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Field emission from atomic size sources

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The source area of electron field emission from ultrasharp $\langle 111 \rangle$ -oriented tungsten tips terminated by a few (down to one single) atoms is confined to the very end of the tip, thus forming an atomic size electron source. The most remarkable emission property of such tips, the angular dispersion of the emitted electron beam, is measured both qualitatively and quantitatively for well-defined atomic arrangements at the tip apex and also for a wide range of applied electric fields and currents. It is shown that only pyramidally shaped tips emit electrons in sharp beams, with small opening angles down to less than 3° . The results suggest, in contrast to existing theories, that mesoscopic parameters, as the overall shape of the tip apex (on a scale of a few nanometers) and the associated patch field distribution, might play a crucial role in focusing the beam.

I. INTRODUCTION

Fabrication of ion sources of atomic dimension was achieved a few years ago.¹ These sources are ultrasharp field emission tips, with noble gas ions being produced by field ionization above the atoms, possibly above one single atom, located at the very end of the tip. From field ion microscopy, which gives atomic resolution, it is clear that the emission area may be as small as the size of an atom. Electrons can also be produced from these tips by field emission, but because of the lack of atomic resolution with electrons, the actual size of the source for electron emission has not yet been measured, but is expected to be of atomic dimension.

One remarkable property of these sources, that was mentioned from the very beginning, is the small opening angle Θ of the ion and electron beams that these sources can produce. These angles can be as small as 0.5° with ions and less than 3° with electrons. (In this paper the emission half-angle Θ is defined as the ratio of the radius of the spot produced by the beam on the screen to the tip-to-screen distance.) Electron emission then appears different from that of classical field emission tips, i.e., tips with a typical radius of 100 nm, where opening angles are one order of magnitude larger.

The development of these atomic size electron sources, especially with their outstanding emission properties, has roused hopes of practical applicability in, for instance, electron microscopy² and electron holography.³ Although some theoretical work has been done recently, we are still unable to predict the influence of the various parameters of these systems on the emission characteristics of ultrasharp tips. This is one of the reasons why the purposeful optimization of these devices and, consequently, their functionality is still very restricted.

Apart from these more application-oriented fields,

atomic size sources are a very interesting system because the quantum mechanical processes responsible for the emission can be investigated by varying the parameters over a wide range (the tunneling current, for instance, can be varied over 10 orders of magnitude). Theoretical predictions may thus be largely checked by performing the corresponding experiments.

Theoretical understanding of the electron emission divergence in such an experimental situation has been sought by some authors.^{4,5} The general idea of these attempts is to examine the focusing of particles of wavelength λ when they go through windows of similar lateral dimensions. It was shown that such models can explain focused beams by suppressing the tunneling of particles with high transverse momentum.

However, there is a lack of experimental data. The only data presently available are purely qualitative and were obtained by measuring the size of the spot on a screen placed behind a microchannel plate, necessary to amplify the intensity. Limitations come mainly from the small dynamics and the nonuniformity of such a detector.

The aim of this paper is to provide experimental data concerning the opening angle of the electron beam for different atomic arrangements at the tip apex, and for a wide range of applied electric fields and currents.

II. EXPERIMENT

The experimental chamber used in this study is basically a combined field-ion/field-emission microscope (FIM/FEM) as outlined schematically in Fig. 1.

The emitter employed in this investigation is a $\langle 111 \rangle$ -oriented tungsten single-crystal electrochemically etched *ex situ*. It is then mounted on a loop (a) that can be resistively heated. A copper cylinder (b) serves as both a heat shield and as a counterelectrode to the tip. It is connected to ground potential in order to keep the space between the cylinder and the analytical devices free of field. Two plates (c), fixed at the exit bore of the cylinder and connected to symmetrical potentials relative to ground, are used to correct a possible misorientation of the tip axis. By

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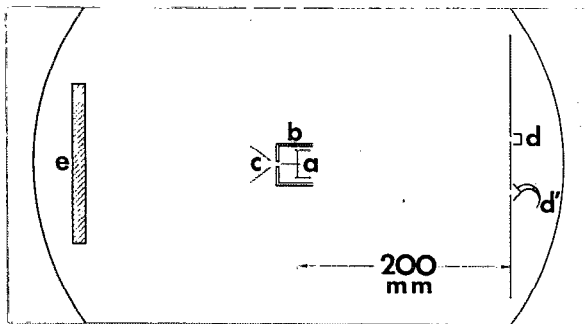


FIG. 1. Schematic of combined field-ion/field-emission microscope for both qualitative and quantitative measurements of the opening angle of charged particle beams emitted from ultrasharp tips. See text for description.

observation of the FIM pattern one can check that the charged-particle trajectories show no distortion. In order to obtain clear FIM images, the loop can be cooled with liquid nitrogen. The entire device can be rotated over the analytical devices to scan the emitted beam.

A Faraday cup (d) and a channeltron (d') used to quantitatively analyze the spatial distribution of the emitted beam are placed in the plane of rotation, as is a channel plate (e) for qualitative analysis and FIM. The acceptance angles of channeltron and Faraday cup allow angle resolved measurements with $\approx 0.2^\circ$ resolution.

One of the advantages of our combined FIM/FEM lies in the possibility of switching rapidly between FIM and FEM mode. The former permits analysis of the geometry of the tip apex with atomic resolution and the modification of this shape *in situ*. The latter is employed to measure the field emission properties of the tips prepared and characterized in this way.

By comparing the apparent angles between different facets as measured in FIM images to the corresponding crystallographic angles of the W crystal, the so-called compression factor of the imaging system can be analyzed (Fig. 2). This factor is mainly due to the actual form of the electric field as formed by the copper cylinder serving as the counterelectrode. The field is not radially symmetric and thus further focuses the emitted charged particles. This factor is, of course, the same for ions in FIM and for electrons in FEM. The emission half-angles as measured using the final spot size of the beam can therefore be corrected to obtain the true emission half-angles.

It is worth emphasizing that these true emission half-angles correspond to opening angles at very small, i.e., microscopic, distances from the tip. Therefore, a comparison of theoretical calculations limited to trajectory calculations in the close vicinity of the tip is possible. In order to stick to the experiments, we shall, however, give the measured angles.

The experimental method of investigation in the case of electrons consists mainly of the following cycle, which is carefully repeated for each data set: (1) Preparing a special atomic arrangement at the tip apex using FIM methods like field evaporation, sputtering, and thermal buildup.

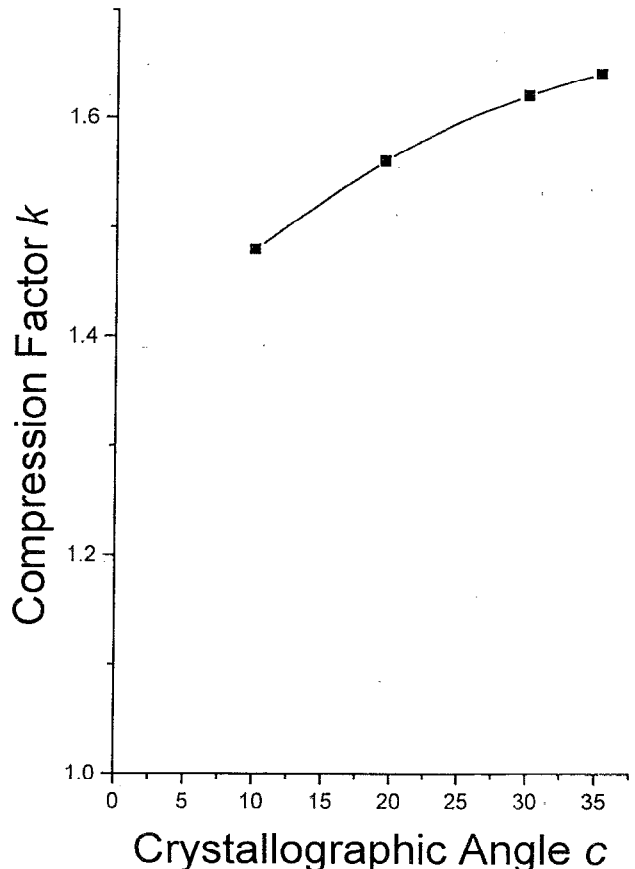


FIG. 2. Compression factor k vs the crystallographic angle c , defined as the ratio of c to the measured angle m between the same facets. Note that for $c=0$ by definition $k=1$.

(2) Pumping the imaging gas while leaving the tip at positive potential. (3) Switching to FEM mode only under UHV conditions; then measuring the spatial distribution of the intensity of the emitted electron beam at various tip potentials. For each potential the tip holding device is rotated, thereby scanning the beam over the Faraday cup and the channeltron. From these intensity versus angle curves we get Θ by measuring the full width at half maximum of the peak. (4) Turning back to positive potential, then letting in the imaging gas, and checking the unchanged atomic arrangement at the tip apex to verify whether the conditions during the measurements have been stable. Between different cycles the tip can be resharpener from time to time using preferential neon ion sputtering,⁶ followed by careful annealing of the tip.

Another means of "quality control" is provided by the analysis of Fowler-Nordheim plots derived from the data points also obtained in step (3): tunneling current versus applied potential. These curves often exhibit steps indicating sudden changes in tip geometry.

Apart from cathode sputtering (see below), adsorption of atoms from the gas phase plays a special role even under UHV conditions. The existence of any adsorbate cannot be deduced unequivocally by the FIM images as taken in step (4) of the cycle because of the low level of field necessary

to field-evaporate these species. However, the (non)linearity of the Fowler–Nordheim plot offers a means to sort out data sets of tips which have undergone changes due to adsorption. The same holds, of course, for any evaporation effect taking place during the blind phase, in the absence of the imaging gas.

All in all, about 90% of the data sets have been eliminated according to the given criteria. Only the remaining data sets were used and can be considered to be gathered under stable tip conditions.

To allow comparison of data sets gathered with different tips we use a fictive field $F^* = U/r_{\text{FN}}$, which is proportional to the real field at the tip surface. U is the effective potential of the tip, r_{FN} the mean radius of the tip as calculated with the classical procedure⁷ using the Fowler–Nordheim current versus potential plot.

Comparison of r_{FN} with the radius of curvature at the very end of the tip as deduced from FIM images⁸ shows a clear tendency: r_{FN} normally is found to be about two times larger than the radius of curvature between the {110} and the (111). Typical r_{FN} values of the tips used are of the order of 100 Å. Finer tips can be prepared but are difficult to be imaged by FIM as they tend to be very unstable.

The fictive field F^* varied in our experiment between 1.1 and 3.6 V/Å. Assuming a correction factor of 5 as in classical FE microscopy,⁷ this corresponds to real fields in the range of 0.2–0.7 V/Å, which is in good agreement with the corresponding fields known from the FIM mode. These should be larger by one order of magnitude.

Changing the field by a factor > 3 causes the emitted current to vary over more than 10 orders of magnitude. The combination of channeltron and Faraday cup allows continuous measurements in FEM mode from single electron emission to emitted currents of up to 10^{-3} A. The upper limit, however, is mainly imposed by the increasing tendency of self-destruction by the tip when used in this regime. The origin of this problem is associated with electron-induced desorption⁹ from surfaces hit by the electron beam. The desorbed particles impinge on the tip and lead to sputtering, thus destroying the atomic arrangement prepared in step 1. Therefore, in reality, we get reproducible results only for emitted currents of up to 10^{-5} A. This still represents a range of over ten orders of magnitude available in the experiments.

III. RESULTS AND DISCUSSION

Following existing theories, one expects the opening angle Θ to increase mainly with increasing field and the decreasing size of the emission area. Assuming that the latter, in our case, is formed by the (111) or, more precisely, by the cluster actually forming the (111) facet (the “terminating multimer”), Θ should increase with a decreasing number n of atoms in this multimer.

We find, however, that the most prominent dependence of Θ is on the local geometry of the tip apex *surrounding* the emission area. Tips with otherwise identical parameters F^* or n may show extremely different emission patterns, as demonstrated qualitatively in Fig. 3.

Quantitative analysis of the round and pyramidal tips shows the expected increase of Θ with F^* , but the most striking feature remains the difference between the two kinds of tips (Fig. 4): above all, small emission angles are obtained only with pyramidal tips.

By round tips we mean tips obtained after field evaporation or heating which have reached their nearly hemispherical equilibrium form. Field ionization occurs on these tips not only above the (111) facet, but also on the surrounding regions. In contrast, pyramidal tips are produced by heating only to such an extent as to allow surface diffusion of W adatoms but not yet mass transport diffusion of bulk atoms. Due to the crystallography of <111>-oriented tungsten tips, these then form large {112} and {110} facets around the (111) one, thus reaching a polyhedral shape, the so-called pyramidal form. There is in general no need to apply a field as for thermal-field built-up tips. On the pyramidal tips field ionization occurs only above the (111) facet.

Having found that round tips give large emission angles, from then on we concentrated on pyramidal tips, and we tried to check whether n is an important factor.

In principle it is possible to heat the tip just enough as to get a perfect pyramid, which means a (111) “facet” consisting of exactly one atom. In this context we speak of the (111) facet even if it consists of only one single atom (the case of a terminating monomer). In many cases, however, one ends up with terminating multimers of up to ten atoms. With some practice one regularly reaches multimers of less than eight atoms and relatively often a terminating monomer.

By controlled field evaporation we diminish the size of these terminating multimers and measure Θ versus decreasing n . In each measurement we use a fixed value for F^* when in FEM mode (more precisely a fixed applied potential, assuming that changes in n do not alter r_{FN} drastically, which is confirmed by the constant potential one needs in order to get field ionization.)

During field desorption of the terminating multimer, we observed hardly any changes in the surrounding area. The local radius of curvature at the summit of this pyramid is so much smaller than the radius of the neighboring facets (mainly the large flat {112}) that the critical evaporation field is only reached above the multimer.

Once again we find Θ increasing with F^* , but no significant dependence upon n (Fig. 5).

These findings show that not only purely microscopic parameters concerning the emission area are of importance, but also mesoscopic factors, as the actual form or geometry of the surroundings of this area.

These factors, in general, are not considered by classical theories based upon the assumption of an idealized atomic size source: A single atom adsorbed on a smooth and plane surface, which means that any other irregularities are neglected compared to the width of the potential barrier. In principle then, only this single atom actually forms the source, whereas the surroundings should not influence the emission properties. These idealized sources

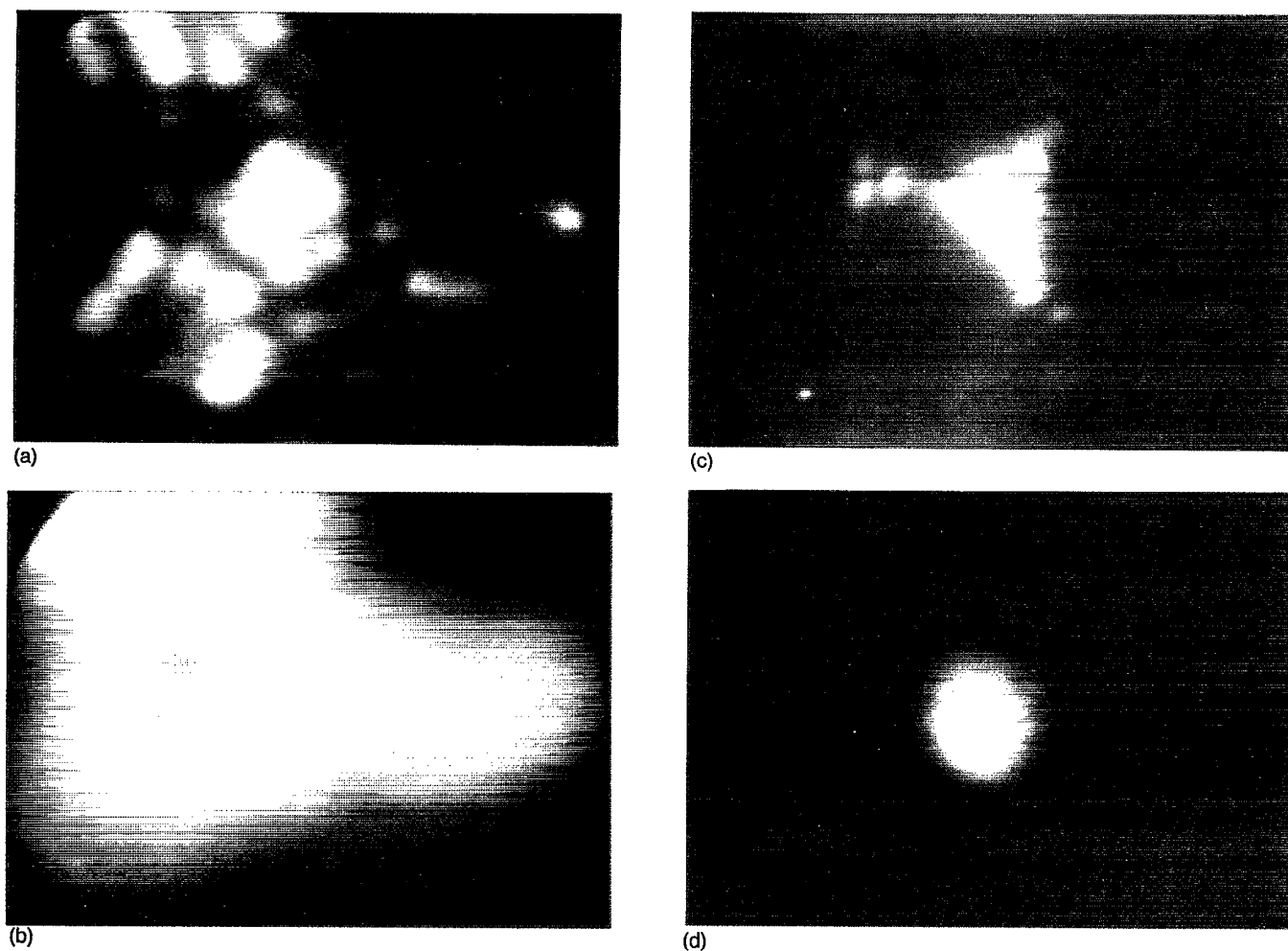


FIG. 3. Dependence of the opening angle of the emitted electron beam upon the mesoscopic form at the tip apex of a $\langle 111 \rangle$ -oriented tungsten tip. (a) Typical FIM image of a round tip (at ≈ 1800 V). (b) Electron emission (FEM) pattern (at ≈ -150 V) from the tip shown in (a). The side peaks towards the $\{112\}$ can clearly be seen. The dark spot near the central maximum is an artifact of the channel plate used here. (c) Typical FIM image of a pyramidal tip (at ≈ 3700 V). (d) Electron emission (FEM) pattern (at ≈ -400 V) from the tip shown in (c). Note that in both cases the microscopic form of the tip apex is the same [trimer forming the (111)]. Both FIM images were taken directly after the preparation of the respective tips shapes. Because of the small dynamics of the channel plate used to record these patterns, the applied potential in (b) and (d) was chosen such as to yield approximately the same current density emitted. In (a) and (c) the potential was chosen slightly above best image field conditions to also make the surroundings of the (111) visible. All images are reproduced using the same scale.

are at best realized by ion sources but not by electron sources.

Furthermore, these theories very often do not consider larger distances. The calculations are performed using a counterelectrode placed only a few \AA in front of the point source, thus avoiding any mesoscopic influences. In order to see why these could influence the focusing of the electron beam, we take a closer look at the round and pyramidal tips.

The main difference between the two is the geometry surrounding the (111) facet. Round tips exhibit a more regular distribution of the electric field on their surface because the overall variation of their local radii of curvature is small. Pyramidal tips, on the other hand, show up an accentuated field maximum at the summit of the pyramid due to its very small local radius of curvature. The sides of the pyramid, however, are formed by flat $\{112\}$ facets so that the field there is strongly attenuated with

respect to the (111) facet. Consequently, the field emission current density is much bigger above the small area forming the summit than over the sides.

For pyramidal tips another fact not taken into account by existing theories might be important: The local work functions Ψ of $W(111)$ and $W(112)$ differ by about 0.5 eV. As, in the case of the multifaceted pyramidal tips, the $\{112\}$ facets are directly adjacent to the (111) facet (which means distances of the order of \AA), the local fields or patch fields^{10,11} are of the order of some tenths of $V/\text{\AA}$. This is the same order of magnitude as that of the field applied from outside. These local fields are therefore no longer small compared to the applied field. Given that $\Psi_{112} > \Psi_{111}$, the effect of these fields is to produce a classical force oriented towards the tip axis. Therefore, a focusing effect is conceivable because the electrons, when passing the very region directly above the surface, still possess low kinetic energy.

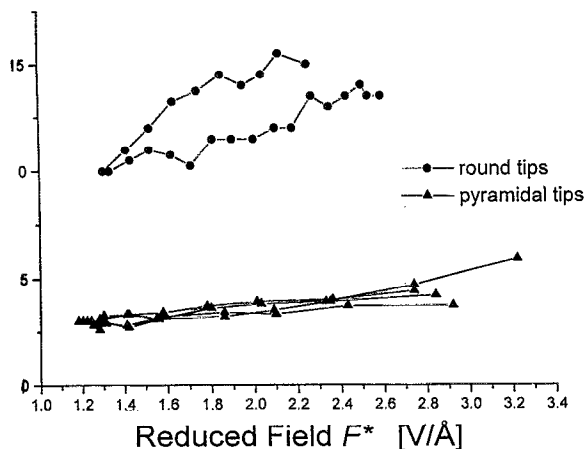


FIG. 4. Opening angle Θ of the emitted electron beam vs reduce field F^* for some typical round and pyramidal tips. Real fields are about $5\times$ smaller. Both round tips are terminated by a monomer. Their r_{FN} radii are 92 and 105 Å, respectively. The applied potentials range from 120 to 270 V, corresponding to a maximum emitted current of 2×10^{-7} A. The pyramidal tips are terminated by 1–3 atoms. Their r_{FN} radii vary from 290 to 305 Å. The applied potentials range from 350 to 950 V, corresponding to a maximum emitted current of 3×10^{-6} A. All tips are used directly after their preparation.

A more sophisticated approach is necessary because these patch fields appear in a “layer” around the tip apex which is of about the size of the (111) facet, i.e., approximately 10 Å. This means that it is still inside the tunnel barrier, the width of which is between 10 and 20 Å.

Instead of a classical trajectory calculation, a correct approach would be to first compute the shape of the tunneling barrier and then the “quantum focusing” in such a potential distribution. An elementary computation made on a truncated cone with a uniform surface dipole density

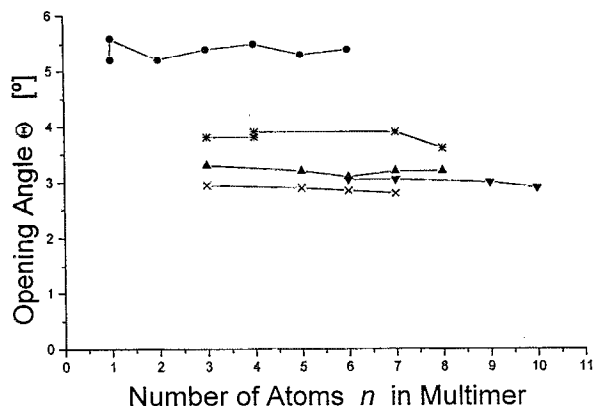


FIG. 5. Opening angle Θ of the emitted electron beam from typical pyramidal tips vs the number n of atoms in the terminating multimer. Curves for four different fields are given with $F^*(\times)=1.28$ V/Å, $F^*(\Delta)=1.42$ V/Å, $F^*(*)=1.69$ V/Å, and $F^*(\bullet)=1.93$ V/Å. Note that directly after their preparation these tips were terminated by ten or more atoms. Then n was reduced by field evaporation, leaving the layer underneath the terminating multimer larger than in the case of the tips used in Figs. 3 and 4 (i.e., the distance from the terminating multimer to the {112} facets is larger). The absolute values of Θ therefore tend to be larger here than in Figs. 3 and 4 because the patch fields are smaller.

on the top plane and a uniform, but different dipole density on the lateral surface, shows that a saddle point in the potential distribution along the tip axis can appear in some cases.¹² Such a saddle point, even if of a completely different origin, was shown to be a crucial point in explaining focusing.⁴

It is worth mentioning that the focusing effect driven by patch fields should obviously decrease when the applied field increases because the patch fields are not dependent on the applied field; this is in agreement with the present observation (Fig. 4).

In the case of round tips, these effects are weaker because of the larger distances between (111) and {112} and the smaller extension of the {112} facets.

It might be noted that the applied fields in FIM are, in general, one order of magnitude larger than in FEM. Patch field effects should therefore be difficult to be seen in FIM.

IV. CONCLUSIONS

We have measured the angular dispersion of a field electron beam emitted from ultrasharp tips terminated by only a few (down to one single) atoms for different atomic arrangements at the tip apex and for various applied fields and currents.

It is found that the opening angle Θ is strongly dependent upon mesoscopic geometrical factors and that pyramidal tips predominantly emit focused electron beams. For such tips the microscopic geometry, like the size of the emission area (ranging from one to ten atoms), seems not to be of importance.

The opening angle of the beam increases from 3° to 5° when the field is increased by a factor of 3 (which corresponds roughly to ten orders of magnitude concerning the variation of the emission current).

Existing theories are mostly just one or two dimensional, thus disregarding the complex three-dimensional conditions of real tips with atomic size emission areas. They are at the moment not very helpful.

We therefore feel the need for more elaborate calculations based upon more realistic assumptions of tunneling, for instance, in the presence of patch fields.

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