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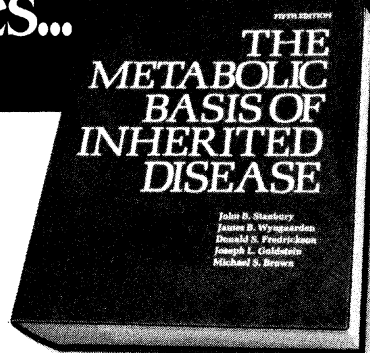
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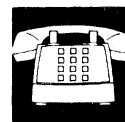
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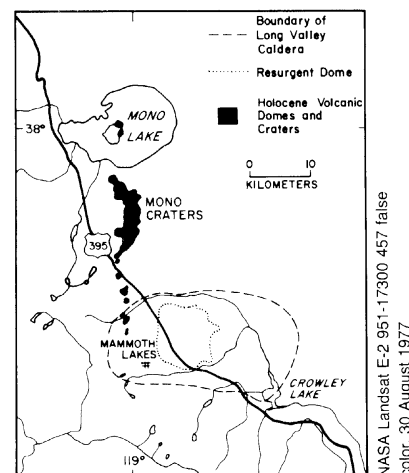
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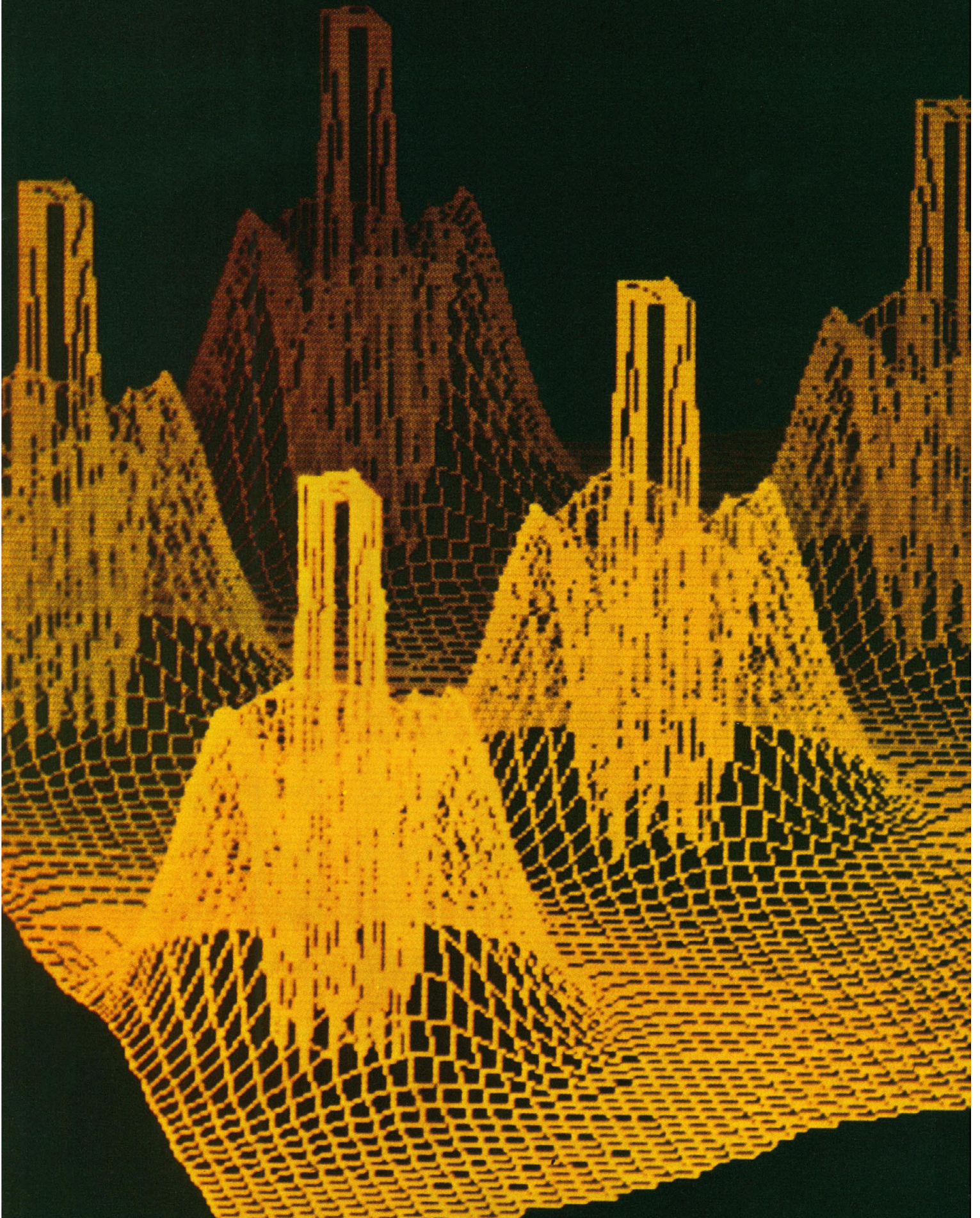
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The Surface Contingent



The Surface Contingent

A recent finding at the General Motors Research Laboratories has changed scientific thinking about the behavior of electrons in metal surfaces. This discovery provides a greater understanding of the fundamental physical processes involved in such surface events as adhesion, corrosion and catalysis.

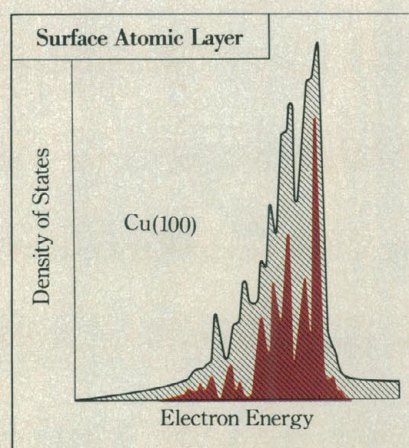


Figure 1: Energy distribution of electrons in outermost atomic layer. Red area indicates electrons in surface states.

Figure 2: Two electron density contour maps of the cross-section of a Cu(100) surface. One map shows a clean copper surface (tan); the other shows a nitrogen-covered copper surface (green).



CONVENTIONAL scientific thought treats virtually all of the valence electrons found in the surface atomic layer of a metal as if they are free to roam throughout the metal's interior. The work of three physicists at the General Motors Research Laboratories suggests otherwise. Through calculations confirmed by experimental data, the theorists have shown that more than a quarter of the valence electrons in the top atomic layer of some metals are effectively trapped in the surface. The presence of so many "surface state" electrons must be considered when analyzing physical and chemical surface phenomena, including such surface events as oxidation leading to corrosion.

Drs. John Smith, Jack Gay and Frank Arlinghaus applied their theoretical analysis to the (100) surface of five metals: copper, nickel, silver, rhodium and palladium. They made bold predictions concerning the percentage of electrons in the surface atomic layer to be found in surface states: Cu(36%), Ni(23%), Ag(23%), Rh(23%) and Pd(19%). The ratio of the red area to the hatched area of figure 1 gives the percentage for copper.

Electrons in surface states are not only abundant, but also highly localized on the surface. Chemisorption on a metal is also confined to the surface region. Figure 2 shows what happens in the case of nitrogen chemisorbed on copper. The two contour maps coincide except in the surface layer, where the interaction is largely exhibited. Localization of the interaction holds for the chemisorption of other gases, including oxygen in the initial stage of metal oxidation. These observations led the physicists to conclude that surface states are important in chemisorption.

One way to probe electrons in surfaces is to chemisorb atoms on a clean metal surface and look for changes in photoemission spectra. Such an experiment was performed at GM for fractional monolayers of nitrogen, oxygen and sulfur on Cu(100). The dominant change in the photoemission spectrum was the disappearance of a large peak whose shape and en-

ergy location was independent of the chemisorbed atom. It was of special interest that the shape and energy location of this peak was nearly identical to the envelope around the surface state peaks in figure 1. This suggests that surface state electrons play a major role in the chemisorption process.

THE THEORETICAL advance at the heart of the discovery is the "Self-Consistent Local Orbital (SCLO) Method" for solving the Schrödinger equation. This new mathematical method was devised by the GM theorists to handle the classic dilemma posed by the self-consistency requirement. The characterization of electron behavior used to complete the equation must be consistent with the behavior predicted by the equation. In other words, one almost needs to know the answer in order to make the calculation.

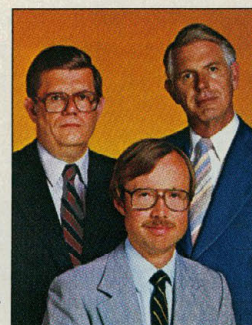
Self-consistent solution of the equation for a metal surface is made exceedingly difficult by the three-dimensional nature of the electron density distribution. The theorists dealt with this challenge successfully by dividing the electron density distribution into two parts—the first part due to overlapping atomic density distributions; the second part equaling the difference between this atomic contribution and the exact density distribution.

One of the more stringent tests of the accuracy of the SCLO method was an angular photoemission experiment conducted by Heimann et al. at the University of Munich, subsequent to publication of the GM research. The German research team confirmed a prominent surface state band predicted by the three GM physicists. This was the first time a surface state band on a solid had been calculated prior to its being seen experimentally. The SCLO method makes possible something that could not be done before—accurate prediction of the actual behavior of electrons whirling around nuclei at the surface of a metal.

"The large body of surface states we found on metal surfaces," says Dr. Smith, "may be a controlling factor in many physical and chemical surface phenomena. By replacing conjecture with calculation, the new surface theoretical methods give us the means to make major steps forward in the analysis of surface and interface properties."

THE MEN BEHIND THE WORK

Drs. Smith, Gay and Arlinghaus are theorists in the Physics Department at the General Motors Research Laboratories.



John Smith (center) and Jack Gay (right) received doctorates in physics; Smith from Ohio State University and Gay from the University of Florida. Frank Arlinghaus received his Ph.D. in physical chemistry from the Massachusetts Institute of Technology.

John Smith, leader of the GM solid state physics group, did postdoctoral work at the University of California in La Jolla. He joined General Motors in 1972. Frank Arlinghaus and Jack Gay joined the corporation in 1964 and 1965, respectively.

Each member of the team brings to the project a different expertise: Smith in surface physics, Gay in solid state theory, and Arlinghaus in bulk band structure calculations.



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Long-Term Changes in the Chemical Industry

A combination of factors has caused the chemical industry to examine its future feedstock and energy sources. At present, petroleum and natural gas are by far the predominant sources of organic carbon and energy. However, increases in the price of oil and natural gas have had impacts on conservation, process design, and choice of future feedstocks. The price of these hydrocarbons will likely tend to rise faster than inflation, creating additional pressure for changes in the industry and in its needs for trained people.

One alternative is coal. As a source of heat, its cost is about one-fifth that of oil or natural gas, and increasingly it is replacing other fuels. Coal can also serve as a source of such compounds as benzene, toluene, phenol, and naphthalene. In future, coal is likely to be consumed in large quantities to make synthesis gas, that is, carbon monoxide plus hydrogen, which can be converted to methanol, ethanol, and a host of hydrocarbons.

But for the long-term future and to meet worldwide needs, the most interesting feedstock is derived from photosynthesis. In contrast to fossil fuels, biomass is renewable, and supplies will likely increase substantially in the coming years. As feedstocks, wood and plant materials are more versatile than coal, and advances in biotechnology are likely to lead to innovative processes for manufacturing chemicals from them.

In terms of ash or sulfur content, plant material is cleaner than coal, and it is converted to synthesis gas more readily. But the really interesting chemicals produced by plants and trees are the diverse natural products, such as rubber, resins, terpenes, carbohydrates (including starch, hemicellulose, and cellulose), proteins, fats, waxes, and lignin. Up to the present, research to improve plants has largely focused on food crops. This effort has been quite successful and indicates that similar efforts would improve the yields of other desired products. Already better management of silviculture in some instances has led to yields five times those of natural stands.

Of all the products of photosynthesis, glucose and its polymers are probably the most important as chemical feedstocks. Glucose is a favorite source of energy and carbon for many microorganisms, both aerobes and anaerobes. They can convert the sugar into hundreds of different chemicals, including ethanol, citric acid, and pharmaceuticals. By use of selected mutants and of recombinant DNA techniques, it should be possible to target specific products and improve their yields.

In spite of the increasing costs of oil and natural gas, the transition to other feedstocks will occur only gradually. The petrochemical complexes are in place. Biotechnology is likely to have its initial expansion in the production of substances that cannot be derived from fossil fuels. But fermentation is beginning to have a small but significant impact on the petroleum industry. Until recently, nearly all industrial alcohol was derived from ethylene. Now an increasing fraction is coming from grain, and some of it is being used in gasoline in an application other than gasohol. Some oxygenated organic compounds, including ethanol, increase the octane number of gasoline and thus serve as a substitute for tetraethyl lead.

For decades, biotechnology was overshadowed by the petrochemical industry. Fermenters producing ethanol, acetone, and butanol were dismantled. The pharmaceutical and food industries kept industrial microbiology alive, but academic training in biotechnology was almost nonexistent. Thus there are few academic experts in the field at a time of expanding opportunity. Some of the nation's most creative people are scientists versed in molecular biology, including recombinant DNA techniques. However, if their work is to lead to the production of useful products for society, biotechnology and biochemical engineers must be involved.

Many of the major chemical companies recognize that important future opportunities involve biotechnology and they are building their staffs accordingly. Students who have inclinations and aptitudes for the natural sciences can confidently seek training in biotechnology with the conviction that the field will expand in significance and usefulness for many decades.

—PHILIP H. ABELSON

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