Comment on "Quantum Scattering of Heavy Particles from a 10 K Cu(111) Surface"

In a recent Letter Althoff *et al.* [1] reported a study of scattering of thermal Ne, Ar, and Kr atoms from a Cu(111) surface in which they assessed the corresponding Debye-Waller factor (DWF) as a function of the particle mass m in a wide range of substrate temperature T. The experiments were interpreted by the semiclassical DWF theory [2–4] in which the projectile moves on the classical recoilless trajectory $\mathbf{r}(t)$ and the surface vibrations are quantized. This gives the DWF in the form $I_{00} = \exp[-2W(m,T)]$ where the Debye-Waller exponent $2W$ (DWE) in the essentially one-dimensional (1D) approach of Refs. [2–4] and the limit $T \rightarrow 0$ depends on the particle incoming energy E_i, surface Debye temperature Θ_D, and the static atom-surface potential $V(z)$, but not on m. On the other hand, in the scattering regime $T \gg \Theta_\tau = \hbar/(k_B \tau)$ and $\Theta_\tau < \Theta_D$, where τ is the effective collision time, it scales as $2W \propto m^{1/2}T$. However, the experiments described in [1] were carried out in the quantum scattering regime in which, as we show below, neither of the above semiclassical scalings holds and the semiclassical DWE significantly deviates from the exact quantum one both in the low and high T limits, irrespective of the form of $V(z)$. Hence, the quantum scattering data [1] cannot be reliably interpreted by the semiclassical but rather by the quantum theory.

To substantiate these statements we carry out a fully three-dimensional quantum [5–7] and semiclassical [4] calculation of the DWF relevant to the experiments of Ref. [1]. We start from the quantum DWE [cf. Ref. [6], Eq. (3)] in which we also include prompt sticking processes because of their large contribution to the quantum DWF and use theoretical $V(z)$'s from [1] which produce a closer fit of the semiclassical DWF to the data. The quantum DWF has the correct semiclassical limit [5] which enables pinpointing the breakdown of the semiclassical description. A comparison of the measured [1] and calculated DWF's shown in Fig. 1 reveals general agreement between the measured and quantum and not the semiclassical values, confirming the validity of quantum approach. A systematic small underestimate at very low T appears because our results are uncorrected for T-independent diffuse elastic scattering. The breakdown of the semiclassical approach is illustrated in Fig. 2 which shows the m dependence of the quantum and semiclassical DWE's calculated in the low (main panel) and high (inset) T limits for fixed $V(z)$ and E_i. It is seen that the semiclassical 1D scaling results for the DWE are reached for masses which largely exceed those of the scattered atoms. Hence, although our calculations support the conjectures of Ref. [1] on the m dependence of the DWF for heavy particle-surface scattering in the *classical* limit, they also demonstrate that the semiclassical approach is not yet applicable in the studied scattering regime and that the *quantum* approach should be used instead.

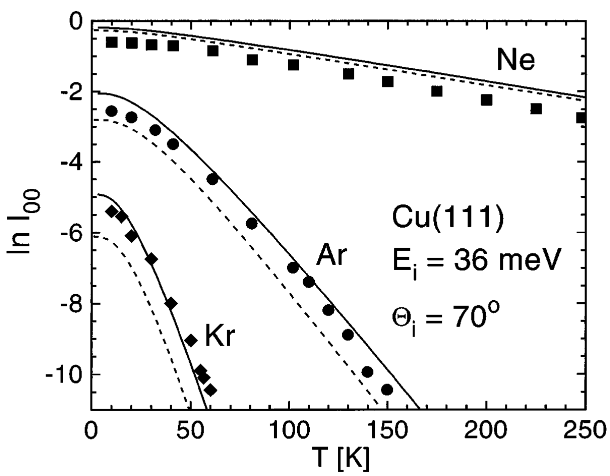

FIG. 1. Calculated temperature dependence of the quantum (full curves) and semiclassical (dashed curves) Debye-Waller factor for Ne, Ar, and Kr scattering from Cu(111) and experimental results [1] (full symbols).

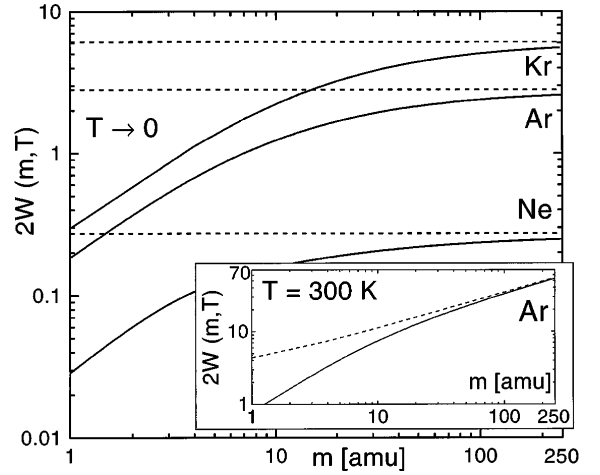

FIG. 2. Dependence of quantum (full curves) and semiclassical Debye-Waller exponent (dashed curves) on particle mass for fixed $V(z)$ and E_i in the low and high temperature limits.

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