Electron tunneling time measurement by field-emission microscopy

S. K. Sekatskii[1,2] and V. S. Letokhov[1,*]
[1]*Institute of Spectroscopy Russian Academy of Sciences, Troitsk, Moscow, 142190 Russia*
[2]*Institute de Physique de la Matière Condensée, Université de Lausanne, BSP, CH-1015 Lausanne-Dorigny, Switzerland*
(Received 5 June 2001; published 27 November 2001)

Electron-tunneling time has been measured using field-emission microscopy (FEM). The analysis of the FEM images of the dopant samarium ions located inside the calcium fluoride coating onto the silicon nanotip gives the value of the perpendicular momentum distribution of emitted electrons. This distribution is a natural measure of the tunneling time: the more time an electron spends under the barrier, the narrower such a distribution is (Larmor clock experiment). For the barrier height of 1.7 eV and electric-field strength ranging from 0.55 to 0.7 V/nm, the tunneling time ranges from 6 to 8 fs.

DOI: 10.1103/PhysRevB.64.233311 PACS number(s): 73.40.Gk, 03.65.Xp, 79.70.+q

The problem of tunneling time is one of the oldest, well known, and controversial in quantum mechanics.[1–3] Probably the best approach for the experimental measurement of tunneling time was formulated long ago by Rybachenko,[4] who modified the idea of Baz'[5] of how to measure the collision time. He proposed to use some additional degree of freedom, changing during the tunneling, to track the particle motion under the barrier (Larmor clock experiment; spin precession due to the magnetic field acting only in the barrier region was considered). Comparing the values of parameters characterizing this degree of freedom before and after the tunneling, one can directly measure the tunneling time (see Fig. 1).

Such an experiment was never performed (although somewhat similar experiments of tunneling of electrons in heterostructures in the presence of magnetic field were reported[6,7]). Moreover, it was shown that Larmor precession is not the main source of the apparent spin rotation for this and other approaches, and the results of such experiments would be difficult to interpret.[8–10] Thus ''Larmor clocks'' should be invented to measure the electron-tunneling time. Recently it was indicated that, due to the three-dimensional character of the tunneling during the field ionization of a particle in the uniform electric field, the perpendicular momentum distribution of the tunneling particle can be used as such a clock.[11] Here we report the experimental realization of such a possibility.

An idea of the method is illustrated in Figs. 1(c) and 2. Let us first consider the tunneling process as taking place for an ensemble of electrons. Each of them tunnels from the spherically symmetric quantum well in a uniform radial electric field F, and in the quasiclassic approximation, usually used to discuss the tunneling, it can be characterized by the energy E and momentum components with the initial values of $p_{z,0}$ (in the direction of field) and $p_{\perp,0}$ (in a transversal direction). When tunneling under the barrier, no force is acting in the transversal direction, and thus the transversal momentum also does not change. But the ''trajectory'' of the tunneling particle depends on the $p_{\perp,0}$ (see Fig. 2). The larger the initial transversal momentum, the longer the distance the electron should travel under the barrier and thus the smaller will be the tunneling probability. (Here we exploit the concept of the ''virtual'' trajectory in a classically forbidden area introduced by Kapur and Peierls[12] by means of substituting an imaginary time $i\tau$ instead of real time t.) Formally this corresponds to the suppression of the initial $p_{\perp,0}$ distribution: after the tunneling, a much narrower $p_\perp$ distribution will appear.

This suppression is obviously related to the fundamental characteristics of the tunneling process, and it can be explained using the concept of the tunneling time as follows. Let us now consider the tunneling as taking place with only one particle characterized (in a momentum space) by the wave function $\psi(p_z, p_\perp)$ of two independent parameters. Because the electric field has only a z component, the tunneling process is defined only by the normal momentum component p_z, while $p_\perp$ could be treated as an independent degree of freedom. The average value of $p_\perp$, however, evolves during

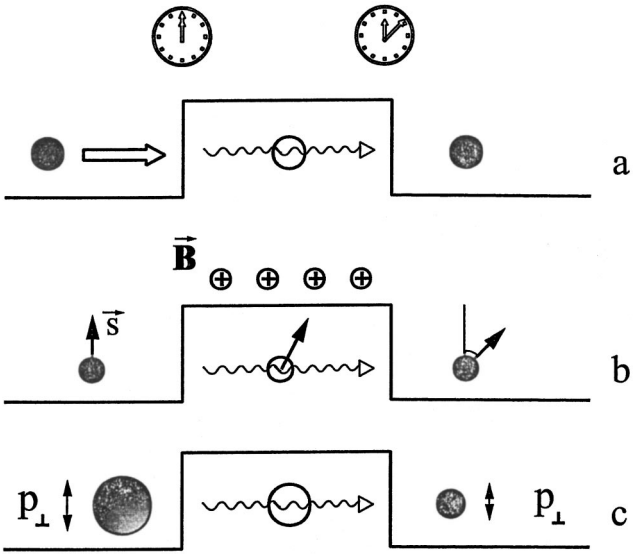

FIG. 1. The idea of the Larmor clock tunneling time measurement. One should invent a clock that starts to measure at the beginning of the barrier crossing and finishes at the end (a). Particle spin vector s rotation in the perpendicular magnetic field **B**, superimposed in the barrier region (b), or transversal momentum suppression (c) can be used as such a clock.

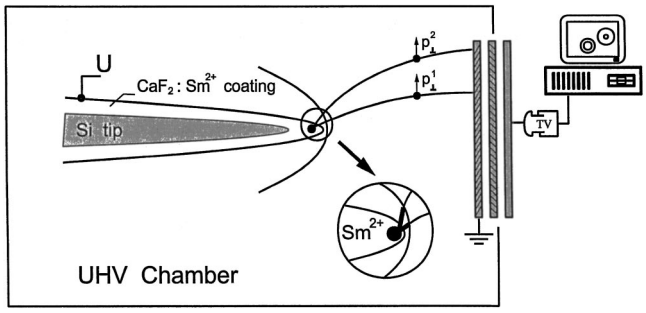

FIG. 2. The laser photoelectron projection microscope (out of the scale; two electron trajectories corresponding to the different transversal momenta $p_\perp$ values are shown). Sharp tips are installed inside the UHV chamber and high-resolution field-emission images of these tips formed when the potential U ranging from 0 to 4 kV was applied were detected using the microchannel plate-fluorescence screen assembly placed at a distance of $L=10$ cm from the tip. The optical image formed at the exit of the assembly was picked up by a television camera operated in conjunction with a Model Argus-50 image processing computer system (Hamamatsu Photonics K. K., Japan). The same experimental setup has been used by us earlier to vizualize single color centers on the $LiF:F_2$ crystal tip surface (Ref. 17) and as a first femtosecond superresolution projection photoelectron microscope. The dotted line indicates the edge of the barrier (the beginning of the classically accessible area); the difference of the lengths of the particle trajectories under the barrier is clarified in the inset.

the tunneling (and stops evolving after it), which enables to measure the tunneling time as a ''stop watch,'' or Larmor clock. The proportionality coefficient between the tunneling time T and the measured transversal momentum spread should be found from the full quantum-mechanical treatment of the problem. Such a treatment was given using the exact quantum-mechanical Green function[11] available for the Schroedinger equation describing the quantum well represented by the contact Fermi potential (pseudopotential) $-V\delta(r)(\partial/\partial r)$ placed in a uniform electric field.[13,14] It also has been discussed that such a model satisfactorily describes a real field-emission experiment.[11,13–15] The corresponding result reads

$$T=\frac{\hbar m}{2\langle p_\perp^2\rangle}=\frac{\hbar}{4E_\perp}, \tag{1}$$

where m is an electron mass and $\hbar$ is the Planck constant, $E_\perp=p_\perp^2/2m$.

In the experiments described we measured the transversal momentum spread of the tunneling electrons using the field-emission/ion microscope shown schematically in Fig. 2. From the consideration of the classical equations of motion of an emitted electron in the electric field of a projection microscope, it is known that when the emitted electron possesses the kinetic energy $E_\perp$ in the transversal direction (see Fig. 2), in the detector plane its image will be shifted to the distance of $l=2L\sqrt{E_\perp/eU}$ from the symmetry axis.[16] In particular, this defines the spatial resolution of the field-emission microscope: a pointlike object on the tip surface that emits electrons with the mean kinetic transversal energy of $E_\perp$ will be seen on the detector plane as corresponding to the spot with the diameter δ on the tip surface:

$$\delta=4r\gamma\sqrt{E_\perp/eU}. \tag{2}$$

Here r is the radius of the curvature of the tip and γ is a numerical coefficient ranging from 1.5 to 2 and appearing because of the difference between the geometry of a real FEM and an ideal case of a spherical capacitor. The same coefficient appears in the formula describing the magnification M of the FEM:[16]

$$M=L/\gamma r, \tag{3}$$

and for our microscope it was calibrated using the samples with the well-defined structure (observation of tungsten tips with one-atom resolution, etc.) as equal to $\gamma=1.5$.[17]

Thus, measuring with the help of the field-emission microscope the transversal momentum spread of the electrons emitted by an isolated center of the tip, one can measure the electron-tunneling time related with the field ionization of this center. It should be noted, however, that usual field-emission experiments performed with metal or semiconductor tips do not deal with the single isolated centers to be field ionized. Instead of this, these experiments are described as the local variations of the work function due to the presence of different crystallographical planes or impurities on the tip surface, and the spatial resolution of the FEM is governed by the statistical distribution of the electron-gas energies inside the metal.[16]

We proposed that ultrasharp silicon tips (radius of a curvature 10–20 nm) coated with 50–100-nm thick $CaF_2:Sm^{2+}$ layers could be used as such necessary field-emission sources, for which tunneling current will be due to the field ionization of single isolated bivalent samarium dopant ions. High-conductivity n-type silicon tips were fabricated in the Institute of Crystallography Russian Academy of Sciences, Moscow, using a vapor-liquid-solid growth technique with the subsequent sharpening procedure.[18] CaF_2 coatings were grown on Si tips by a molecular-beam epitaxy technique in the A. F. Ioffe Institute of Physics and Technology Russian Academy of Sciences, Sanct-Petersburg; CaF_2 containing samarium in a necessary concentration had been evaporated from a Knudsen cell.[19]

These samples were selected by us for the tunneling time measurements for the following two reasons:

(i) The ionization threshold I for the bivalent samarium ions in a calcium fluoride matrix is known to be small. Experiments devoted to the internal[20] and external[21] photoelectric effect for $CaF_2:Sm^{2+}$ crystals give the value $I=1.7$ eV, which is much smaller than the work function characterizing the tip itself—silicon and calcium fluoride. Thus one can hope that the field ionization of the dopant ions will be achieved at lower fields and can be distinguished against the field emission from the matrix.

This was confirmed in the first series of experiments performed with the silicon tips coated by the rather thin (a few nanometers thick) $CaF_2:Sm^{2+}$ layers: at low tip potentials it was shown that the field emission observed is due to the resonant tunneling process via the dopant samarium ions.[22] Similar conclusions have been made in Ref. 23 where analo-

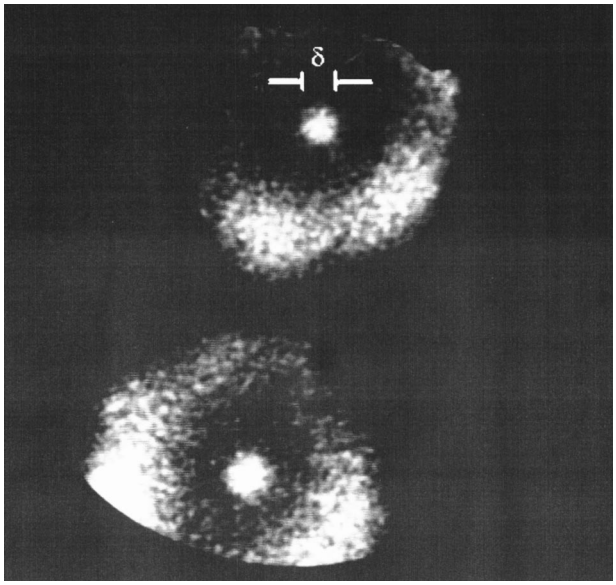

FIG. 3. Field-emission image of the $Si/CaF_2:Sm^{2+}$ tip. Radius of the curvature of the tip $r=70$ nm (SEM data), $CaF_2:Sm^{2+}$ coating thickness—50 nm, and Sm^{2+} dopant ions concentration—0.02 mol %. Tip potential $U=1.9$ kV. The full diameter of the image is equal to 35 nm.

gous ultrasharp silicon nanotips coated by thin surface oxide layers were studied. The measurements of the emitted electron energy showed that all emitted electrons are due to the surface oxide and/or interface states while the direct emission coming from the silicon conduction band cannot be observed.

(ii) An observation of the field ionization of the dopant samarium ions should be facilitated by the extremely high transparency of the calcium fluoride matrix for low-energy electrons (the photoelectron escape depth of 260 nm was reported[24]) and an atomic perfection of the Si/CaF_2 interface.[19,25] These circumstances could be important to ensure the efficient recombination of the field-ionized dopant ions due to the passage of electrons from the silicon conductive core.

A typical field-emission image of one of the $Si/CaF_2:Sm^{2+}$ tips is presented in Fig. 3, where two double-ring structures (each composed from a central spot and a ring) are clearly seen. These structures could be observed in a rather narrow range of the voltage applied (in a low-current regime, from ~1.8 to ~2.5 kV): the emission current was unmeasurable at lower potentials, while at higher potentials it was not possible to discern similar structures against intense quasiuniform background due to the field emission from the silicone core or/and CaF_2 valence zone. Similar double-ring structures were also never observed for the cases of both pure uncoated silicon tips and Si/CaF_2 tips that contain no samarium dopant ions, and were attributed to the field ionization of dopant bivalent samarium ions; thus two different single Sm ions are imaged in Fig. 3. (It is worthwhile to note that the typical FEM image caused by some kind of a microprotrusion on the tip surface looks like a structureless and rather broad spot in striking contrast with the data presented in Fig. 3.)

The presence of two rings was recently explained by us[15] as a direct visualization of two neighboring quantum levels of Sm^{2+} ions in CaF_2 matrices: $^7F_0$ and $^7F_1$ characterized by the total angular moments $J=0$ and $J=1$, respectively. These levels result in a bimodal distribution of the transversal momentum, which is imaged as a double-ring structure. In this paper we will concentrate on the central ring (image of the spherically symmetric $^7F_0$ state of the dopant ion), for which an elaborate tunneling theory exists and whose width gives us a tunneling time.

The experimental measurements of δ by processing the images of the central rings in Fig. 3 and analogous images [the formula (3) and scanning electron microscopy (SEM)-

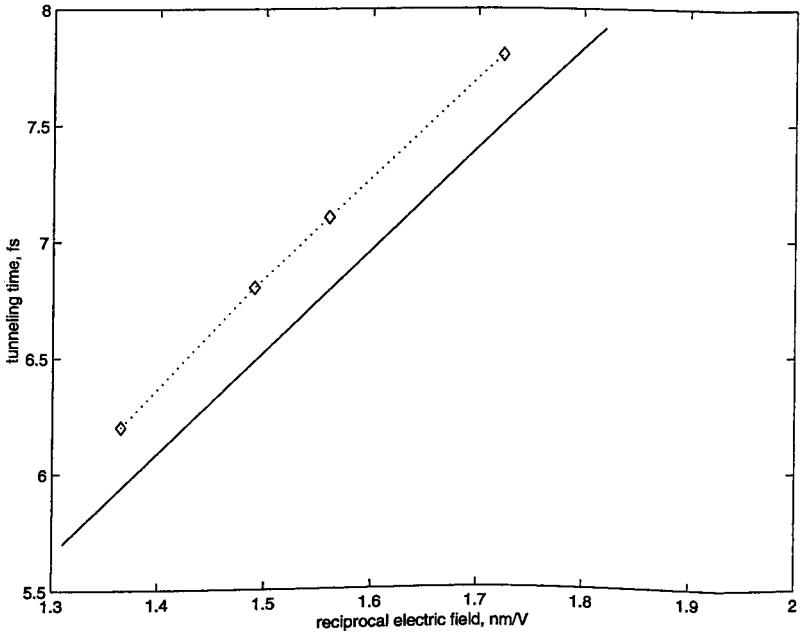

FIG. 4. The dependence of the measured tunneling time on the reciprocal electric-field strength (diamonds). The theoretical dependence [Eq. (5)] is shown by the solid line.

measured radius of curvature of the tip were used] give the value $\delta = 1.4 \pm 0.05$ nm (for $U = 1.9$ kV; this is the result of an averaging for both Sm ions seen in Fig. 3). Expressing the mean transversal kinetic energy $E_\perp$ from Eq. (2) as a function of δ (for our case $E_\perp = 0.021$ eV) and substituting the result into Eq. (1) we can write the following relation to determine the tunneling time:

$$T = \frac{\hbar}{eU}\left(\frac{2r\gamma}{\delta}\right)^2, \tag{4a}$$

which can be further simplified using Eq. (3) to express the time as a function of the mean diameter of the central spot observed on the detector d,

$$T = \frac{\hbar}{eU}\left(\frac{2L}{d}\right)^2. \tag{4b}$$

Only the easy-to-measure experimental parameters are contained in Eq. (4b), which gives the tunneling time value $T = 7.8$ fs.

It is interesting to compare this value with the calculated value of barrier crossing time, which can be obtained for the model used[11,13,14] (the attractive contact Fermi potential of the height I and the triangular barrier with the thickness $t = I/F$, where F is the electric-field strength):

$$T = \frac{\sqrt{2mI}}{eF}. \tag{5}$$

The electric field acting on the dopant atom for the emitter used can be calculated as follows:[26] $F = r_0 U/k(\varepsilon r_0 + h)(r_0 + h)$. Here k is a numerical coefficient describing the electric field of the uncoated conducting tips[16] calibrated by us as equal to 5,[17] and r_0 is the radius of curvature of the conductive silicon ''core'' coated with the layer of calcium fluoride with the dielectric constant $\varepsilon = 6.8$ and thickness $h = 50$ nm; thus $r_0 = r - h = 20$ nm. For the case in hand we have $F = 0.854$ V/nm, and substituting this value in Eq. (5) we obtain the value $T = 7.5$ fs, which is close to the experimental data given above.

Thus, the direct measurement of the electron-tunneling time (Larmor clock experiment) has been performed using a field-emission method. The values received in the paper as well as the dependence of the *measured* tunneling time on the reciprocal electric-field strength (see Fig. 4) are relatively well described by the existing tunneling theories. The small systematic deviation is due to the imprecise calibration of the field (and also may be due to the difference between the free-electron mass and effective electron mass inside the calcium fluoride coating) and can be eliminated by adjusting the field value. Nevertheless we believe that the results obtained are important for better understanding the tunneling time problem, and hope that further refinements of the measurements could lead to improvements in the theory.

The authors thank V. N. Konopsky for assistance during the beginning of these experiments, V. V. Zhirnov and E. I. Givargizov for the submission of silicon nanotips, N. S. Sokolov and J. C. Alvarez for the preparation of calcium fluoride coatings, and Hamamatsu Photonics, Japan, for lending us the experimental equipment. This work was financially supported by the Russian and Swiss Funds of Fundamental Research and the Department of Defense of the United States of America.

*Also at the University of Lund, Sweden.

[1] L. A. MacColl, Phys. Rev. **40**, 621 (1932).

[2] E. H. Hauge and J. A. Stovneng, Rev. Mod. Phys. **61**, 917 (1989).

[3] R. Landauer and T. Martin, Rev. Mod. Phys. **66**, 217 (1994).

[4] V. F. Rybachenko, Yad. Fiz. **5**, 895 (1967) [Sov. J. Nucl. Phys. **5**, 635 (1967)].

[5] A. J. Baz', Yad. Fiz. **5**, 229 (1967) [Sov. J. Nucl. Phys. **5**, 161 (1967)].

[6] P. Guéret, A. Baratoff, and E. Marclay, Europhys. Lett. **3**, 367 (1987).

[7] P. N. Roy, A. P. Singh, and B. P. Singh, Indian J. Phys., A **66A**, 629 (1992).

[8] M. Büttiker, Phys. Rev. B **27**, 6178 (1983).

[9] J. P. Falck and E. H. Hauge, Phys. Rev. B **38**, 3287 (1988).

[10] M. Deutsch and J. E. Golub, Phys. Rev. A **53**, 434 (1996).

[11] C. Bracher, W. Becker, S. A. Gurvitz, M. Kleber, and M. S. Marinov, Am. J. Phys. **66**, 38 (1998).

[12] P. L. Kapur and R. Peierls, Proc. R. Soc. London, Ser. A **163**, 606 (1937).

[13] B. Gottlieb, M. Kleber, and J. Krause, Z. Phys. A: Hadrons Nucl. **339**, 201 (1991).

[14] Yu. N. Demkov and V. N. Ostrovsky, *Zero-Range Potentials and Their Applications in Atomic Physics* (Plenum, New York, 1988).

[15] S. K. Sekatskii, Surf. Sci. **453**, L303 (2000).

[16] R. Gomer, *Field Emission and Field Ionization* (American Institute of Physics, New York, 1993).

[17] V. N. Konopsky, S. K. Sekatskii, and V. S. Letokhov, Opt. Commun. **132**, 251 (1996).

[18] E. I. Givargizov, J. Vac. Sci. Technol. B **11**, 449 (1993).

[19] N. S. Sokolov, J. C. Alvarez, and N. L. Yakovlev, Appl. Surf. Sci. **60/61**, 421 (1992).

[20] C. Pedrini, F. Rogemond, and D. C. McClure, J. Appl. Phys. **59**, 1196 (1986).

[21] S. K. Sekatskii, V. S. Letokhov, and S. B. Mirov, Opt. Commun. **95**, 260 (1993).

[22] V. N. Konopsky, S. K. Sekatskii, and V. S. Letokhov, J. Phys. IV **6-C5**, 125 (1996).

[23] J. Shaw, J. Vac. Sci. Technol. B **18**, 1817 (2000).

[24] B. Quiniou, W. Schwarz, Z. Wu, R. M. Osgood, Q. Yang, and J. M. Phillips, Appl. Phys. Lett. **60**, 183 (1992).

[25] S. K. Sekatskii, D. V. Serebryakov, V. S. Letokhov, V. V. Zhirnov, A. L. Meshcheryakova, J. C. Alvarez, and N. S. Sokolov, Appl. Surf. Sci. **140**, 46 (1999).

[26] A. L. Pregenzer, K. W. Bieg, J. E. Olson, and J. A. Panitz, J. Appl. Phys. **67**, 7556 (1990).