Calculation of quantum tunneling for a spatially extended defect: the dislocation kink in copper has a low effective mass.

Tejs Vegge[*,†], James P. Sethna[‡], Siew-Ann Cheong[‡], K. W. Jacobsen[*], Christopher R. Myers[¶], Daniel C. Ralph[‡],
[*] Center for Atomic Scale Materials Physics (CAMP) and Department of Physics,
Building 307, Technical University of Denmark, DK-2800, Kgs. Lyngby, Denmark
[†]Materials Research Department, Risø National Laboratory , DK-4000 Roskilde, Denmark
[‡]Laboratory of Atomic and Solid State Physics (LASSP), Clark Hall,Cornell University, Ithaca, NY 14853-2501, USA
[¶]Cornell Theory Center, Cornell University, Ithaca, NY 14853, USA

Several experiments indicate that there are atomic tunneling defects in plastically deformed metals. How this is possible has not been clear, given the large mass of the metal atoms. Using a classical molecular-dynamics calculation, we determine the structures, energy barriers, effective masses, and quantum tunneling rates for dislocation kinks and jogs in copper screw dislocations. We find that jogs are unlikely to tunnel, but the kinks should have large quantum fluctuations. The kink motion involves hundreds of atoms each shifting a tiny amount, leading to a small effective mass and tunneling barrier.

Tunneling of atoms is unusual. At root, the reason atoms don't tunnel is that their tunneling barriers and distances are set by the much lighter electrons. The tunneling of a proton over a barrier one Rydberg high and one Bohr radius wide is suppressed by the exponential of $\sqrt{2 M_p R_y a_0^2} = \sqrt{M_p/m_e} \sim 42.85$: a factor of 10^{-19}.

The atomic tunneling which dominates the low-temperature properties of glasses [1] occurs in rare regions [few parts per million] where an atom or group of atoms has a double-well potential with a low barrier and asymmetry. Tunneling defects in crystals [2] involve either nearly spherical molecules or weakly off-center defects with low barriers and tunneling distances. Quantitative modeling of these spatially localized tunneling defects has been frustrated by the demands for extremely accurate estimates of energy barriers, beyond even the best density functional electronic structure calculations available today.

There is much evidence that quantum tunneling is important to the properties of undoped, plastically deformed metals. Quantum creep [3], glassy low-temperature behavior [4], and two-channel Kondo scaling seen in the voltage and temperature-dependent electrical conductivity in nanoconstrictions [5] have been attributed to quantum tunneling associated with dislocations. It has never been clear how this can occur, given the large masses of the metal atoms involved.

We show here using a classical effective-medium interatomic potential that quantum fluctuations can indeed be important in the dynamics of one particular defect: a kink in a split-core dislocation — in this case a screw dislocation in copper. The motion of the kink involves a concerted motion of hundreds of copper atoms, leading to a dramatic decrease in its effective mass [6]. Because our important conclusions rest upon this delocalization they are qualitatively much less sensitive to the accuracy of our potential than calculations for spatially localized tunneling defects. We assert that these kinks are likely the only candidate for quantum tunneling in pure fcc metals.

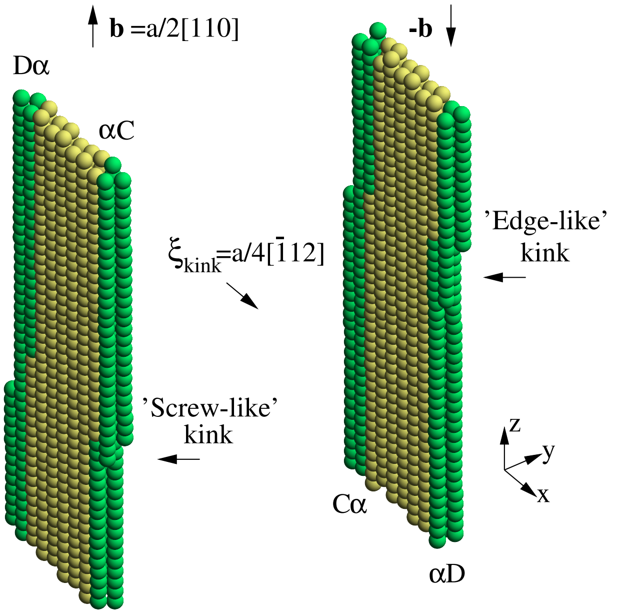

FIG. 1. **Kink geometry.** Broadly speaking, the screw dislocation represents the locus where planes of atoms form a helix. In copper it spreads out into a ribbon along the x-axis to lower its energy. The kink we study shifts the ribbon by one atom in the x direction. More specifically, the $\mathbf{b}=\frac{a}{2}[110]$ dislocation on the left dissociates on the $(1\bar{1}1)$ plane into the Shockley partials: $\alpha\mathrm{C} = \frac{a}{6}[121]$ and $\mathrm{D}\alpha = \frac{a}{6}[21\bar{1}]$ respectively. The kink is introduced with line vector $\xi_{\mathrm{kink}} = \frac{a}{4}[\bar{1}12]$, and dissociates into a wide screw-like- and a bulky edge-like kink located on the partial dislocations. The lighter atoms are on the stacking fault (hcp local environments) and the darker atoms are along the partial dislocations (neither hcp nor fcc).

Quantum creep estimates [3] of double-kink nucleation will change significantly using our low kink effective mass.

The glassy low-temperature properties [4] of deformed metals likely arise from kinks tunneling between pinning sites. (Dislocation-rich metals are not glasses, and glasses don't have kinks, but there has been speculation [7] that tunneling in glasses is collective.) We don't have an explicit model for the scaling seen in nanoconstrictions [5], but we do claim that kinks are the only active atomic degrees of freedom.

The kink simulation consists of two screw dislocations with opposite Burgers vectors $\mathbf{b}=\pm\frac{a}{2}[110]$, allowing periodic boundary conditions giving us the perfect translational invariance necessary to measure energy differences to the accuracy we need. The two dislocations are placed in different $(1\bar{1}1)$ planes separated by 20 $(1\bar{1}1)$ planes (4.4 nm), see figure 1. The system is 86 planes wide (19.3 nm) in the two (non-orthogonal) directions, and extends 44.5 $\mathbf{b}$ (11.4 nm) along the dislocations. We introduce kinks or jogs on the dislocations by applying skew periodic boundary conditions to system, *i.e.* we introduce a small mismatch in the dislocation cores at the interface to the next cell. The procedure also introduces a row of interstitial atoms between the kinked dislocations, which is subsequently removed from the system, leaving us with a total of 329 102 atoms. The kinks have a net line vector of $\xi_{\text{kink}} = \frac{a}{4}[\bar{1}12]$, with a the lattice constant.

To show how unusual the properties of the kink are, we also study the properties of a dislocation jog. The jog simulation, and the associated energy barrier calculation, is similar and is described elsewhere [8]. The elementary jog we study is introduced with a line vector oriented in the $(\bar{1}11)$ glide plane of the screw dislocation, $\xi_{\text{jog}} = \frac{a}{4}[1\bar{1}2]$, which then transforms into an obtuse lower energy configuration: $\frac{a}{4}[1\bar{1}2] \rightarrow \frac{a}{2}[101] + \frac{a}{4}[\bar{1}\bar{1}0]$. This jog is expected to be the most mobile of the jogs, second only to the kink in mobility.

We introduce the two kinked dislocations directly as Shockley partial dislocations, see figure 1, and relax using the MDmin algorithm [9], using Effective Medium Theory (EMT): a many-body classical potential [10], which is computationally almost as fast as a pair potential, while still describing the elastic properties well. The elastic constants of the potential are: $C_{11} = 176.2$ GPa, $C_{12} = 116.0$ GPa and $C_{44} = 90.6$ GPa with a Voigt average shear modulus of $\mu = 66$ GPa, and an intrinsic stacking fault energy of $\gamma_{\text{I}} = 31$ mJ/m^2.

We present three quantities for the kink and jog: the Peierls-like barrier for migration along the dislocation, the effective mass, and an upper bound for the WKB factor suppressing quantum tunneling through that barrier. Since the motion of these defects involves several atoms moving in a coordinated fashion, we use instantons: the appropriate generalization of WKB analysis to many-dimensional configuration spaces [11,12,6]. An upper bound for the tunneling matrix element is given by the effective mass approximation [12,13],

$$\Delta \leq \hbar\omega_0 \exp\left(-\int \sqrt{2M^*(Q)V^*(Q)}dQ/\hbar\right), \qquad (1)$$

where ω_0 is an attempt frequency, $V^*(Q)$ is the energy of the defect at position Q with the neighbors in their relaxed, minimum energy positions $q_i(Q)$, and

$$M^*(Q) = \sum_i M_i(dq_i/dQ)^2 \qquad (2)$$

is the effective mass of the defect incorporating the kinetic energy of the surrounding atoms as they respond adiabatically to its motion. The effective mass approximation is usually excellent for atomic tunneling. The method is variational, so equations 1 and 2 remain upper bounds using other assumptions about the tunneling path $q_i(Q)$ (such as the straight-line path between the two minima described below for the kinks).

The difficulty of finding models for atomic tunneling is illustrated rather well by the properties of the jog we study. The barrier for migration was determined to be 15 meV [8]: lower than other jogs, or even than surface diffusion barriers calculated with the same potential. The effective mass for the jog, estimated by summing the squared displacement of the 200 atoms with largest motion, is $M^*_{\text{jog}} \simeq 0.36\ M_{\text{Cu}}$: the jog is spatially localized (it doesn't dissociate into partials), with a few atoms in the core of the jog carrying most of the motion. The WKB tunneling matrix element for the jog to tunnel a distance $Q = 2.5$ Å over a barrier $V = 0.015$ eV is suppressed by a factor of roughly $\exp(-\sqrt{2M^*_{\text{jog}}V}Q/\hbar) \simeq 10^{-14}$. Jogs don't tunnel much.

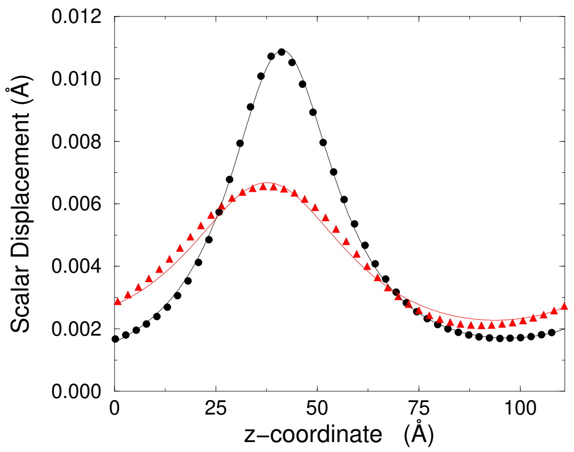

FIG. 2. The magnitude of the atomic displacement field as the kinks move on the partials, along the cores of the two partial dislocations, as a function of the z-coordinate. The data points are fitted to 'periodic' Lorentzian distributions. The full-width at half maximum of the two partial kinks are $\Gamma_{\text{edge}} = 13\mathbf{b}$ (bullets) and $\Gamma_{\text{screw}} = 21\mathbf{b}$ (triangles). The center of masses of the two partial kinks are separated: the screw-like partial kink is shifted by $\delta z = -1.5\mathbf{b}$.

For the kinks, we take a relaxed initial configuration and define a final configuration with the kink migrated by one lattice spacing along the dislocation. The final position for each atom is given by the position of the neighboring atom closest to the current position minus the kink migration vector $l_{\rm migr.} = \frac{a}{2}[110]$ which represents the net motion of the kink. This automatically gives the correct relaxed final position, which is otherwise difficult to locate given the extremely small barriers. The width of the kinks is the traditional name for their extent along the axis of the screw dislocation. We can measure this width by looking at the net displacement of atoms between the initial and final configurations. We find that the displacement field is localized into two partial kinks, localized on the partial dislocation cores, see figure 2. These two partial kinks are quite wide (13 **b** and 21 **b**). They differ because the partials are of mixed edge and screw character; it is known [14] that the kink which forces a mixed dislocation towards the screw direction will be wider and have higher energy. This is wider than the $w \leq 10\mathbf{b}$ predicted for slip dislocations in closed-packed materials by Hirth and Lothe, and Seeger and Schiller using line tension models [15]. The kinks will merge together for kink densities above one per kink width, or dislocation radius of curvature smaller than the width squared over **b** (say, $< 300\mathbf{b}$), validating the traditional description of continuously curved dislocations.

Notice that the maximum net distance moved by an atom during the kink motion in figure 2 is around 0.01 Å. Summing the squares of all the atomic motions, and using equation 2, we find an effective mass $M^*_{\rm kink} \simeq M_{\rm Cu}/130$ within the straight-line path approximation. This remarkably small mass can be attributed to three factors. (1) The mass is decreased because the screw dislocation is split into two partial dislocations [16]. (2) The cores of the partial dislocations are spread transversally among $W_T \simeq 4$ planes of atoms, figure 3; this factor seems to have been missed in continuum treatments. These first two factors each reduce the total distance moved by an atom as the kink passes from $z = -\infty$ to $+\infty$. (3) The kinks on the partials average $W_L \simeq 17$ planes wide (above), so the total atomic motion is spread between around 17 kink migration hops [17]. Thus when the kink moves by x, the atoms in two regions $1/W_L$ long and $1/W_T$ wide each move by $x/(2W_L W_T)$, reducing [17] the effective mass by roughly a factor of $2W_L W_T \sim 136$.

Evaluating the energy at equally spaced atomic configurations and linearly interpolating between the initial and final states (along the straight-line path) yields an upper bound to the kink-migration barrier of 0.15 μeV, figure 4. We attribute this extremely small barrier to the wide kinks on the partials: for wide kinks the barrier V should scale exponentially with the ratio of the kink width to the interplanar distance: $V \propto \exp(-W_L)$ [18].

This small barrier is not only negligible for thermal activation (two mK), but also for quantum tunneling. The WKB factor suppressing the tunneling would be $\exp(-\sqrt{2M^*_{\rm kink}V}Q/\hbar) = \exp(-0.0148) = 0.985$. Even at zero temperature, the kinks effectively act as free particles, as suggested in the literature ([14], among others).

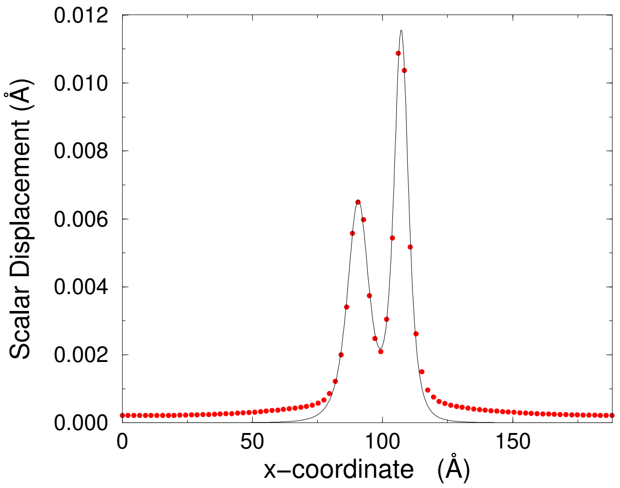

FIG. 3. The magnitude of the atomic displacement field as the two partial kinks move, along the cores of the two partial dislocations, as a function of the x-coordinate. The core regions are fitted to two squared Lorentzian distributions. The partial core widths $W_T \simeq 4d$ (4 $(1\bar{1}1)$ lattice planes), significantly reduces the effective mass of the kink.

Our estimated kink migration barrier is thus 10^5 times smaller than that for the most mobile of the jogs. How much can we trust our calculation of this remarkably small barrier? Schottky [19] estimates using a simple line-tension model that the barrier would be $\sim 3\ 10^{-5}$ eV in fcc materials, using a Peierls stress $\sigma_{\rm P} = 10^{-2}\mu$ and a kink width $w = 10$ **b**. This value is a factor of 200 higher than the barrier we find. On the other hand, both experiments and theoretical estimates predict $\sigma_{\rm P} \simeq 5\ 10^{-6}\mu$ for Cu [18], yielding barriers orders of magnitude lower than ours. Using a classical potential is valid for our purposes: electronic quantum fluctuations are well treated in the Born-Oppenheimer approximation, and atomic quantum fluctuations are explicitly treated in the instanton approximation. The interatomic potentials we use do not take into account directional bonding. This is usually a good approximation for noble metals; however, small contributions from angular forces may change the kink width. The kink width is like an energy barrier, balancing different competing energies against one another: in analogy, we expect it to be accurate to within twenty or thirty percent. Our small value for the effective mass, dependent on the inverse cube of the spatial extent of the kink, is probably correct within a factor of two. The energy barrier is much more sensitive: if we take the total exponential suppression to be 10^5 (using the jog as a "zero-length" defect) then each 20% change in the width would yield a factor of 10 change in the barrier height.

The qualitative result of our calculation, that the barriers and effective masses are small, are robust not only to the use of an approximate classical potential, but may also apply to other noble metals and perhaps simple and late transition metals.

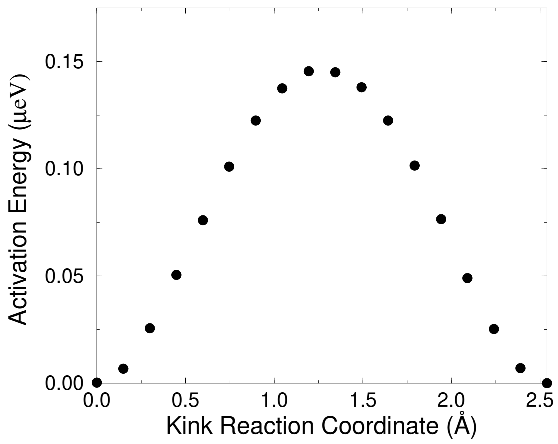

FIG. 4. The activation energy as a function of the straight-line distance moved by the kink on one dislocation, with an associated barrier of $E_{\mathrm{act}} = 0.15$ μeV. Notice that this activation energy is about one part in 10^{13} of the total system energy.

In summary, we have used an atomistic calculation with classical potentials to extract energy barriers and effective masses for the quantum tunneling of dislocation jogs and kinks in copper. For jogs, the atomic displacements during tunneling are primarily localized to a few atoms near the jog core, each moving a significant fraction of a lattice spacing. Consequently, the tunneling barrier and effective mass are relatively large, and tunneling is unlikely. However, the kinks in screw dislocations are much more extended: as a kink moves by one lattice spacing, hundreds of atoms shift their positions by less than 1% of a lattice spacing. Both the energy barrier and the effective mass are reduced, to the extent that tunneling should occur readily. Kinks are likely the only candidate for quantum tunneling in pure crystalline materials. They may explain measurements of quantum creep, glassy internal friction, and non-magnetic Kondo effects seen in plastically deformed metals.

Acknowledgments. This work was supported by the Digital Material project NSF #9873214 and the Danish Research Councils through Grant #9501775 and the Center for Structural Characterization and Modelling of Materials in Risø. The Center for Atomic-scale Materials Physics (CAMP) is sponsored by the Danish National Research Foundation. An equipment grant from Intel and the support of the Cornell Theory Center, including through grant NSF #EIA-9972853, are also gratefully acknowledged. We had helpful conversations with Nicholas Bailey and Jakob Schiøtz.

---

[1] R.C. Zeller and R.O. Pohl, *Phys. Rev. B* **4**, 2029 (1971).

[2] V. Narayanamurti and R.O. Pohl, *Rev. Mod. Phys.* **42**, 201 (1970).

[3] B.V. Petukhov and V.L. Pokrovskii, JETP Lett. **15**, 44 (1972); V.V. Pustovalov, Fiz. Nizk. Temp. **15**, 901 (1989), translated in Sov. J. Low Temp. Phys.; A. Hikata and C. Elbaum, *Phys. Rev. Lett.* **54**, 2418 (1985).

[4] T. Kosugi, D. McKay and A.V. Granato, J. Phys. IV **6**, 863 (1996), X. Liu, E-J. Thompson, B.E. White, Jr., and R. O. Pohl, *Phys. Rev. B* **59**, 11767 (1999), and Boston MRS symposium L, to be published, 1999.

[5] D.C. Ralph, A.W.W. Ludwig, J. von Delft, and R.A. Buhrman, *Phys. Rev. Lett.* **72**, 1064 (1994); R.J.P. Keijsers, O.I. Shklyarevskii, and H. van Kempen, *Phys. Rev. Lett.* **77**, 3411 (1996); G. Zárand *et al.*, *Phys. Rev. Lett.* **80**, 1353 (1998); O.P. Balkashin *et al.*, *Phys. Rev. B* **58**, 1294 (1998); S.K. Upadhyay *et al.*, *Phys. Rev. B* **56**, 12033 (1997).

[6] James P. Sethna and Stephen Kivelson, *Phys. Rev. B Rapid Communications* **26**, 3513 (1982).

[7] K. K. Mon and N. W. Ashcroft, *Solid State Comm.* **27**, 609 (1978).

[8] T. Vegge, O. B. Pedersen, T. Leffers and K. W. Jacobsen, Mat. Res. Soc. Symp. Proc. **578**, 217 (2000).

[9] P. Stoltze, *Simulation Methods in Atomic-scale Materials Physics* (Polyteknisk forlag, Lyngby, 1997).

[10] K.W. Jacobsen, J.K. Nørskov, and M.J. Puska, *Phys. Rev. B*, **35**, 7423, (1987). K.W. Jacobsen, P. Stoltze, and J.K. Nørskov, *Surf. Sci.* **366**, 394, (1996), and references therein.

[11] S. Coleman, 1977 Erice lectures, reprinted in his *Aspects of Symmetry*, Cambridge Univ. Press, Cambridge, 1985, p. 265.

[12] James P. Sethna, *Phys. Rev. B* **24**, 698 (1981), **25**, 5050 (1982).

[13] W.P. Su, J.R. Schrieffer, and A.J. Heeger, *Phys. Rev. Lett.* **42**, 1698 (1979); *Phys. Rev. B* **22**, 2099 (1980).

[14] Hirth and Lothe, *Theory of Dislocations* second ed., Krieger, Malabar Fl., 1992: quantum kink tunneling p. 533ff.

[15] J. Lothe, J. P. Hirth, Phys. Rev. **115**, 543 (1959), and A. Seeger and P. Schiller, in *Physical Acoustics*, edited by W. P. Mason, Academic Press (New York), 1966, Vol. 3A, pp. 361; J. D. Eshelby, Proc. R. Soc. London Ser. A **266**, 222 (1962).

[16] F. Kroupa and L. Lejček, Czech. J. Phys. B **25**, 65 (1975)

[17] Hirth and Lothe [14] constant line tension p. 252. A. Seeger and P. Schiller, and J. D. Eshelby [15] recognize that the effective mass is reduced by a factor of the longitudinal width of the kink, which Hovakimian *et al.* use to calculate depinning of dislocation kinks (L.B. Hovakimian, Phys. Rev. Lett. **68**, 1152 (1992), L.B. Hovakimian, K. Kojima and I. Okada, Phys. Rev. B **51**,

6319 (1995)).
[18] B. Joós, M. S. Duesbery, Phys. Rev. Lett. **78**, 266 (1997), and references therein. They deal with the exponential dependence of the ordinary Peierls barrier on width, rather than the corresponding barrier for the kink.
[19] G. Schottky, Phys. Stat. Sol. **5**, 697 (1964)