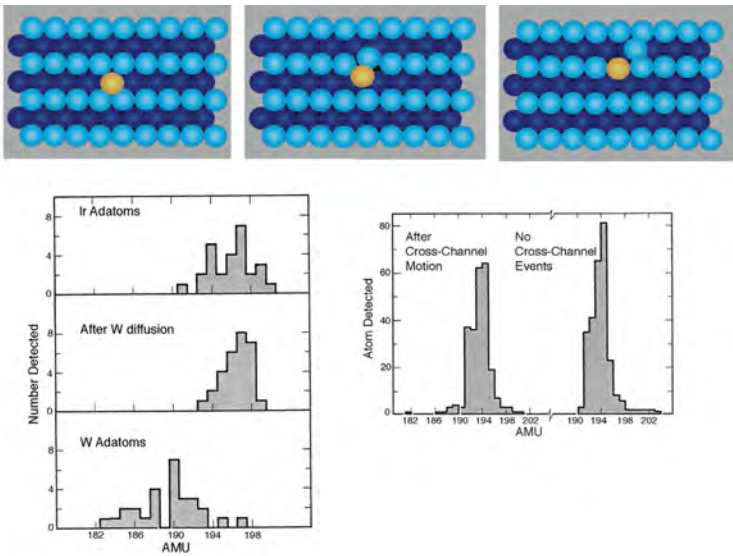

Figure 2. Atom probe investigation of exchange mechanism of W on Ir(110).[27]

In summary, the efforts over almost six decades of Gert and his research group to understand surface diffusion, in combination with similar efforts from around the world, have

evolved from what first appeared to be a hopelessly complicated collection of surface phenomena into reasonably understandable surface behavior that has significant technological value. His last effort with his last graduate student was a project, still in progress, that combines many of the principles described here: diffusion by a combination of jump and exchange mechanisms and the creation of single vacancies on reconstructed W(100) [29].

An accomplished but modest man

Gert received numerous honors during his career. Among them were the Medard W. Welch Award from the American Vacuum Society in 1979 "for contributions to our understanding of the microscopic-force laws by which atoms residing on solid surfaces interact with the substrate and each other;" and the Kendall Award in colloid or surface chemistry from the American Chemical Society in 1982 "for his fundamental work on atomic and molecular events on solid surfaces, including diffusion of individual atoms, bonding and clustering of adsorbed atoms, and molecular disassociation on metal surfaces." He was a Guggenheim Fellow in Harvard's Division of Applied Sciences in 1984–85 and was elected to the National Academy of Sciences in 1986. Gert was a University of Illinois Scholar, 1987–90, and in 1992 he received a research award from the German Alexander von Humboldt Foundation that he spent at the Fritz-Haber-Institute in Berlin. There he worked on boundary-layer and interfacial chemistry with Gerhard Ertl and Jochen H. Block. Gert was a fellow of the American Physical Society and of the New York Academy of Sciences; a member of the American Chemical Society, the American Vacuum Society, and the Faraday Society; and was actively involved in affairs of the Field Emission Symposium, Physical Electronics Conference, and International Workshop on Surface Physics.

Figure 3. A group photo taken during the Gert Ehrlich Symposium on Surface Science, which celebrated Gert Ehrlich's 80th birthday.

Because of Gert's intense modesty, it was difficult for members of his group to draw him out for celebrations. Except for the occasional prelim-exam party to mark the achievements of someone else in the group, the only "celebration" that he dearly cherished was the weekly donut hour at 11:00 a.m. on Fridays (absolutely every Friday!), established by him in 1968. This 45-year tradition continues even to this day in his honor, although—lacking his leadership—weeks are often skipped.

Gert worked tirelessly and continuously during his 44 years at Illinois. He arrived every morning well before 8:00 a.m.—except for Sunday, when he "slacked off" until 9:00 a.m. Occasionally he would attend or listen to a classical opera on a weekend afternoon. Thereafter, for the next week or so, the other occupants of the lab would be (pleasantly) subjected to a light whistling of his favorite arias from the opera. Except for working sabbaticals, he took only two short vacations while living in Illinois. The image of a "workaholic" might come to mind, but that would not be a fair assessment; Gert was a person whose play and leisure were synonymous with his teaching and research. He treated most of his group members as a family and was always available for assistance. When asked a research question, however, he would usually respond only with hints, or perhaps with a leading question of his own, to challenge the asker.

In 2006, after many failed attempts to celebrate earlier milestones, Gert finally acquiesced to having a Gert Ehrlich Symposium on Surface Science in honor of his 80th birthday. Although he may first have been somewhat apprehensive, he was visibly moved that so many members of his research group, spanning several decades, managed to attend and give presentations, some of which had nothing to do with surface science [Figure 3]! However, most have applied Gert's rigorous rules of careful experimentation to their own fields and are the better for it.

In 2007, Gert realized that perhaps it would be a good idea if he wrote a book on surface diffusion. He sent a tentative letter to an editor at Cambridge University Press, asking if there might possibly be any interest in such a book. The editor chose not to respond immediately but instead to send Gert's letter out for review! The overwhelming response from the reviewers was a definite "yes" and that Gert would be the best author for such a book. Written with coauthor G. Antczak, it was published in 2010 as the definitive work on *Surface Diffusion: Metals, Metal Atoms, and Clusters* [30].

Although Gert was not widely known beyond the field of surface science, he was indeed famous and highly respected within that field. When a recent postdoctoral fellow was

asked why he chose to venture out to the unglamorous flatlands of Illinois, he simply smiled and said only, "THE Gert, THE Ehrlich."

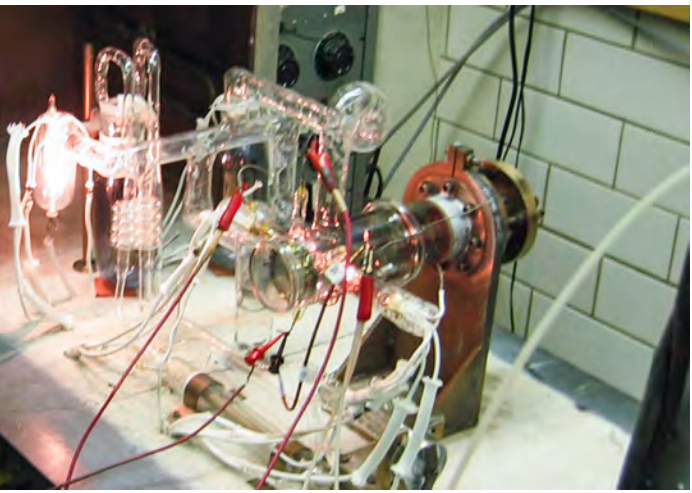

Figure 4. A typical Ehrlich field ion microscope.

Not surprisingly, Gert was secretive about his final illness; only his wife knew that he had leukemia until—literally—he was unable to sit in his office chair (about five weeks before he died). His laboratory has recently been shut down and the last of his working field ion microscopes [Figure 4] has been donated to the Deutsche Museum in Munich. He is survived by his wife and lifelong partner, Anne.

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