Comparison of transverse-electron-focusing and scanning-tunneling-microscopy measurements on Ag(001) and (011) surfaces

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Chemically etched Ag (001) and (011) surfaces have been studied from inside the sample by transverse-electron-focusing (TEF) experiments and from outside the sample with a scanning tunneling microscope (STM). The TEF experiments give mean reflection coefficients of 0.37 ± 0.03 for the (011) surface and 0.64 ± 0.02 for the (001) surface. This difference is confirmed by the STM data, which show a hilly (011) surface, while the (001) surface consists of large, atomically flat terraces. With the additional information from the STM, the unexpected high specular reflectivity, which is often observed in TEF experiments, is explained.

I. INTRODUCTION

Recent advances in manipulating conduction electrons within metals by point contacts and magnetic fields (transverse electron focusing) on one hand and the invention of the scanning tunneling microscope on the other hand should make it possible to study one and the same surface from the inside and from the outside of the metal using the conduction electrons. It is the purpose of this paper to report on this sort of investigations and to use the comparison of the two methods to throw light on a long-standing problem in the field of transverse-electron-focusing experiments.

It has been shown before[1–10] that the transverse-electron-focusing technique (TEF) can be used to determine the probability for specular reflection q for electrons incident normal to the surface of a metal or semimetal. This technique has been applied successfully to a variety of different metals and semimetals: Bi,[1,2] Sb,[3] W,[4–6] Cu,[4,7] Ag,[8,9] Al,[9] and Zn.[10] In many of these experiments the reflection coefficient q turned out to be surprisingly high, despite the sometimes apparent roughness of the surface and the crude preparation techniques. The high specularity implies a surface smooth on the scale of the de Broglie wavelength of the conduction electrons (5 Å for Ag). One way out of this contradiction is to assume that the surface consists of atomically flat terraces, separated by steep slopes, such that the optical appearance is that of a rough surface. In search of a method to confirm this hypothesis, we have applied the scanning tunneling microscopy (STM) technique to these samples, because this technique has proven to be a powerful tool in matters of surface analysis. (For a review of the pioneering work of Binnig and Rohrer, and their collaborators, on this subject, see Ref. 11.) The STM technique is very suitable for obtaining detailed information on the surface roughness of bulk samples, on a scale sufficiently small to compare with the specularity obtained from the TEF experiments.

In this paper the STM technique is compared with the TEF technique, using Ag(001) and (011) surfaces. We first describe briefly the principles of the two techniques, the sample treatment, and the experimental methods. Next, the results of both experiments are presented, leading to a discussion in which the data are compared and interpreted.

II. PRINCIPLES OF THE TEF AND STM TECHNIQUES

The principle of the TEF technique is as follows: A current is injected into a metal single crystal through a point contact (the emitter). A uniform magnetic field in the plane of the surface is used to bend the paths of the electrons to bring them to a second contact (the collector). Here they produce a field-dependent voltage across the resistive junction between the contact and the surface (Fig. 1). The electrons of primary interest are those leaving the emitter, moving with a velocity perpendicular to the surface.[1] At certain values of the magnetic field these electrons reach the collector, either directly ($B=B_0$) or after one or more specular reflections from the crystal surface between the point contacts ($B=2B_0$, $3B_0,\ldots$), resulting in voltage peaks. When a fraction q of the electrons incident on the surface is reflected specularly, then each

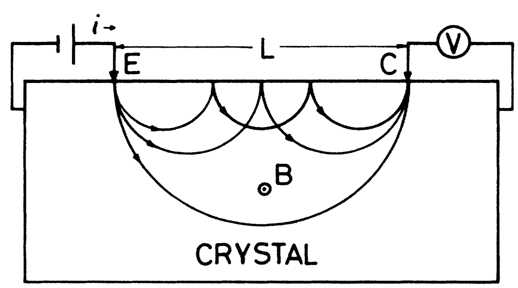

FIG. 1. Experimental setup of the TEF experiment. Electrons are injected through a point contact E and focused on another point contact C by means of a uniform magnetic field B. The electrons can also be focused on C after one or more specular reflections from the crystal surface. The point-contact distance L ranges from 50 to 500 μm, the injection current i is 300 mA, and the collector voltage V is 0.1 to 800 nV.

voltage peak is lower than the previous one by a factor q. So, it is obvious that q gives a measurement of the roughness of the sample surface.

In the case of STM, electrons tunnel through a vacuum barrier of a few angstrom from the crystal to a sharp metal electrode, under influence of a small voltage difference. The tip is fixed to a three-dimensional displacement device built up of piezoelectric parts.[12,13] While the tip is scanned along the surface, the tunnel current, and therefore the distance, are kept constant via a feedback voltage to the piezo element which moves the tip perpendicular to the surface. Thus, information on the topography can be obtained by monitoring the feedback signal.

III. EXPERIMENT

A. Samples

The samples used were Ag single crystals of 99.9999% purity,[14] spark cut from a single-crystal rod with one surface perpendicular to the [001] axis and the other perpendicular to the [011] axis. Ag has been used, firstly because it is obtainable in high purity, leading to a large electron mean free path at low temperatures. Secondly, the Fermi surface approaches the free-electron sphere very well, except in the $\langle 111 \rangle$ directions where it bulges out to make contact with the Brillouin-zone boundary.[15] The samples were etched chemically in a NH_3 solution ($>25\%$) of about 20 ml, with five to ten drops of a 40% H_2O_2 solution added. This yielded a shiny surface to the naked eye, terracelike for the (001) crystals, but rather rough for the (011) ones. The samples were annealed for eight hours at 850 °C in 10^{-4}-Torr air[16] and, finally, again a short etch cycle was applied with the same solution. This procedure yielded a residual resistivity ratio [R(300 K)/R(4.2 K)] of 15 000, leading to an electron mean free path l of about 700 μm at low temperatures.

B. TEF

The TEF measurements were performed at liquid-helium temperatures (≤ 4.2 K) and the insert was constructed in such a way that both the point contacts and the sample could be handled from outside while at low temperatures. Since the magnetic field was directed horizontally, the samples were mounted so that their surfaces were as horizontal as possible (typically within a few degrees). The point contacts consisted of 0.1-mm-diam W wires etched electrolytically in a 5N KOH solution, resulting in sharp points with a radius of about 0.5 μm. The distance between the point contacts ranged from about 50 to about 500 μm. After immersion in the helium bath the point contacts were spot-welded on the sample surface, using a 90-V battery with a 1-MΩ series resistance. This gave a rather stable contact, also in magnetic fields, with a resistance on the order of 0.1 Ω. The point contacts could be lifted again from the sample surface in the helium bath, e.g., to rotate the crystal or to renew the contact. Because of the stiffness of the tungsten, the wires kept their shape during these operations.

The insert was placed in a glass cryostat with an unsilvered tail, with the direction of the line connecting the point contacts such that their distance could be measured in the helium bath. The errors in direction and distance were strongly dependent on the quality of the sight in the bath, but were typically 1° and 0.01 mm, respectively.

The measurements were made using a current source, which supplied ac injection currents of about 300 mA rms at a frequency of about 24 Hz. The collector voltage was amplified with an impedance-matching transformer and a low-noise preamplifier and fed into a phase-sensitive detector, of which the output was plotted versus magnetic field on an x-y recorder.

C. STM

The STM experiments were performed in an ultrahigh-vacuum environment (2×10^{-10} Torr) at room temperature. The samples were mounted to a rough positioning device, opposite a 0.1-mm-diam W wire, which was treated as in the TEF experiment. The point was scanned along the surface in a constant-current mode (1 nA), with a voltage of 50 mV across the junction by means of the piezoelectric driving mechanism. In order to increase the density of information, successive mappings of the same area (1450×1300 Å^2) were made with mutually perpendicular scan directions. This was possible because of the high thermal stability of the device.[13] Each mapping was stored by a computer into a field of 40×200 points, i.e., 40 scans of 200 data points each. During the experiment the sample was displaced several times over a large distance (10—30 μm) in order to get an overall idea of the surface roughness.

IV. EXPERIMENTAL RESULTS

Figure 2 shows results obtained with the TEF technique for the two samples. Clearly, the peaks due to direct focusing (indicated with 0) can be seen, as well as peaks due to focusing after one or more specular reflections from the crystal surface (1, 2, etc.). Also a continuously rising background is present, which has to be subtracted in order to determine the peak heights. In Fig. 2(a) a measurement of the (001) surface with a high q (0.85) has been displayed, with the magnetic field directed along the [100] axis, and in Fig. 2(b), a measurement of the (011) sample with the field along the [111] axis. From about 100 experiments on different places of each surface, the ratios of the height of the first reflection peak to the directly focused peak were taken and subsequently averaged, yielding a mean coefficient for specular reflection q of 0.64 ± 0.02 for the (001) surface and 0.37 ± 0.03 for the (011) surface. The mean distance between the point contacts, L, was 150 ± 10 μm. Notice, that in previous publications[8,9] q was estimated from the maximal ratios of the measured peak heights. This was done to eliminate the influence of the surface roughness and to determine q as a function of the type of orbit the electron follows in the metal.[8] As we now actually look at the surface roughness, we have to take the average value of q.

Figure 3 shows results obtained by the STM technique. In general, the contours of the (001) plane showed large,

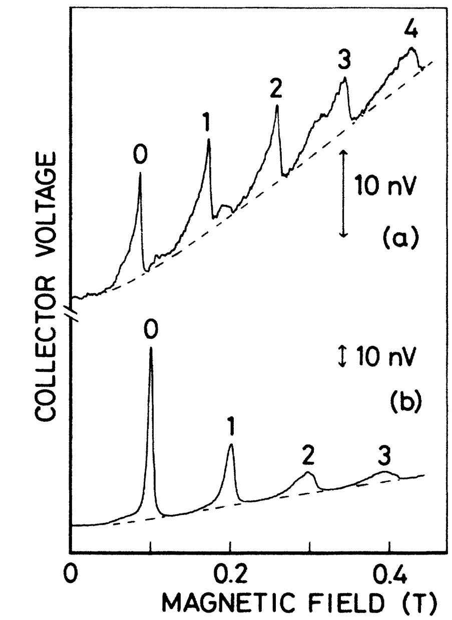

FIG. 2. Reflection measurements with TEF. Clearly, the peaks caused by direct electron focusing (0) and those caused by focusing after one or more reflections from the surface (1,2, . . .) can be seen. The dashed lines indicate the estimated background signal. (a) result from Ag(001), **B**||[100], $L=150$ μm; (b) result from Ag(011), **B**||[111], $L=200\,\mu$m.

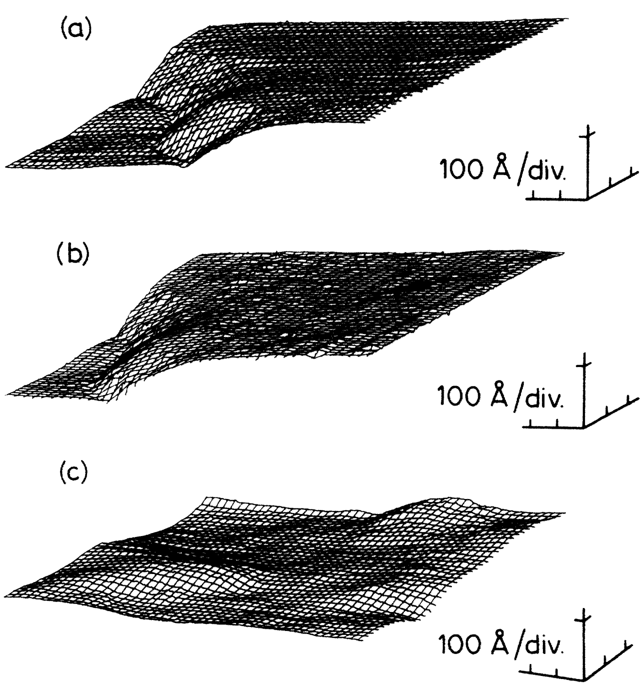

FIG. 3. STM mapping of (a) a Ag(001) surface of 1450×1300 Å^2 showing atomically flat regions separated by a large step (approximately 50 Å), (b) the same area scanned in a direction perpendicular to the previous one, (c) a Ag(011) surface of 1450×1300 Å^2 showing a smoothly varying structure. Note the different scales used for the in-plane and out-of-plane axes.

atomically flat terraces ranging from 300 to 1000 Å [Fig. 3(a)]. The step heights differed quite drastically between 4.0 Å (one unit cell) and about 100 Å. Great care had to be taken to prevent the tip from hitting the surface. A sudden collision resulted in a severely damaged Ag surface, showing large and steep edges in all directions.[17] The surface roughness of the (011) sample [Fig. 3(c)] cannot be described in as simple a way as the (001) sample. The topography changes from atomically flat regions via smoothly curved ones to highly corrugated regions. No decisive description can be given, although a general tendency of roughness in comparison with the (001) sample prevailed.

V. DISCUSSION

In order to make a comparison between the two techniques, a further analysis of the mechanisms involved and the data is necessary. In the TEF experiment, consider the point contacts to lie on the x axis and the applied magnetic field along the y axis. Now we calculate the deviation in the orbit of an electron which has been reflected from an oblique surface element between the point contacts, instead of a flat one. Suppose that the reflection at the tilted area takes place at the origin, with the emitter at a distance $-\frac{1}{2}L$ and the collector at $+\frac{1}{2}L$, with $\frac{1}{2}L$ the diameter of the cyclotron orbit. The normal of the surface element is given in polar coordinates (θ,ϕ). Consider the electron incident along the z axis. It will then be reflected over 2θ in the direction of ϕ. Because the distance traveled in the x direction is only dependent on the z component of the velocity just after reflection, it will be a factor of $\cos(2\theta)$ less than $\frac{1}{2}L$. This makes the deviation from the collector be

$$\Delta L_x = -\tfrac{1}{2}L[1-\cos(2\theta)] . \quad (1)$$

The deviation in the y direction is determined by the velocity v_F of the electron and its time of flight t after reflection:

$$\Delta L_y = v_y t , \quad (2)$$

where

$$v_y = v_F \sin\phi \sin(2\theta) \quad (3)$$

and t is the time necessary to travel along a segment of one circular cyclotron orbit:

$$t = \left[\pi - \frac{2\arctan[\cos\phi \tan(2\theta)]}{2\pi} \right] T_c , \quad (4)$$

where T_c is the cyclotron period. Furthermore, we can write v_F in terms of the cyclotron frequency

$$v_F = \tfrac{1}{4} L\omega_c . \quad (5)$$

When we substitute Eqs. (3)–(5) in Eq. (2), we find

$$\Delta L_y = \tfrac{1}{4}L \sin\phi \sin(2\theta)\{\pi - 2\arctan[\cos\phi \tan(2\theta)]\} . \quad (6)$$

In the case of the Ag(001) surface, the electrons which are focused on the crystal surface form a broad front resembling the front formed by electrons running over a spherical Fermi surface.[9] Therefore, the deviations of the electron orbits in the y direction do not play a role and ϕ can be taken as zero. Consequently, the detected reflection signal is mainly dependent on the slope of the surface in one direction only. The electrons, which reflect at the (011) surface, are confined in narrow channels due to the presence of the necks on the Fermi surface of Ag.[15] Therefore, an electron reflected from a (011) surface outside the plane perpendicular to the direction of the applied magnetic field is not compensated anymore by another electron, as in the (001) case. In order to account for the more complicated behavior of the electrons at the Ag(011) surface, one can regard the reflection coefficient to be dependent on two angular dimensions rather than on one.

From the STM mappings we now calculate the fraction of the surface which has an angle of inclination smaller than a certain given angle. First, the global angle of inclination of each area is estimated. Next, the relative tilt angles of both the line segments connecting two neighboring points in the scanning direction (a total of 40×200 line segments, one-dimensional approach) and the triangles, defined by three neighboring points (a total of $2\times40\times200$ triangles, two-dimensional approach), are calculated. Finally, the fraction of lines or triangles with a relative tilt angle smaller than a given cutoff angle gives a measurement of the reflection coefficient in one or two dimensions. Because noise fluctuations greatly affect the spread in the calculated angles of these small elements, each data point of a scan is averaged with its ten neighbors. The average values of a large number of measurements for different cutoff angles are shown in Fig. 4. As can be seen from these data, in the one-dimensional (1D) limit the fractions for Ag(011) are smaller than the fractions for Ag(001). One can conclude from this that the (011) surface is more corrugated than the (001) one. This tendency was noticed already by a visual inspection of the STM pictures [cf. Figs. 3(a) and 3(c)].

For a further comparison of the two experiments, we limit ourselves to the (001) surface, because a quantitative interpretation of the data in the (011) case is problematic with the available data, due to the complexity of the Fermi surface, as mentioned before. The TEF result for the (001) surface ($\overline{q}=0.64$) is represented by the dashed line in Fig. 4. The intersection with the 1D data leads to a critical tilt angle of 2.7°, which means that [via Eq. (1), with $\overline{L}=150$ μm) electrons with a deviation of less than 3300 Å are detected at the collector. When we assume that the point contacts are Sharvin junctions,[18] we can relate the contact resistance R_S and its radius b:[19]

$$R_S=\frac{4\rho l}{3\pi b^2}\ , \tag{7}$$

with ρ the resistivity of the metal and l the electron mean free path. For Ag, with $R_S=0.1$ Ω, we find $b=600$ Å, which should be compared with $\frac{1}{2}\Delta L_x(\theta=2.7°)=1650$ Å. This discrepancy is not unexpected for two reasons. Firstly, the finite top angle of the tip causes a tendency of the observed sloping parts to spread out. This will be a small effect because we can conclude from our data that only a small fraction of the tilted parts of the surface ($\simeq0.2\%$) has an angle approaching the top angle of the tip.[17] Secondly, because the point contact is spot-welded, it is not an ideal Sharvin contact, which means that the diameter will be larger than that deduced from the resistance.

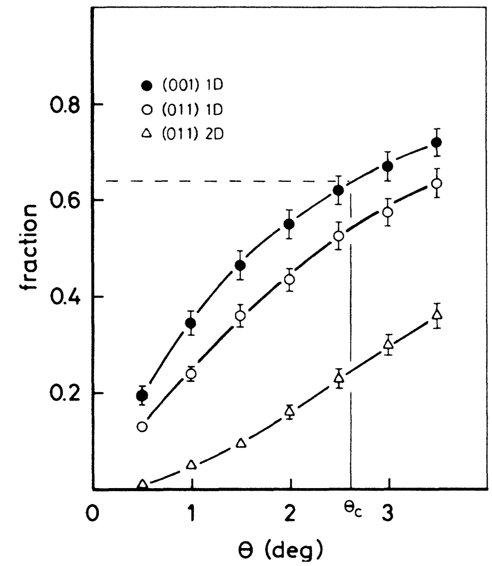

FIG. 4. Total surface fraction of Ag(001) (solid symbols) and Ag(011) (open symbols) with a tilt angle less than a maximum angle θ (solid lines) versus θ, as calculated from the observations by the STM method. The results are interpreted by two methods: a one-dimensional analysis (circles) and a two-dimensional analysis (triangles) (see text). The analysis has been done for fixed angles indicated by the symbols; the lines have been drawn to guide the eye. If the specular reflection coefficient q from the TEF experiment is identified with the plotted fraction, then the observed $\overline{q}=0.64$ for the (001) surface leads to a critical angle $\theta_c=2.7°$ as indicated.

VI. CONCLUSIONS

The hypothesis that the high specular reflectivity for conduction electrons of etched surfaces of metal samples is due to the fact that the surfaces consist of atomically flat terraces is confirmed for the Ag(001) surface by STM measurements. The Ag(011) surface, which shows a consistently lower specularity than the (001) face, has a clearly more hilly surface, as revealed by the STM. The described experiments have shown that the same surface can be studied from the inside and from the outside, and that useful complementary information can be obtained.

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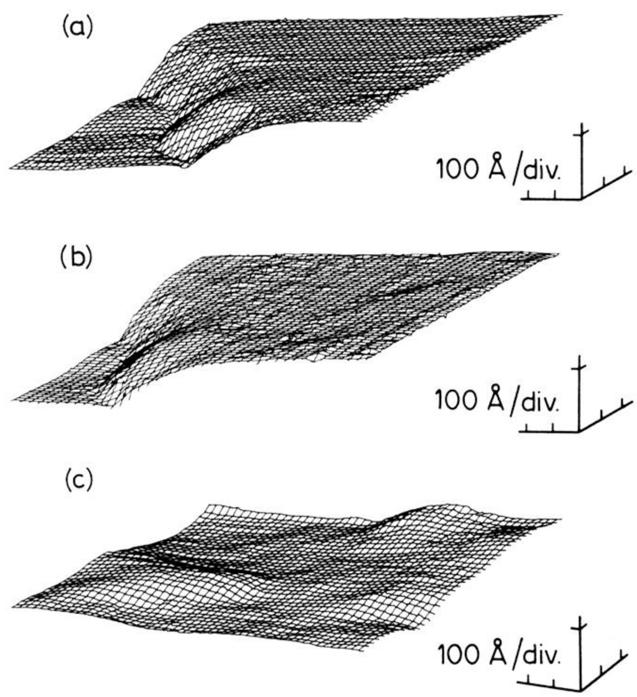

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