Roos and Tringides Reply: In response to the Comment of Morgenstern and Besenbacher [1], first we compare all the experiments [1–6] carried out to measure the interlayer probability $p = (\nu_s/\nu_t e^{-(\Delta E_s/kT)})$ for Ag/Ag(111). In the second part we address the questions raised about the diffraction experiment.

The main conclusion of [5] is the large prefactor ratio $\nu_s/\nu_t \gg 1$ for Ag/Ag(111). This conclusion is reached by other experiments, besides the RHEED experiment, over widely different length scales (from 3 to 50 nm). Experiments with $\nu_s/\nu_t \gg 1$ [3,5,6] are carried out at $T < 150$ K, while experiments with $\nu_s/\nu_t \sim 1$ [2,7] are carried out around $T = 300$ K. In [5] combining the results of the three independent experiments to reduce the uncertainty a specific value $\nu_s/\nu_t = 100$ is obtained. Since there is a large temperature difference of 150 K between experiments with different ν_s/ν_t ratios, it is not clear whether a single Arrhenius form should describe interlayer diffusion over such a wide temperature range. Possibly ν_s/ν_t decreases with temperature. In addition, nucleation processes can also be governed by long range interactions, especially at low temperatures, which can affect the measured prefactors in second layer nucleation experiments as in submonolayer experiments.

There is an indication that a single Arrhenius form might not apply over the whole temperature range. If one combines the measurement of the detachment rate β [2] and the terrace diffusion on Ag/Ag(111) [3] ($D_t = 2 \times 10^{11}(\mathrm{s}^{-1})e^{-0.1\ \mathrm{eV/kT}}$), an extremely low equilibrium value $\rho_\infty = (\beta/2\pi D_t n) = 10^{-9}$ atoms/nm^2 results. The equilibrium concentration difference at the inside and outside islands will be less than 10^{-9} atoms/nm^2 at 300 K. It is difficult to generate steady state diffusion currents in the region between the two islands driven by so small concentrations. The large difference between β and D_t has implications about the comparison of the decay rates of the vacancy vs adatom island since at 300 K the time to detach from the island ($\sim$4 sec) is much larger than the hopping time on the terrace (10^{-9} sec) and over the barrier (10^{-7} sec).

About the diffraction experiment it is important to emphasize that the RHEED experiment of [5] is carried out not on a macroscopic Ag(111) crystal but on a 40 ML Ag(111) film, grown epitaxially on Si(111) similar to other experiments [8]. The lattice constant and step height measured with spot profile analysis have the expected bulk values.

The average terrace size as measured from the FWHM of the (00) spot is 50 nm, while in [8] it is even smaller 20 nm. These sizes are smaller than the local average terraces shown in [1] and are obtained by averaging globally a much larger number of terraces ($\sim 10^7$) typical of diffraction. Also it is not clear if the step bunched terrace morphology described in [1] applies to the epitaxially grown crystals, since the annealing temperatures are different. From the Gaussian shape of the (00) spot we deduce a terrace size distribution peaked at the average value and not the distribution implied by the step bunched morphology. The intensity drop will be less on smaller terraces and more on larger terraces, so the average drop will be approximately the intensity drop on the average terrace. In addition the "1-island" regime extends over a finite temperature range, so even for terraces differing in size by a factor of 2 or 3 (which they are a small fraction in our size distribution), there will be an overlap in their "1-island" regimes. Since the experiments carried out on the epitaxially grown crystals give prefactor ratios consistent with the ratios on macroscopic crystals [3] at the same low temperatures, we believe the temperature of the experiment is more important than the substrate preparation.

Figure 2 of Ref. [6] shows that the steps are not fractal after Ag deposition of 0.1 ML. If, indeed they are fractals this implies that the intensity drop, due to the increase in the island perimeter, which is deduced from the experiment, is even smaller than what was deduced in [5]. Using a square lattice with no anisotropy in corner rounding results in nonfractal islands in the simulation. If fractal islands were modeled in the simulation, they will give a larger intensity drop, since the perimeter of a fractal island is larger than the perimeter of a compact island, for the same number of atoms. Thus both changes in the analysis result in a larger difference between the intensity drop in the simulation vs the experiment and require an even larger prefactor ratio.

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