FROM THE CURRENT LITERATURE

Vacuum tunneling microscopy and spectroscopy

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> "It reveals the mysteries of nature
> to the astonished soul"1)
> J. W. von Goethe

One of the most significant achievements of the 1980s in physics was the invention of a new and very powerful method of topography and analysis of solid surfaces—scanning tunneling microscopy. Now, after five years have elapsed since the first publication on this topic,[1] the stream of papers describing new versions of tunneling microscopes and their use in science is growing rapidly. The significance of these investigations, which have opened a new page in microscopy, extends beyond the bounds of solid state physics and touches upon many areas of our knowledge. The best support for this assertion is the awarding of the 1986 Nobel prize in physics to the inventors of the scanning tunneling microscope (STM), G. Binnig and H. Rohrer (along with E. Ruska, honored with this award for the development of the electron microscope).

Improvements in the construction of the STM have resulted in fantastic resolution, reaching 0.02 Å normal to the sample surface and 2 Å in the plane of the sample. However, its advantage lies not only in these values of resolution, which are a record for microscopy. In contrast to the electron microscope, the STM examines the surface directly; it does not damage or change the sample, it operates in a gaseous or liquid medium (with minor degradation of the resolution compared to that in vacuum), and it can be combined with other apparatus to obtain devices with qualitatively new characteristics.

The success that has been attained in this field has brought about a situation in which, as stated in Ref. 2, "many investigators have been infected by the STM louse." The STM has so far been developed in a number of laboratories in the USA and Europe (see e.g., Refs. 2–4). The first Soviet STM is operating successfully at the S. I. Vavilov Institute of Physics Problems, Academy of Sciences of the USSR.[5–8] While the STM at first was used to determine structures of metal surfaces, it is now used for studying semimetals, semiconductors, adsorbed particles,[9] and even biological objects, which do not require, as in ordinary electron microscopy, outgassing or coating with a thin metallic layer[10] (of course, in all cases a necessary condition is the presence of a surface of a good conductor on which the object to be studied may be placed). A broad range of possibilities has been opened up for performing elastic and inelastic tunneling spectroscopy,[11] both localized and scanning. The development of a low-temperature tunneling microscope[12] permits the study of the electronic structure of the surface of superconducting materials, nucleation of superconducting regions on the surface, and the determination of the elementary excitation spectrum. These experiments take on special interest today because of the recent discovery of high-temperature superconductivity.[13]

In industry at the present time, 5–10% of the gross national product is expended on losses due to friction and wear. The scanning tunneling microscope is the ideal device for studying these processes and for the study of corrosion and catalysis, which is the governing factor in the larger part of modern chemical technology. All these phenomena are controlled by the structure of the first atomic layers of the surface, to which the conventional electron microscopy is absolutely insensitive. Modification of the STM for operation in the field emission mode[14] holds promise for its use for lithography and other types of material treatments which are of wide use in microelectronics (submicron lines have already been made by this means). The unique energy characteristics of tunneling electrons lie in the same range as the majority of chemical processes, and therefore by creating a beam of electrons of a specified energy one can induce a desired chemical reaction,[9] for example, one that causes a modification of the surface structure.[15] The invention of the scanning tunneling microscope, in the words of S. Johansson, the chairman of the Nobel committee on physics, "opens up unlimited possibilities for science."

Before turning to a description of the construction of the STM and the new results that have been obtained with it since the publication of the first notice of the STM in the January 1984 issue of Uspekhi Fizicheskikh Nauk,[16] let us discuss briefly the history of the problem. The idea of electron tunneling, which appeared in the early stages of the development of quantum mechanics, concerns the passage of an electric current through a vacuum gap separating two metal electrodes.[17] However, an experiment of this sort did not seem feasible at first because the distance between the metals would have to be of the order of 10 Å or less; i.e., a distance comparable to the nonuniformities on the surfaces of the conductors. In this connection the thoughts of Giaever, expressed in his Nobel address, are of interest: "...we decided early in the game not to attempt to use air or vacuum between the two metals because of problems with vibration. After all, we had training in engineering!".[18] For the potential barrier Giaever proposed to use the thin oxide layer that forms naturally on the surface of one of the metals. Although subsequently the devices of tunneling spectroscopy were exclusively metal-insulator-metal Giaever-type structures,[17,19] attempts to produce a vacuum gap did not stop. This persistence can be attributed to the obvious advantages of this type of barrier: the electrical or mechanical properties of the insulator have no effect on the tunneling characteristics; one is rid of effects due to inelastic tunneling through impurity centers in the barrier; there are no contact phenomena at the metal-insulator interface; and finally, as subsequent investigations have shown, it is possible to vary in a controlled way the spacing between the electrodes.

The first attempt to create a vacuum barrier by forming a gap between electrodes as a result of placing together two separately prepared samples was undertaken at the Institute of Radioelectronics, Academy of Sciences of the USSR by Lutskiĭ and his coworkers.[20] The first device with a variable vacuum gap was developed by Young *et al.*,[21,22] who used a piezoelectric transducer for scanning an emitter with a radius of about 1000 Å over the sample surface and a feedback system for supporting the emitter a given distance from it. The resolution of this device, which was used for the microtopography of metal surfaces, was 30 Å normal to the sample and 4000 Å in the plane of the surface.[22]

The first spectroscopic investigation using this sort of experimental technique[21,22] was undertaken by Poppe[23] on superconducting samples of $ErRh_4B_4$. The gold or silver tip was pressed onto the surface of the crystal being studied, and then it was raised with a piezoelectric transducer a distance sufficient to generate a tunneling current across the vacuum gap that had been formed. The clear observation of the energy gaps in the dependence of the differential conductivity on the bias in this and other superconducting materials indicates the tunneling nature of the current that is produced in the system.

Radical progress in the field of vacuum tunneling was achieved in the work reported in Ref. 1, carried out by workers at the IBM research laboratory in Zurich, Switzerland. These investigators improved the shielding from internal and external vibrations, they substantially improved the stability of the surface sample and the tip, reduced considerably the width of the tunneling gap, and they developed in detail the mechanism for moving the emitter. Let us discuss briefly the operating principle of this device. With the use of a piezoelectric transducer the metal tip is moved along the normal z to the surface of a conductor until there is a finite tunneling resistance. Since the electron wave function falls off exponentially in the potential barrier that is formed, the tunneling resistance R and current $I = RU$ are very sensitive to the gap width d (in Å):

$$R = R_0 \exp\,(1{,}025\ d\varphi^{1/2}),$$

where φ, in eV, is the average of the work functions of the two materials that form the tunnel junction. This expression is valid for a gap voltage $|U| \ll \varphi/e$. At a constant voltage, the tip is moved above the surface. The tunneling current and, consequently, the resistance R and the product $d\varphi^{1/2}$ are held constant by means of a feedback system which controls the piezoelectric transducer. Then the voltage u_z applied to the controlling peizoelement is varied in accordance with the relief of the sample surface. If φ is constant along the surface, then the signal u_z that is obtained while scanning gives directly information on the topography of the surface on an atomic scale.

The construction of the STM described in Ref. 1 contains three main elements: a coarse mechanism for bringing the tip and the sample together (more than 1 μm); a fine-motion mechanism (less than 1 μm), and a system for vibration isolation and damping. For coarse stepwise lateral motion, i.e., in the x-z plane, a "spider"-type piezoelectric drive is used. The tip is moved smoothly up to the surface with the use of a ceramic piezodrive, which moves the tip over a range of a few thousand angstroms (in the first STM a standard piezoceramic made by the Philips company was used to move the electrodes in three mutually perpendicular directions). The "spider" and the motion along the x, y, and z axes are controlled by computer, whose task includes presenting the tip to the sample surface nondestructively, reaction to substantial nonuniformities, orientation of the scanning plane, compensation for thermal drift, etc, as well as storing the information in memory and reading it out to a disk.

The resolution that the STM can attain is governed by a number of technical factors, among which we may point out the following: a) thermal drift; b) external vibrations; c) obtaining a tip with the minimum curvature; and d) achieving ultrahigh vacuum.

The first problem was solved successfully in the work reported in Ref. 2. To prevent thermal drift in the z direction a compensating structure was used, and for thermal drift in the x-y plane a symmetric network of piezoelectric transducers was used. These measures, along with a compact scanning unit, made it possible to reduce the sensitivity to vibrations; they resulted in a resolution of at least 10 Å in the scanning plane and a reaction time constant of at most only 0.3 ms; i.e., a rapid rate of scanning (400 points per second).

The resolution along the normal to the surface is determined mainly by vibrations of the tip relative to the plane. It should be noted that the system of vibration isolation and damping has been undergoing continuous modification and has been simplified as the construction of the STM has been perfected. In the first generation of STMs a superconducting suspension was used for this purpose.[1,24] In the second[25] and third[9] generations the suspension systems for the STM were two-stage spring systems differing in their basic dimensions. Finally,[26] a miniature STM was described, in which coil springs and damping magnets were completely absent. In this case the system for vibration isolation was a stack of stainless steel plates, one on top of the other, with three (or more) Viton damping spacers between each pair of adjacent plates. This arrangement proved to be sufficient for a tunneling gap stability of the order of a few angstroms in an STM operating in an atmospheric pressure of air on an ordinary laboratory bench. Of course, additional shielding from sound and large-amplitude vibrations can improve the stability by at least an order of magnitude. Besides isolation of the STM from external vibrations, an important characteristic of the system is its rigidity and compactness which serve to increase the resonance frequencies of the instrument.[2]

The resolution in the plane of the sample that the scanning tunneling microscope can attain depends mainly on the shape of the tip: the smaller the radius of curvature the higher the resolution of the instrument. According to Ref. 27, it is best to form the tip *in situ*. Simply touching the tip to the sample surface is sufficient to obtain a resolution the order of 10 Å. By means of heating and by passing large currents from the tip to the sample one can obtain a resolution in the x-y plane of the order of 2 Å (Ref. 27), while in the z-direction the best resolution[28] is 0.02 Å (these values were cited at the beginning of this paper).

Of course, the resolution can be improved if the measurements are carried out in vacuum: in the first experiments the vacuum was $\sim 10^{-6}$ torr, while in subsequent experiments it was $\sim 10^{-10}$ torr. Nevertheless, it should be emphasized that the tunneling microscope can also operate directly in air with fairly good resolution (see e.g., Ref. 26).

Up until now we have discussed the tip-plane type of STM construction. Another way of creating a gap between two metals has been developed[29] (see also Ref. 30). The authors of Ref. 29 pointed out that for spectroscopic purposes a gap stability of 0.01 Å is required. To solve this problem they suggested a method of producing a tunnel junction by squeezing two crossed electrodes deposited on two substrates separated by a thin-film spacer. By means of an electromagnetic device that is placed in a dewar with liquid helium the gap between the metals can be varied smoothly and the substrates are deflected by such an amount that a tunneling current is set up between them. Measurements carried out at 1.2 K revealed the energy gap of the superconducting plates in the voltage-current characteristics of lead-vacuum-lead and lead-vacuum-aluminum junctions; this observation served as proof of the tunneling nature of the current across the junction. Without any feedback system and with minimal means of suppressing vibrations, structure due to lead phonons was reliably observed in the second derivative of the tunneling current with respect to the voltage, and was completely identical to similar features observed previously with Giaever junctions with a barrier of aluminum oxide. This result indicates that the junctions that were produced had an extremely high stability, which, nonetheless, was insufficient for observing molecular vibrations by use of the inelastic tunneling effect.[29]

A tunneling microscope which combines construction elements of the scanning tunneling microscope described in Ref. 1 with a device for squeezing the tunnel junctions[29] has been described in detail in Ref. 31. In developing this instrument the authors of Ref. 31 aimed at two main goals: attaining maximum rigidity of the system and minimum thermal drift. The coarse motion of the tip in this arrangement was provided by a screw with a very fine thread, and piezoelectric transducers were used for scanning in the plane of the sample and for fine motion along the z axis. High resolution was attained in liquid nitrogen, and images of individual atoms on the surfaces of close-packed layers were obtained. The resolution in the scanning plane was better than 3.4 Å and the best resolution in the vertical direction was of the order of 0.1 Å.

Let us turn now to a description of the first Soviet scanning tunneling microscope.[5] The tungsten tip in this STM is moved by means of a titanium ceramic piezoelectric transducer. The STM is suspended by a shock absorber under the bell jar of a vacuum system (the vacuum of 10^{-2}–10^{-5} torr provides acoustic isolation) which is in turn supported by a spring suspension and rubber pads. The STM is controlled, and the results of the scanning are stored, by a "Mera-60" computer. According to Ref. 6 the device is scanned over a relatively large area of $10\times 10\,\mu$m with close to atomic resolution. This microscope was used to study for the first time the relief of the Si–SiO_2 interface in a metal-insulator-semiconductor (MIS) structure,[6] a study that is extremely important for the adequate description of the properties of this object. Long-period irregularities in relief with a characteristic height of 10–30 Å and hundreds of angstroms across were discovered with the STM. According to Ref. 6, these irregularities are the principal source of electron scattering, and this phenomenon must be incorporated into the corresponding theories that describe electronic processes in MIS structures. This same microscope has been used to study the surface of niobium films,[7] from which niobium-niobium oxide-lead thin film structures are prepared. The nature of the formation of the tunnel junctions, with a multitude of microbridges penetrating the oxide layer, was discovered: it was found that protuberances—crystallites about 40–50 Å high—appeared on the surface of niobium films deposited on a heated substrate, and these protuberances probably serve as the microbridges in the oxide layer. Definite progress is represented by the development of a fast STM,[8] which permits study of kinetic phenomena in real time.

The most impressive progress up until now is that in scanning tunneling microscopy applied to studying the details of the electron structure of metal and semiconductor surfaces. The atomic arrangements in the 7×7 cell structure of the silicon surface[9] and in the surfaces of germanium, GeSi,[32,33] and GaAs[34] have been studied extensively, and the Si(001) surface has also been studied.[35] The successful resolution of structure at the atomic level shows that the new microscopy opens up new and unprecedented possibilities in the study of the physics and chemistry of surfaces. The microscopy is not limited to just vacuum-solid interfaces, but can be also applied to gas-solid and liquid-solid interfaces, which play an important role in catalysis, corrosion, lubrication, and biology. The ability of the scanning tunneling microscope to operate in ordinary air at ordinary pressures more than compensates in many cases for the certain amount of loss in resolution. An important feature is that the STM makes it possible in principle to study, as they are developing, such surface phenomena as the dynamics of dislocations, superconducting regions, living organisms, chemical processes, etc. A promising direction also is the combination of the STM wih other instruments. For instance, investigators have recently been able to combine an STM with an ordinary scanning electron microscope,[26] and a device, based on an STM, has been developed that can measure ultrasmall forces (to 10^{-18} N).[36] The technique of transverse electron focusing together with scanning tunneling microscopy makes it possible to obtain information on the same surface from without and from within, such that the two types of information obtained complement each other.[37]

Further improvement in the construction of the STM, and, in particular, improvement in the stability of the vacuum gap, is opening up a broad field for the application of elastic and inelastic tunneling spectroscopy. Here a principal interest is in the study of elementary excitations in "exotic" superconductors, layered and nonuniform superconducting structures, and magnetic superconductors (the results of the first experiments on superconducting samples are reported in Ref. 37). The scanning tunneling microscope permits a detailed study of the anisotropy of the superconducting properties of materials. Binnig *et al.*[38] have discussed the possibility of observing inelastic tunneling processes with the STM, a technique that would allow one to determine the chemical nature of individual adsorbed molecules by observing the corresponding features in the tunneling curves (see also in Ref. 39 a calculation of the cross section for inelastic scattering of electrons by a molecule located on the surface of a planar electrode). Estimates show[38] that for a barrier height of the order of 2 eV it is necessary to achieve a stability of 0.02 Å in the vacuum gap (the attainment of the corresponding parameters has been reported in Ref. 28). It appears possible to use the STM for investigating resonance tunneling of electrons through a small aggregate of point defects in an insulating film that covers a conducting plane.[40]

The experimental progress that has been attained in a comparatively short time has stimulated the development of the theoretical foundation of vacuum tunneling microscopy and spectroscopy. Let us formulate some of the questions that are raised in these investigations, taking these questions particularly from Ref. 41.

1. What requirements are imposed on the tips that are used for one of the electrodes?
2. How must the experimental data be analyzed in order to obtain accurate information on the electronic and geometric characteristics of the sample surface?
3. Are there any theoretical limits to the vertical and transverse resolution of the STM?

To answer these questions a consistent theory of tunneling in the tip-plane system is necessary[41–44] (see also the discussion in Ref. 45). Let us point out that Leavens and Aers[46] have adapted the theory developed in Ref. 42 to take into account thermal vibration of the atoms of the crystal lattice, an important consideration for the interpretation of results obtained with the STM at room temperature. We should also mention the work of Binnig *et al.*,[47] who have pointed out the imporant role of the image potential.

Theoretical investigations have addressed particular attention to the resolution of the tunneling microscope. In the case of small-scale irregularities the resolution of the STM is determined not only by geometrical dimensions—the radius of the tip and its average distance from the surface—but also by the decay length of the wave function in the vacuum barrier.[44,48]

Research during the time since the appearance of the first publication can be characterized as an intensive search for solutions to problems of increased sensitivity, stability, immunity to vibrations, the development of various mechanisms for moving the injector tip, and others. The principal achievement—a controllable vacuum gap of atomic dimensions—has, however, become a reality, and with the use of the scanning tunneling microscope a resolution hitherto unrealized has been attained. The great Goethe was indeed correct: "Let the imagination stand numb in terror before the fatal threshold."[1)]

[1)] Retranslated from the Russian text.

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