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## Comment on "Non-Arrhenius behavior of surface diffusion near a phase transition boundary" - Reply

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**Vattulainen *et al.* Reply:** In their Comment [1] to our Letter [2], Uebing and Zhdanov (UZ) argue that the non-Arrhenius (NA) behavior of  $D_C$  in the O/W(110) model system we studied [2] cannot be predominantly due to the average transition rate  $\Gamma$ . Instead, based on mean-field (MF) arguments and Monte Carlo (MC) simulations of an unrealistic model [3], UZ claim that the NA behavior of  $D_C$  in *all cases* arises solely from the thermodynamic compressibility factor  $\kappa_T \propto \langle(\delta N)^2\rangle/\langle N\rangle \equiv 1/\xi$ . This issue has already been discussed by us in a recent paper, where we have shown this claim not to be correct [4]. Since UZ seem to ignore this work, we present some of the discussion here.

UZ claim that the NA behavior we observed [2] results from our system sizes being too small. This is definitely not the case as explained in detail in Ref. [4]. To further emphasize this point, we note that the compressibility exponent in model C [5], where  $\theta$  is not the order parameter, depends on the specific heat exponent  $\alpha$  which is generally small. Furthermore, for the symmetry class of a 2D XY model in a cubic anisotropy field to which the present system belongs,  $\alpha$  is nonuniversal and *negative*. It is thus expected that near  $T_c$  there is just a rounded cusp for  $\kappa_T$  whose size dependence is very weak. In fact, the data of UZ for large system sizes  $L \times L$  completely agree with this. The fact that UZ do not observe the cusp for  $L = 64$  is probably due to the low quality in their simulations. As far as our simulation work is concerned, in Fig. 1 we show  $D_C$ ,  $\xi$ , and  $\Gamma$  for  $L = 30$  and  $120$ , the latter size being typical in realistic surfaces where the average terrace length is around 100 lattice spacings. Near  $T_c$ , the cusp for  $\kappa_T \propto 1/\xi$  we find shows only very weak dependence on  $L$ , while the jump rate  $\Gamma$  remains virtually the same. This shows that even for system sizes comparable to experiments, the behavior of  $D_C$  and the corresponding effective barrier  $E_A$  near  $T_c$  in *this case* is indeed dominated by the jump rate  $\Gamma$ , and not by the compressibility  $\kappa_T$ . These results clearly contradict the claims of UZ [1].

The so-called “general theory” of UZ concerning the critical behavior of  $\Gamma$  is based on a MF approximation together with the adoption of initial value (IV) dynamics. This choice of dynamics is totally unrealistic for an interacting system near a critical region. UZ use a slight variation of the IV dynamics in their MC simulations, and it is not surprising that their simulation results are in agreement with their own “general theory” for  $\Gamma$ . What UZ do not seem to recognize is that a dynamical quantity such as  $\Gamma$ , unlike the equilibrium properties, does not depend only on the lattice-gas Hamiltonian but also depends crucially on the details of the stochastic dynamics chosen in MC studies. Our simulations were performed with a more realistic TDA (transition dynamics algorithm) dynamics [4], and that is the main reason why we capture the prominent NA behavior in  $\Gamma$ . In-

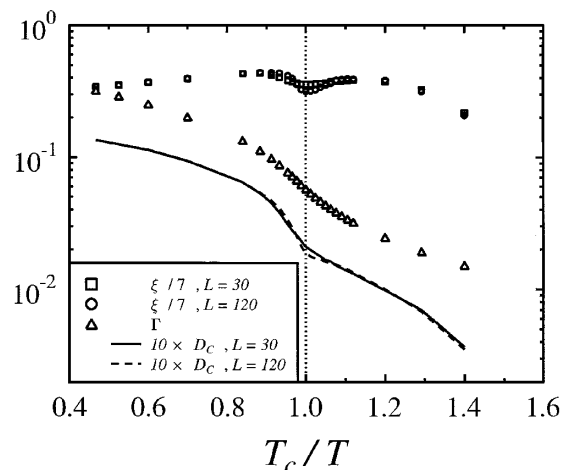


FIG. 1. Results for  $D_C$ ,  $\xi$ , and  $\Gamma$  as a typical Arrhenius plot in the O/W(110) system at a coverage of  $\theta = 0.45$ .

stead, UZ observe only a near-Arrhenius behavior for  $\Gamma$  as reflected in their results for  $D_T$ . In fact, we have repeated our studies with the dynamics employed by UZ, as well as three other dynamics often used in MC simulations. We find that the dynamics used by UZ yield an effective barrier that has just a very small cusp at  $T_c$ . Other dynamics such as Metropolis and Kawasaki are consistent with TDA results and show a prominent NA behavior as found in our published works [2,4]. Furthermore, we have recently studied the validity of MC dynamics through MD simulations and find that the UZ dynamics does not describe the activation barrier qualitatively correctly in all cases.

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