Non-stochastic behaviour of surface atomic diffusion at all temperatures

J. Ferrón
INTEC-CONICET, Univ. Nacional del Litoral, 3000-Santa Fe, Argentina

L. Gómez
Fac. Cs. Exactas e Ingeniería, Univ. Nacional de Rosario, Instituto de Física Rosario, 2000-Rosario, Argentina

R. Miranda and J.J. de Miguel*
Dpto Física de la Materia Condensada, Universidad Autónoma de Madrid, Cantoblanco, 28049-Madrid, Spain

Atomic diffusion is usually understood as a succession of random, independent displacements in which the adatom would gain thermal energy from the substrate, use it to overcome the diffusion barrier and reach a nearby potential well where it would become thermalized, lose memory of its previous movement and start the process again. Nevertheless, from an analysis of Molecular Dynamics simulations of self-diffusion on Cu(111) using Embedded Atom interatomic potentials it is found that a large proportion of the jumps making up the diffusion path are in fact correlated. At high temperatures the adatom never reaches thermal equilibrium with the surface, and its displacements can be considered ballistic, consisting of "long jumps" spanning several surface unit cells along the high symmetry directions of the substrate. On the contrary, at low temperatures recrossing events predominate, in which after a first nearest-neighbor jump the adatoms preferentially return to their former position. In all cases, only a minority of the total number of jumps can be considered truly statistically independent. For thise reason the atomic movements cannot be correctly described in general in terms of a random walk model. This fact has a profound impact on the determination and interpretation of diffusion coefficients.

*E-mail: juanjose.demiguel@uam.es

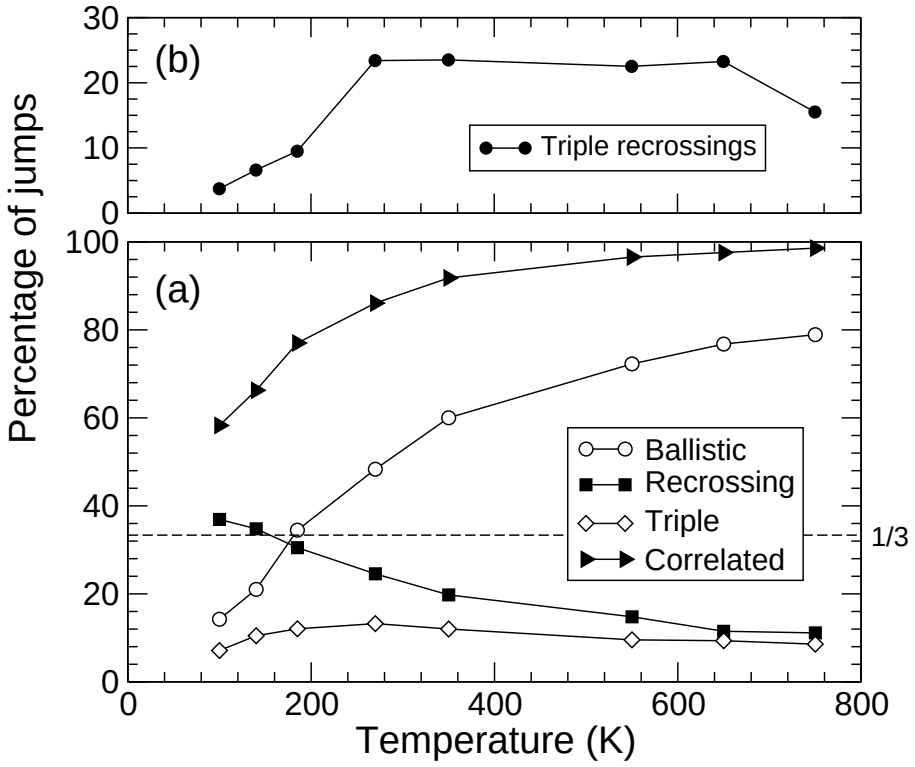

Figure 1: Statistical analysis of correlations in atomic jumps. The total fraction of correlated jumps is given by the triangles; even at 100 K this figure exceeds 0.5, reaching nearly 1.0 at the higher temperatures.