Dynamical Transitions in Correlated Driven Diffusion in a Periodic Potential

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(Received 25 November 1996)

The diffusion of a two-dimensional array of particles driven by a constant force in the presence of a periodic external potential exhibits a hierarchy of dynamical phase transitions when the driving force is varied. This behavior can be explained by a simple phenomenological approach which reduces the system of strongly interacting particles to weakly interacting quasiparticles (kinks). The richness of the strongly coupled system is, however, not lost because, contrary to a single Brownian particle, the array shows an hysteretic behavior even at nonzero temperature. The present investigation can be viewed as a first step toward understanding nanotribology. [S0031-9007(97)02426-5]

PACS numbers: 66.30.–h, 05.70.Ln, 46.10.+z, 63.20.Ry

The diffusion of a Brownian particle, driven by an external force and subjected to a periodic potential, is a situation which arises in several fields of science [1] such as solid-state physics, surface physics [2], chemical physics, and even communication theory. It is now well understood and provides a simple example of an out-of-equilibrium phase transition, between a locked and a running state. The case of many interacting particles is even more interesting because collective effects modify qualitatively the picture giving rise, in some parameter range, to a dynamical state which is very reminiscent of *a traffic jam at the atomic scale*. We discuss here this behavior in the context of solid-state friction because it provides a typical example which may be amenable to experimental tests, but the basic ingredients required to observe the phenomenon are simple and can be found in many physical systems.

Understanding the diffusion and mobility of strongly interacting atoms subjected to a periodic potential and driven by an external force is a first step toward understanding solid friction at the atomic level [3]. For noninteracting atoms the problem would be simple since it essentially reduces to the diffusion of a Brownian particle in a periodic potential. Under the influence of a dc force F it will preferably diffuse in the direction of the force and in average there will be a drift velocity $\langle v \rangle$ which depends on F. The mobility B is then defined as $B = \langle v \rangle / F$. If, for small forces, B is independent on F (the linear response regime), for arbitrary ones a nonlinear response takes place, and the task is to calculate this nonlinear mobility.

The total potential experienced by the Brownian particle is the sum of the periodic potential and the potential $-Fx$ due to the driving force, i.e., it corresponds to a corrugated plane, with an average slope determined by F. At small forces the potential has local minima, therefore the particle is static and its mobility vanishes. On the contrary, for large forces there are no stable positions, and the particle slides over the corrugated potential, reaching its maximum mobility $B_f = (m\eta)^{-1}$, where m is the mass of the particle and η is the friction coefficient. A simple calculation shows that, denoting by ε the height of the periodic potential, a the lattice constant, and c a constant depending on the shape of potential, the critical force for which the stable positions disappear is $F_r^0 = c\,\varepsilon/a$.

However, for the underdamped case, the system may also have a running solution, even if the minima of the potential do exist. Indeed, because of their momentum the particles may overcome the next hill, which is lower than the one from which they are falling due to the $-Fx$ contribution to the potential, if the gain in potential energy is greater than the energy dissipated during this motion. One finds that this second critical force is $F_\eta^0 = c'\eta\sqrt{m\varepsilon}$, where c' is a constant which depends on the potential shape. As the particle is either locked or running, depending on its initial velocity, the system exhibits bistability, and the transition between these two states shows hysteresis. However, *for a single particle the bistability disappears in the presence of an external noise such as thermal fluctuations, no matter how small the noise is,* because the fluctuations can kick the particle out of the locked state by usual thermal activation. Thus the Brownian motion of a single particle driven by an external force shows hysteresis only for zero temperature.

The case of interacting particles in a periodic potential is a much more difficult problem; interesting numerical results were obtained for high damping, when the time-independent Schmoluchowsky equations may be reduced to a one-particle equation with an effective on-site potential and then solved numerically by the transfer-integral method [4]. The results show that there is a region of highly nonlinear mobility, but without a bistability phenomenon. Besides, recently Persson [5] has used molecular dynamics to study a 2D system of interacting atoms subjected to a periodic potential. In the underdamped case, he observed a dynamical phase transition similar to the one-particle case. Recalling the well-known Aubry transition from the pinned state to the freely moving state in the Frenkel-Kontorova model with an incommensurate atomic concentration [6], Persson supposed that his results could be explained in a similar way.

In the present Letter, we study the driven motion of interacting atoms in a 2D potential for arbitrary damping.

First, we show that the final state observed by Persson corresponds in fact to the sliding motion of atoms in an inclined potential. Then we demonstrate that the transition from the locked to the sliding state passes through a hierarchy of hysteretic depinning transitions. An important point is that *hysteresis persists in the presence of thermal fluctuations*. Finally, we analytically compute the critical forces and the corresponding mobilities using a phenomenological approach [7] which treats a system of strongly interacting atoms as a system of weakly interacting quasiparticles (kinks).

Although it is still oversimplified, the generalized Frenkel-Kontorova model that we consider provides a rather complete description of a layer of atoms adsorbed on a 2D crystalline surface when the motions in the vertical dimension are also taken into account. The parameters are chosen for the adsystem Na-W(112), and this model was proposed [8] to explain the intriguing experimental results obtained for the dependence of the diffusion coefficient of strongly interacting adatoms when the concentration varies.

The model considers the following Langevin equation for the x coordinate of ith atom,

$$m\ddot{x}_i + m\eta\dot{x}_i + \frac{d}{dx_i}\left[V_x(x_i) + V_y(y_i) + \frac{1}{2}m\omega_z^2 z_i^2 + \sum_{j\neq i} V_0 \exp(-\beta_0|\vec{r}_i - \vec{r}_j|)\right] = F^{(x)} + \delta F_i^{(x)}(t)\,, \qquad (1)$$

and similar equations for the coordinates y_i and z_i. The particles are therefore in a periodic rectangular potential,

$$V_\alpha(\alpha) = \frac{\varepsilon_\alpha}{2}\,\frac{(1+s_\alpha)^2[1-\cos(2\pi\alpha/a_\alpha)]}{1+s_\alpha^2-2s_\alpha\cos(2\pi\alpha/a_\alpha)}\,, \qquad (2)$$

where α is x or y, and parameters $s_x = 0.2$, $s_y = 0.4$, $\varepsilon_x = 0.46$ eV, $\varepsilon_y = 0.76$ eV, $a_x = 2.74$ Å, $a_y = 4.47$ Å are chosen in relation to the highly anisotropic *channeled* surface W(112) [8]. Atomic mass is $m = 1$, and we put $\omega_z = 1.84$, $\eta = 0.165$ in the corresponding units. The exponential interaction law with $V_0 = 10$ eV, $\beta_0 = 0.85$ Å^{-1} corresponds to the repulsion of the adatoms at rather high concentration, $F^{(x)}$ is the external force applied *along the channels*, and δF is a Gaussian random force simulating the interaction with a thermal bath. We start with a ground state of the system, then the temperature and later the force are increased adiabatically. We then compute the mobility for different values of F and T.

An important parameter is the atomic concentration θ, corresponding to the ratio between the number of particles N and the number of available sites M. In this Letter we present data at several generic concentrations in order to show various aspects, although the general statements are common for the wide range of atomic concentrations.

Let us first consider the case of $\theta = 21/41$ modeled by 30 channels, each with $N = 105$ and $M = 205$. As this concentration, close to $\theta = \frac{1}{2}$, is not a simple $1/q$ value with q integer, the ground state corresponds to large regions where the effective concentration is $\frac{1}{2}$, equidistantly separated by zones of compression [8]. Since these zones in the standard Frenkel-Kontorova (FK) model are called kinks (or antikinks in the case of localized expansions), we use this terminology here, too. The number of kinks is proportional to the difference between θ and the closest value $1/q$ [7]. All the parameters, such as the mass of kink, Peierls-Nabarro potential, and energy of creation of kink-antikink pair can be easily evaluated [8]. In spite of the obvious difference with the mathematical description of soliton behavior in nonlinear partial differential equations, we will show that there are surprising similarities and, moreover, that they allow us to explain *qualitatively* and *quantitatively* the intriguing behavior of the system in a simple way.

A generic evolution of the mobility as a function of the external force is shown on Fig. 1. One first notices a hysteresis and distinguishes four different regions when the external force is increased. In the very low force range, the mobility is zero. Above a critical force F_k, the mobility jumps to a first plateau at B_k. Then there is a second plateau around B_m for $F > F_{\rm pair}$, and finally for a force higher than F_r, the system reaches the maximum mobility B_f. When the force is decreased, the systems jump directly to the static state for forces lower than F_η. Let us explain these different states.

In the first region, the force is too low, and neither the kinks nor the individual atoms move. Both are trapped in their wells and the system is in the *locked state*. When

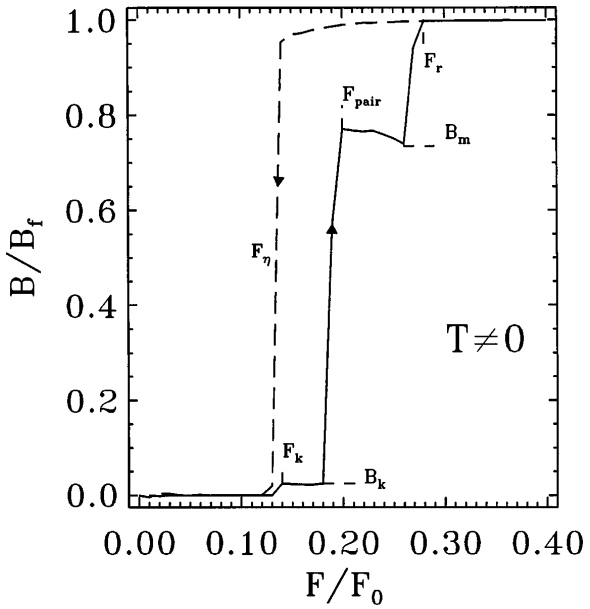

FIG. 1. Mobility versus force for a concentration $\theta = 21/41$ ($N = 105$, $M = 205$). The solid curve corresponds to increasing force and the dashed curve to decreasing force.

the force reaches a critical value F_k, the mobility jumps to a nonzero value B_k. This state corresponds to the *kink-running state*. Indeed, a careful study of the time dependence of positions of all atoms indicates clearly that the compressed zones are moving but not the individual atoms, except when a kink passes through their site. A simple analogy with a single particle in the periodic potential allows one to compute this critical force. Owing to the lattice discreteness, a kink in the FK model moves in the periodic Peierls-Nabarro potential, whose barrier E_k could be easily determined [8]; in addition, the potential is tilted due to the external force. This quasiparticle (kink) will be trapped until the last stable state disappears, i.e., when $F_k = cE_k/a$. Besides, as we know the number of kinks θ_k in the system, the mobility of the kink-running state is $B_k = \theta_k B_f$. This approach is very successful as shown in Fig. 1, where the values B_k and F_k are shown.

In the kink-running domain, the atoms are static contrary to the kinks since the energy barrier ε is always greater than the Peierls-Nabarro barrier E_k. Physically this phenomenon signifies that it is easier to move a dislocation coherently than to move all the atoms. In addition, a detailed study shows a decrease of the relative distance between the kinks. One finds a tendency for the kinks to bunch like the cyclists in a "peloton." The probable reason is that, as the kinks are not exact solutions of the system and therefore radiate waves, the oscillatory tail of a kink could help the following kink to overcome the Peierls barrier and to catch up with the previous one. A second point to notice is that this motion of kinks shows also a hysteresis even for nonzero temperature. If one decreases the force when this first plateau is reached, the system goes back to the locked state for a critical force lower than F_k. This indicates that kinks have a behavior more complex than a single Brownian particle, although the plateau at B_k is well predicted by the one-particle picture.

In the range of force corresponding to the second plateau, at $F > F_{\text{pair}}$, additional kinks and antikinks could be created since the energy barrier for the nucleation of new pairs vanishes. Therefore, we have to take into account not only the geometrical (ground state) kinks but also the "force-excited" kinks [7]. Figure 2 presents the time dependence of the positions of all atoms. Two different regions can be seen, mobile and immobile ones. The finite time interval between the snapshots results in a stroboscopic effect giving a wrong impression for atomic trajectories; in order to show one actual trajectory we marked one of atoms by black diamonds while others are indicated by unfilled diamonds. The picture is very reminiscent of a traffic jam: The particle is trapped into an immobile zone, until being first in this region; then the particle ballistically moves to the next high-density zone, where it is stopped again. The velocity probability presents a two-bells shape, corresponding to static and moving atoms: this is a *coexistence regime*.

A careful examination of the $B(F)$ dependence in this range of F shows that the mobility is even slightly decreasing with increasing force. Indeed, after the kink-antikink nucleation threshold F_{pair}, the kinks start to bunch into compact groups, as in the context of Josephson junction [9]. A simple phenomenological theory gives $B_m(\theta) \propto B_f\ (1 - \theta)/\theta$, which agrees very accurately with the numerical results [10]. Moreover, the study shows that the mobile subsystem corresponds to a bunch of the antikinks, whereas the immobile one corresponds to a bunch of the kinks.

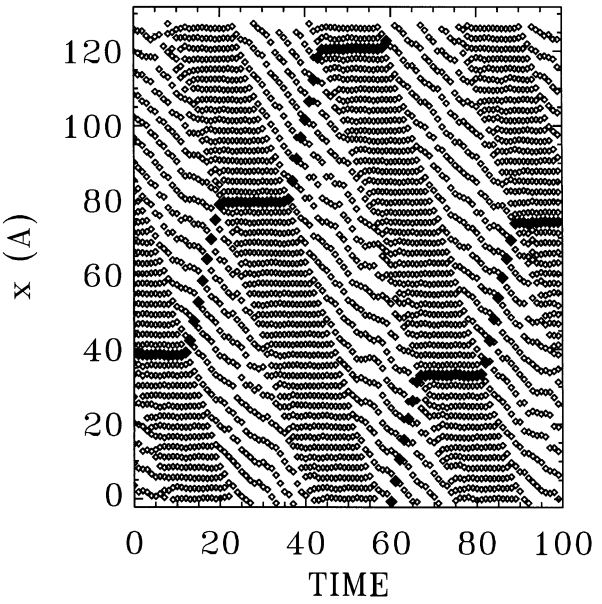

FIG. 2. Atomic trajectories in a given channel for the 2D system with $\theta = 34/47$ at $F = 0.24$. The black diamonds correspond to the trajectory of one atom.

Finally, after the coexistence regime, for high enough force, all atoms are sliding over the periodic potential and the mobility reaches its maximum value B_f. When the force is reduced, the description of the dynamics in terms of kinks has lost its meaning since no special organization subsists. This is why the system goes back directly to the locked state at the critical value F_η.

Thus the system of strongly interacting particles in an anisotropic external potential does present a dynamical phase transition when the dc external force is varied. We are able to explain the multiple steps by the *hierarchy of depinning*: first, the geometrical kinks, then the "force excited" kinks, and finally the atoms. But it is more remarkable that this behavior does not disappear with thermal fluctuations as the hysteresis of a single driven particle in a periodic potential. Figure 3 attests that the hysteresis survives for finite temperatures.

Let us derive an approximate expression for the critical force F_η versus temperature. At zero temperature the back transition approximately corresponds for low concentration to the external single-particle threshold F_η^0. For nonzero temperatures, the system gets locked when the probability for the velocities to be lower than $F_\eta^0/m\eta$ is greater than a threshold P_c. As a result, the critical

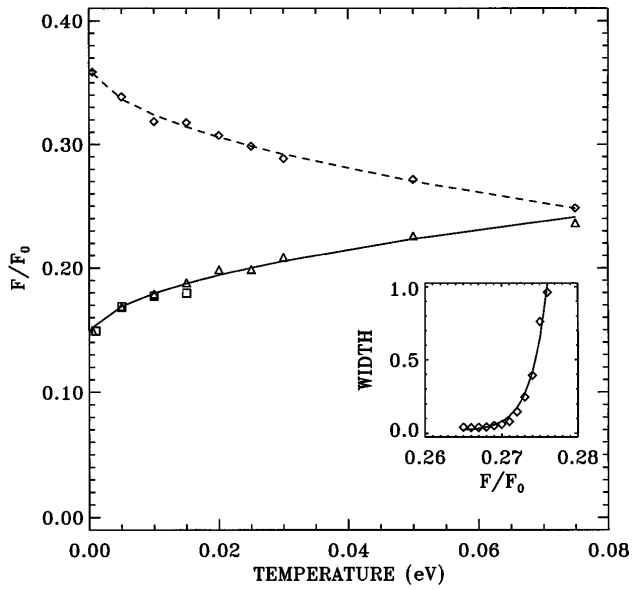

FIG. 3. The diamonds (triangles) correspond to the position of the transition to the running (locked) state for different temperatures in the case $\theta = 21/31$, while the squares correspond to the transition to the locked state for $\theta = 21/41$. The threshold was chosen to be $B = 0.9B_f$. The solid and dashed curves correspond to the phenomenological approach discussed in the text. The inset shows the width of the "river" (the moving neighboring channels) in the 2D case as a function of the driving force, and the solid line corresponds to the approximate expression.

force follows the law

$$F_\eta = F_\eta^0 + \sqrt{2mk_BT\eta^2}\,\mathrm{erf}^{-1}(1 - 2P_c) = F_\eta^0 + \delta\sqrt{T}, \tag{3}$$

where erf^{-1} is the inverse of the error function. With $F_\eta^0 \simeq 0.144$, the solid curve in Fig. 3 shows that this expression scales very accurately with the above expression if $\delta = 0.35$.

Unfortunately, we do not have a complete understanding of the evolution of the forward transition versus temperature. Because of temperature fluctuations, the particles feel a smoother potential and a smaller barrier to overcome; therefore the system jumps to the final running state for lower external forces. Numerical results plotted in Fig. 3 for the case $\theta = 21/31$ scale with the expression $F_r = F_r^0 - \xi\sqrt{T}$, if we chose $F_r^0 = 0.37$ and $\xi = 0.44$. While such a law is valid for other concentrations as well, the parameters significantly depend on the concentration contrary to the law for F_η.

The behavior of the 1D version of the problem is almost identical to the 2D case. The only difference that we have to notice is in the transition to the final sliding state. The exact critical force depends slightly on the external conditions, which means that the transitions do not occur simultaneously in all the channels. A careful examination of the behavior in different channels shows two interesting results. First, when a channel has jumped to the sliding state, it enhances the probability for the neighboring channels to jump due to interatomic interactions. This first channel corresponds therefore to a nucleation event. Indeed, the width of the "river" of neighboring moving channels grows faster than the number of independent rivers. The inset of Fig. 3 presents the numerical results which scale very well with an exponential law $\exp[(F - F_0)/\Delta F]$ with $F_0 = 0.276$ and $\Delta F = 0.002$. Of course, the value of ΔF attests that it is a very thin effect, but this phenomenon reveals an enhanced transition to sliding state due to cooperative effects in the second dimension. On the contrary, the transition of a channel to the locked state is almost independent of neighboring channels.

In conclusion, a driven system of interacting particles in a 2D anisotropic external potential exhibits several dynamical transitions successfully explained by a hierarchy of depinnings. This behavior, which is much richer than the behavior of a single Brownian particle, illustrates the interesting properties of complex systems. The analysis of the transitions can be put in the framework of a single particle theory by introducing collective excitations which have particlelike properties, although the strong discreteness of the system does not allow us to consider these excitations as solitons. The complex behavior of the multiparticle system is, however, not lost because, after the transition to the full running state, the collective organization is completely destroyed so that the backward transition does not look like the upward sequence of transitions. This explains why the complex system can maintain an hysteresis at nonzero temperature contrary to the single Brownian particle. This result, which emphasizes the role of hysteresis at microscopic scale, should be related to the main role of hysteresis found in solid friction [11]. Work along this line is in progress.

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