Roos and Tringides Reply: There are three questions raised in [1] about the interlayer probability $p = (\nu_s/\nu_t e^{-(\Delta E_s/kT)})$ for Ag/Ag(111): (i) Is the prefactor ratio $\nu_s/\nu_t \gg 1$ as suggested in [2–4]? (ii) What is the correct theory to describe second layer nucleation? (iii) Why does the revised theory [5,6], when applied to Ref. [3], give unphysically high values for ν_s/ν_t and ΔE_s? Since Ref. [2] deals with question (i), our reply focuses on $\nu_s/\nu_t \gg 1$. We have expressed the main conclusion based on second layer nucleation experiments of Ref. [2] as $\nu_s/\nu_t \gg 1$ since the result $\nu_s/\nu_t \gg 1$ does not depend on the theory but a specific value does. A specific value $\nu_s/\nu_t = 100$ is deduced by considering two additional experiments [2]. Although the revised theory [5] gives a different value, it reinforces the conclusion $\nu_s/\nu_t \gg 1$. Question (ii) was discussed in the criticism [5,6] of the earlier theory [7]. The third question is still an open challenge.

The conclusion $\nu_s/\nu_t \gg 1$ for Ag/Ag(111) is supported by three independent experiments: the analysis of second layer nucleation experiments [2–4], the decay of the reflection high-energy electron diffraction (RHEED) intensity, and the size of the denuded zone [2]. Furthermore, there are three methods of analyzing the second layer nucleation experiments. Reference [1] discusses only one of the three methods, a small part of [2]. This method is the least robust because it uses only part of the experimental information; however, we will show that, even with this method, $\nu_s/\nu_t \gg 1$ holds.

In the original second layer nucleation experiment [3], fits to the fractional occupation curves have shown that $\nu_s/\nu_t = 50$, but with large uncertainty. Later, a method was proposed [4] that allows the extraction of ν_s/ν_t and ΔE_s uniquely. Since this method is far more exact than Eq. (1) of Ref. [2] (criticized in [1]), we review the method. From the expression of the nucleation rate $\Omega = (\pi^2/2)F^2R^5/pD_t$ [5] and the limiting values $\Omega_{\max}$ (when all the islands in the ensemble have second layer islands) and $\Omega_{\min}$ (when none of the islands in the ensemble have second layer occupation) in the experiment of [3], the dependence of the corresponding island sizes $R_{\max}$, $R_{\min}$ vs p is plotted in Fig. 1. By using the measured values of ($R_{\min}$, $R_{\max}$) [(2 nm, 4 nm) at $T = 120$ K and (3 nm, 7.5 nm) at $T = 130$ K [3]], we search for the values of p with the best agreement between measured and calculated values $\nu_s/\nu_t = 10^9$, $\Delta E_s = 0.32$ eV for the theory of [5] and $\nu_s/\nu_t = 10^3$, $\Delta E_s = 0.13$ eV for the theory of [7].

A third approximate method in Ref. [2] is based only on $R_{\min} = 3$ nm at $T = 130$ K by estimating the number of successful hops f over the step edge barrier, within the time between atom depositions $\tau = 1/FA$. The number of step interrogations as pointed out in [1] is $2\tau D_t/R_{\min}$ which implies $f = 2\tau p D_t/R_{\min}$. In Ref. [2] (and in [1]), $f > 1$ was taken as the condition for a deposited atom to descend the $R = 3$ nm island. However, the experiment was carried out on an ensemble of islands, not a single island.

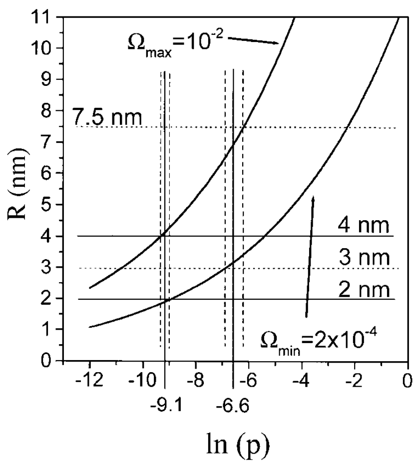

FIG. 1. Dependence of $R_{\max}, R_{\min}$ on p.

Since for time $f\tau$ no islands in the ensemble have second layer nucleation, this time should be larger than the inverse of the minimum nucleation rate $\Omega_{\min}^{-1}$. This gives a much stronger condition on $f > \Omega_{\min}^{-1}(1/\tau)$ than $f > 1$. From this condition, we derive $\nu_s/\nu_t > 7$ (using $\Omega_{\min} = 2.8 \times 10^{-4}$ s^{-1}, $\Delta E_s = 0.13$ eV). This lower bound is less than $\nu_s/\nu_t = 100$ because only the data at $T = 130$ K are used. If one uses the data at 130 K, 120 K, $\nu_s/\nu_t = 100$ results (with the other experiments).

For the other experiments (RHEED, denuded zone), which are the major part of [2], Ref. [1] discusses only whether $\Delta E_s = 0.13$ eV is justified. This is the value consistent with all the experiments [2,3,8,9]. Additional constraint $\Delta E_s > 0.1$ eV (and $\nu_s/\nu_t > 10$) can be deduced from the absence of diffraction oscillations at $T = 500$ K in Ag/Ag(111) and assuming the maximum value $n = 1$ in $R_{\min} \sim p^n$ (and therefore the lowest value of ΔE_s).

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