Comment on "Determination of Interlayer Diffusion Parameters for Ag/Ag(111)"

In a recent Letter [1] Roos and Tringides (RT) presented evidence in support of the hypothesis that the preexponential factor ν_s in the interlayer jump rate on Ag(111) exceeds the corresponding quantity ν_t for inlayer transport by about 2 orders of magnitude. As part of the argument they provide a simplified analysis of an experiment carried out by Bromann *et al.* (BBRK) [2], in which second layer nucleation on top of predeposited Ag islands on Ag(111) was investigated. Here I point out a mistake in the analysis of RT, and show that the correct application of their idea does not allow them to conclude that $\nu_s/\nu_t > 1$. I then discuss more broadly our present understanding of interlayer diffusion on Ag(111) in view of a recent reanalysis [3] of the BBRK experiment.

RT estimate the probability f that an atom deposited during the second dose descends from the island within the time τ between successive deposition events. They write $f = (\lambda/d)p$, where $\lambda = \sqrt{D_t \tau}$ is the diffusion length, d is the island diameter, and p is the probability that an atom poised at an edge site jumps down from the island. The ratio λ/d is referred to by RT as "*the number of edge interrogations.*" In fact this number is much larger than λ/d. Provided that $\lambda \gg d$, which is the case of interest here, the number of edge interrogations is given by the number of diffusion jumps $D_t\tau = \lambda^2$ multiplied by the fraction L/A of edge sites among all sites on the island; L is the island perimeter and $A \sim d^2$ is the island area in units of the lattice constant, so that $(L/A)D_t\tau \sim L(\lambda/d)^2 \gg \lambda/d$.

To obtain the correct expression for f, note that the probability that an atom on the island descends in a small time interval dt is $(L/A)D_s(1 - f)dt$, where $D_s = pD_t = (\nu_s/\nu_t)D_t \exp(-\Delta E_s/kT)$ is the interlayer hopping rate. Integrating up to time $t = \tau$ then yields

$$f = 1 - \exp[-(L/A)D_s\tau], \tag{1}$$

which corrects Eq. (1) of RT. By using the numbers given by RT, for the case of islands of radius 30 Å at temperature $T = 130$ K, we obtain $f = 1 - \exp[-130(\nu_s/\nu_t)]$ for $\Delta E_s = 0.13$ eV. Thus $f = 1$ provided that $\nu_s/\nu_t \geq 0.1$.

In addition to the BBRK experiment, RT base their conclusion on the analysis of two other growth situations involving interlayer transport. In all three cases they work at a single temperature, which implies that information about ν_s can be extracted only if ΔE_s is known. The value of ΔE_s used by RT was obtained in [2] by analyzing the dependence of the fraction of islands with second layer nuclei (a quantity somewhat similar to $1 - f$) as a function of the radius of predeposited islands, at two different temperatures $T = 120$ and 130 K. The analysis was based on a theory of second layer nucleation due to Tersoff *et al.* [4], which was recently shown to be quantitatively incorrect [3,5,6]. Other groups [7] have estimated ΔE_s assuming that the adatom density at second layer nucleation is comparable to that at which first layer islands nucleate, which is generally not true [3].

A reanalysis [3] of the BBRK data using the correct expression for the rate of second layer nucleation yields a very large step edge barrier $\Delta E_s \approx 0.32$ eV, accompanied by a prefactor $\nu_s \approx 8 \times 10^{19}\ \mathrm{s}^{-1}$, which would imply $\nu_s/\nu_t \approx 4 \times 10^8$. Since such a large preexponential factor is hard to justify physically, this suggests that a single pair of diffusion parameters is insufficient to describe interlayer transport on Ag(111). Assuming $\Delta E_s = 0.13$ eV as in [1], the reanalysis yields $1 < \nu_s/\nu_t < 10$.

In this situation additional second layer nucleation experiments at variable temperatures are called for. A temperature-dependent measurement of interlayer transport based on the decay of vacancy islands was performed by Morgenstern *et al.,* who estimate $\Delta E_s = 0.13$ eV and $\nu_s/\nu_t = 10^{-0.6\pm0.5} < 1$ [8]. The discrepancy between these numbers and the (correctly analyzed) BBRK experiment indicates that, despite considerable effort, interlayer diffusion for Ag(111) remains an open problem.

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