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## Two-stage melting in tribological systems

Alexander V. Zhukov <sup>a,\*</sup>, Maxim V. Paliy <sup>a</sup>, Oleg M. Braun <sup>b</sup>, Thomas F. George <sup>c</sup><sup>a</sup> Department of Chemistry, University of Western Ontario, London, Ontario, N6A 5B7, Canada<sup>b</sup> Institute of Physics, National Academy of Sciences of Ukraine, 03028 Kiev, Ukraine<sup>c</sup> Office of the Chancellor/Departments of Chemistry & Biochemistry and Physics & Astronomy, University of Missouri, St. Louis, MO 63121, USA

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### Abstract

A melting phase transition in a thin lubricant film confined between two substrates in moving contact is considered. General theoretical arguments are usually used to prove that ordinary bulk melting is always a first-order phase transition. However, this is not the case for systems with static and dynamic asymmetries. It is shown theoretically that a given tribological system can exhibit a novel two-stage continuous second-order transition, which is confirmed by a Langevin molecular dynamics study.

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The interest in the problem of sliding friction between two substrates in moving contact is due to both its applied engineering importance and fundamental physical features [1–3]. Usually it is believed that liquid lubricants are more appropriate for reducing friction than solid ones. However, when the width of the lubricant film decreases to a few atomic layers, it usually solidifies due to the high load and influence of substrates [1]. Thus, one of the crucial questions in tribology is the mechanism of the liquid–solid transition in thin systems subjected to an external sliding force. This question has been addressed in different experimental and theoretical studies (see the first book in Ref. [1] for a review), but there is still no clear comprehensive picture of this phenomenon because of its richness and high sensibility to external conditions and types of materials used.

The common picture of sliding dynamics with a solid lubricant is as follows. At low driving velocities, the motion is stick-slip-like, where short fast-sliding events due to the film

melting are separated by long sticking ones when the film solidifies back [4,5].

Stick-slip friction as well as solid–liquid transitions in general have been studied theoretically in e.g. Refs. [6–8]. In spite of the obvious success of the above cited and other theoretical descriptions, these do not cover all the many possible scenarios for liquid–solid transitions. The common feature of existing theories is a treatment of the lubricant as a strongly-interacting system, i.e., when the strength of potential of interaction between particles in the lubricant  $g_{ll}$  is much larger than, e.g., an effective lubricant-driving substrate interaction  $g_{ls}$  (we will return to this point later). In such a case, friction leads to effective isotropic heating of the lubricant, which finally melts when its temperature  $T$  reaches the melting temperature of the lubricant  $T_m$ , similarly to how it happens in an ordinary bulk crystal.

However, the picture can be quite different when the interaction between lubricant particles in a very thin film is weak enough (in the sense that we define more precisely below). To explain the general picture, let us first consider a simplified, trivial system consisting of a lubricant whose particles do not interact with each other but strongly interact with a substrate. For simplicity, we consider a single layer where the lubricant particles are initially placed to form a perfect quasi-2D crystal.

\* Corresponding author.

E-mail address: [alex.zhukov@gmail.com](mailto:alex.zhukov@gmail.com) (A.V. Zhukov).

<sup>1</sup> Present address: Faculty of Science, Palacký University, 17, Listopadu 50, 77200 Olomouc, Czech Republic.

If we increase the driving force along one of the crystal planes, then at some point the particles of the lubricant start to disorder along the direction of dynamical anisotropy (friction force). Obviously in our idealized system there is no transversal force because lubricant particles are non-interacting. Thus, the system remains crystalline-like in the direction perpendicular to the driving friction force. In other words, effective 1D melting occurs in the system. It might look confusing that the system forms a stable 1D crystal in the transverse direction, since it was proved long time ago that stable 1D and 2D crystals cannot exist in nature [11]. Indeed, let us imagine a crystal with periodic density function  $\rho(z)$  consisting of atoms arranged in parallel straight rows, where these rows are all equally oriented along the  $z$ -axis, but completely randomly placed with respect to each other. It can be shown that the change of free energy  $\Phi$  in this case is determined by the  $z$ -component of the displacement vector  $u_z$  only,  $\Delta\Phi \propto u_{z\mathbf{k}}^2 k_z^2$ , where  $\mathbf{k}$  is the reciprocal lattice vector. The mean square of  $u_{z\mathbf{k}}^2$  scales as  $\langle u_{z\mathbf{k}}^2 \rangle \propto k_z^{-2}$ . In order to find the fluctuation of the displacements, we have to do a summation over all the wavevectors or integrate in the Debye limit,  $\langle u_z^2 \rangle \propto \int k_z^{-2} d^3k$ , which diverges like  $1/k_z$  when  $k_z \rightarrow 0$ . However, this is not the case in a finite system, where we have to do a summation starting from some non-zero vector  $k_z \sim 1/d$ , where  $d$  is the characteristic size of the system in the  $z$ -direction. Therefore, there are no infinite range fluctuations in real finite systems, but we still can speak about a *quasi*-stable 1D crystal only, because the fluctuations will apparently destroy the order on certain time scales.

Some indications of the above-mentioned two-stage melting mechanism have been observed in Langevin MD simulations for soft lubricants [1,3,5,9], but have not been studied systematically. Similar effects were studied also in several papers (see, e.g., [10]) in the context of vortex or defect dynamics in 1D channels using different artificial models. Instead, we treat here quasi-1D melting generally, without restrictions to the model used.

Now let us formulate our idea more precisely for a realistic system driven frictionally in terms of the effective *longitudinal*  $T_{\parallel}$  and *transverse*  $T_{\perp}$  temperatures of the lubricant, compared to its melting temperature  $T_m$ . It is clear that two-stage melting will occur in the system if the following conditions are met:  $T_{\perp}(g_{ll}, g_{ls}) < T_m(g_{ll}) < T_{\parallel}(g_{ll}, g_{ls})$ . By the parentheses we express the *important* dependencies of all three temperatures on the main system parameters (such as driving strength, interactions, etc.). While  $g_{ll}$  can be rather well defined through the static interaction potential between atoms of the lubricant, we introduce the *effective* strength of the lubricant–substrate interaction as  $g_{ls} = g_{ls}^{(s)} + g_{ls}^{(d)}$ , where the first term is due to static interactions, while the second one is due to driving. Thus, the lubricant can go from a more “hard” to more “soft” case as  $g_{ls}^{(d)}$  increases, even if the static interactions are fixed. Obviously,  $T_m(g_{ll})$  is expected to increase monotonically as  $g_{ll}$  increases (and it depends only weakly on  $g_{ls}^{(s)}$ ). Once  $g_{ll}$  is fixed, both  $T_{\parallel}(g_{ll}, g_{ls})$  and  $T_{\perp}(g_{ll}, g_{ls})$  are generally expected to increase with  $g_{ls}$  increasing in such a way that initially, at infinitesimally small driving  $g_{ls}^{(d)} \rightarrow 0$ , we have  $T_{\perp}(g_{ll}, g_{ls}) \rightarrow$

$T_{\parallel}(g_{ll}, g_{ls}) \rightarrow T_0(g_{ll}, g_{ls}^{(s)}) < T_m(g_{ll})$ . Therefore, the system should be rather “hard”, i.e.,  $g_{ls}^{(s)}$  should not exceed some upper threshold value,  $g_{ls}^{(s)} < g_{\max}(g_{ll})$ , which guaranties that (i) the static friction force, i.e., the force needed to initiate sliding, is small, and (ii) once the steady motion under driving sets up, the condition  $T_0(g_{ll}, g_{ls}^{(s)}) < T_m(g_{ll})$  is satisfied, or otherwise the system will be unstable already under infinitesimally small driving.

On the other hand, the system should not be too “hard”, in order to be able to develop two substantially different temperatures  $T_{\parallel}(g_{ll}, g_{ls})$  and  $T_{\perp}(g_{ll}, g_{ls})$  at the conditions of the “melting” experiments. We always can introduce an effective time scale for kinetic energy transmission from a substrate to a lubricant as  $\tau_{ls} \propto 1/\sqrt{g_{ls}}$ , and the time corresponding to the lubricant equilibration as  $\tau_{ll} \propto 1/\sqrt{g_{ll}}$ . Note, that here we only discuss the total  $g_{ls}$ , which can vary depending on the driving conditions, while the quantities  $g_{ls}^{(s)}$  and  $g_{ls}^{(d)}$  that play essentially different roles in real systems, and whose definition depends on the particular experiment setup, will be considered in detail elsewhere.<sup>2</sup> It is important to stress now only that in the too “hard” lubricant, the full interaction satisfies  $g_{ll} \geq g_{ls}$  at all driving strengths  $g_{ls}^{(d)}$ , so that the times  $\tau_{ls}$  and  $\tau_{ll}$  are comparable ( $\tau_{ll} \leq \tau_{ls}$ ), the friction leads to effective isotropic heating and results in the first-order melting transition, which is similar to the bulk case.<sup>3</sup> On the other hand, if the lubricant is “soft” enough, so that  $g_{ll} \ll g_{ls}$  at some driving conditions, then the kinetic energy transferred from a substrate is mainly associated with longitudinal motion for a long time, until transverse equilibration occurs due to mutual lubricant interactions on the time scale  $\tau_{ll} \gg \tau_{ls}$ .

To summarize, we state, that for an intermediate “hardness” lubricant, if at some point the effective *longitudinal* temperature  $T_{\parallel}$  reaches a melting threshold  $T_m$ , while the *transverse* temperature  $T_{\perp}$  does not, then we would observe a two-stage phase transition similar to the one described above for the idealized system. Furthermore, below we argue that at least the first stage (effective 1D melting) would be a second-order (continuous) phase transition, in contrast to the classical picture of liquid–solid transitions.

For distinctness, we consider the first stage (quasi-1D melting) of a two-stage phase transition. For convenience, we consider the solidification process instead of the melting one. Let  $\rho_0 = \text{const}$  be the density of the 1D liquid. At the point of phase transition it becomes  $\rho(z) = \rho_0 + \delta\rho(z)$  (we assume sliding in the  $z$ -direction), where  $\delta\rho(z)$  is the natural order parameter of the system. Let us expand in plane waves as  $\delta\rho(z) = \sum_q \xi_q \exp(iqz)$ , where  $q$  are the reciprocal lattice vectors of the quasi-1D crystal along the  $z$ -direction. In the spirit of the original Landau theory [11,12], we suppose that the free energy  $\Phi$

<sup>2</sup> For example, the driving strength  $g_{ls}^{(d)}$  may depend on such other system parameters, as the phenomenological damping  $\eta$  and driving velocity  $v_d$  introduced for a particular model later.

<sup>3</sup> Actually, this has been the case for various effects studied for a long time as discussed, for example, in Refs. [3,6,9]. We do not discuss them here in order to keep the physical picture simple.

can be expanded with respect to the order parameter as follows:

$$\begin{aligned} \Phi(P, T_{\parallel}, \zeta) = & \Phi_0 + \sum_q \alpha_q \zeta_q e^{iqz} + \sum_{q_1 q_2} A_{q_1 q_2} \zeta_{q_1} \zeta_{q_2} e^{i(q_1+q_2)z} \\ & + \sum_{q_1 q_2 q_3} C_{q_1 q_2 q_3} \zeta_{q_1} \zeta_{q_2} \zeta_{q_3} e^{i(\sum_j^3 q_j)z} \\ & + \sum_{q_1 q_2 q_3 q_4} D_{q_1 q_2 q_3 q_4} \zeta_{q_1} \zeta_{q_2} \zeta_{q_3} \zeta_{q_4} e^{i(\sum_j^4 q_j)z} + \dots, \end{aligned} \quad (1)$$

where the coefficients  $\alpha$ ,  $A$ ,  $C$ , and  $B$  are functions of the pressure  $P$  and characteristic longitudinal temperature  $T_{\parallel}$ . Obviously, the expansion (1) can only contain terms for which  $\sum_i q_i = 0$  to ensure the translational symmetry of the free energy  $\Phi(P, T_{\parallel}, \zeta)$ . It is easy to show (see, e.g., [12]) that if the states with  $\zeta_{q_i} = 0$  and  $\zeta_{q_i} \neq 0$  differ from each other by their symmetries, then  $\alpha_q \equiv 0$ . In the symmetric phase, the minimum of the free energy corresponds to  $\zeta_{q_i} = 0$ , in which case it is necessary to have all the coefficients  $A_{q,-q}$  as positive. On the other hand, in the non-symmetric phase, the minimum of  $\Phi(P, T_{\parallel}, \zeta)$  is reachable for  $\zeta_{q_i} \neq 0$ , which is possible only if  $A_{q,-q} < 0$ . This means that at least one of the coefficients  $A_{q,-q}$  must turn zero at the transition point. Suppose this happens for some  $q = q_c$ ; then at the transition point all the terms  $\zeta_{q_i} = 0$  except  $\zeta_{q_c}$ . In other words, the expansion (1) contains only the plane waves with one definite wavelength  $q_c$ . However, this necessarily means that all the coefficients vanish ( $C_{q_1 q_2 q_3} = 0$ ) at the transition point, because there is no way to satisfy the condition  $\sum_i q_i = 0$  with only the vectors  $q_c$  and  $-q_c$ . Therefore, following the general theory [11,12], the phase transition in such a system is second order, in contrast to ordinary bulk melting, which is always first order.

To illustrate these points, we have chosen a simple enough model, which still bears the essential features of the frictional driving. Namely, we consider a 2D layer of atoms (lubricant) embedded between two (top “t”, and bottom “b”) isotropic substrates, moving in opposite directions along  $z$  with the velocities  $\mathbf{v}_t = (+v_d/2, 0)$  and  $\mathbf{v}_b = (-v_d/2, 0)$ . The energy of the isotropic substrate potential is given by  $V(z, y) = \varepsilon/4[3 - \cos(\mathbf{k}_{-1} \cdot \mathbf{r}) - \cos(\mathbf{k}_2 \cdot \mathbf{r}) - \cos((\mathbf{k}_{-1} + \mathbf{k}_2) \cdot \mathbf{r})]$ , where  $\mathbf{k}_{-1} = (2\pi/a_z, -\pi/a_y)$  and  $\mathbf{k}_2 = (0, 2\pi/a_y)$  are two reciprocal vectors of a triangular lattice, and we have chosen  $\varepsilon = 2$ ,  $a_z = 2\pi$ , and  $a_y = \sqrt{3}/2a_z$ . This simple function gives isotropic minima organized into the triangular lattice and separated by an isotropic energy barrier of height  $\varepsilon$ . The frequency of the atomic vibrations near the minima of the potential is also isotropic,  $\omega_z = \omega_y = \omega_0 \equiv (\varepsilon/2m)^{1/2}(2\pi/a_z) = 1.0$  (here  $m = 1$  is the atomic mass). The motion of atoms is governed by the Langevin equations,  $m\ddot{\mathbf{r}}_i + m\eta\dot{\mathbf{r}}_i + \partial_{\mathbf{r}_i} V_t(\mathbf{r}_i - \mathbf{v}_t t) + \partial_{\mathbf{r}_i} V_b(\mathbf{r}_i - \mathbf{v}_b t) + \partial_{\mathbf{r}_i} \sum_{j \neq i} V_i(|\mathbf{r}_i - \mathbf{r}_j|) = \delta\mathbf{F}_i$ , where  $\mathbf{r}_i = (z_i, y_i)$  for the  $i$ th atom, and  $\delta\mathbf{F}_i$  is the Gaussian random force with the correlation function  $\langle \delta F_i^{(\alpha)}(t) \delta F_j^{(\beta)}(t') \rangle = 2\eta \times mk_B T_L \delta_{\alpha\beta} \delta_{ij} \delta(t - t')$ .<sup>4</sup>

We assume an exponential interaction between the atoms corresponding to the repulsion of atomic cores,  $V_i(r) = V_0 e^{-\beta r}$ , where  $r$  is the distance,  $\beta$  determines the interaction range (in the simulation, we use  $\beta = 1/\pi$  so that a “diameter” of the atom is  $2\pi$ ). The strength of this interaction is taken as  $V_0 = (\pi e)^2$ , which gives the characteristic frequency of the vibrations associated with the interatomic interactions  $\omega_{\text{int}}^2 \equiv V_i''(2\pi) = \omega_z^2 = 1.0$ . In terms of the previously introduced general parameters, this model setup corresponds to an intermediate case between very “hard” and very “soft” lubricant,  $g_{ls}^{(s)} = 0$ ,  $g_{ll} = \omega_z^2 = 1$ , and with a moderate variation of  $g_{ls}^{(d)}$  (the latter is the stronger, the higher is the value of  $V_0$ ). The periodic boundary conditions are imposed in both the  $z$ - and  $y$ -directions in order to fix the atomic density, chosen to correspond to the perfect monolayer (one atom per potential well).

We study normally two systems, with  $16 \times 18 = 288$  lubricant atoms, and with  $32 \times 36 = 1152$  lubricant atoms. This allows us to estimate possible size effects and to see whether or not the system’s size affects qualitatively our concepts. In the selected runs we check for the absence of the finite-size effects with a system four times as large,  $64 \times 72$  atoms.

In the simulation, we first look for the minimum-energy state of the lubricant layer (perfect triangular lattice) when the two substrates are immobile and unshifted with respect to each other. Then, starting from a very high value of the driving speed,  $v_d > 10$  (where the substrates move with a rate much faster than characteristic system frequencies, so that the lubricant film is basically free, decoupled from the substrates, i.e.,  $g_{ls}^{(d)}$  is small), we adiabatically decrease  $v_d$  (thus increasing  $g_{ls}^{(d)}$ ). In the free or “perfect sliding” state at high  $v_d$ , the weakly-oscillating lubricant layer still has a perfect triangular structure, imposed by repulsive interaction and periodic boundary conditions. However, this state becomes unstable due to the first-order parameter resonance [13] at a certain value of the driving velocity  $v_{\text{crit}}$  that depends, in particular, on the value of the damping constant of the system  $\eta$  (see [13] and references therein). At  $v_d < v_{\text{crit}}$ , the fluctuations of the atoms start to grow, and finally the perfect structure of the layer becomes disordered.

While the details of the instabilities leading to disordering is of importance here, it is the disordering process itself that we monitor. In order to do so, we calculate the structure factor  $S(\mathbf{k}) = \langle \sum_{i,j} \exp(i\mathbf{k}[\mathbf{r}_i(\mathbf{t}) - \mathbf{r}_j(\mathbf{t})]) \rangle$  (here  $\langle \dots \rangle$  stands an average over time) for three reciprocal vectors  $\mathbf{k}$  of the triangular structure, which may be attributed to the longitudinal (along atomic rows in the  $z$ -direction)  $\mathbf{k}_{\pm 1} = (2\pi/a_z, \pm\pi/a_y)$  and transversal  $\mathbf{k}_2 = (0, 2\pi/a_y)$  orders in the lubricant layer, respectively. The results are shown in Fig. 1 for different values of the phenomenological constant  $\eta$ . This constant in the Langevin equations describes the rate with which the energy is taken away from the lubricant layer by the substrates. Therefore, one might expect that different values of  $\eta$  will re-

<sup>4</sup> The Langevin temperature  $T_L$  plays an auxiliary role in our simulation, where we have taken it to be *small*,  $T_L = 0.05$ , compared to the melting tem-

perature of the lubricant  $T_m$  and to the typical temperatures produced by the driving itself, just to avoid the locking of the system in the metastable states.

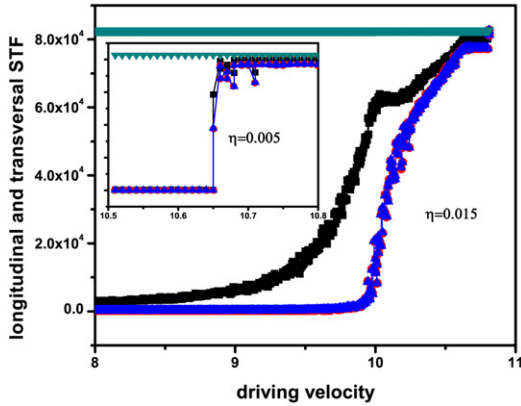


Fig. 1. The system with  $16 \times 18$  lubricant atoms. Heights of the longitudinal  $\mathbf{k}_{\pm 1} = (2\pi/a_z, \pm\pi/a_y)$  (blue and red colors) and transversal (black color)  $\mathbf{k}_2 = (0, 2\pi/a_y)$  structure factor peaks for the triangular structure of the lubricant layer versus the driving velocity  $v_d$ , with a damping of  $\eta = 0.015$ . The same data for  $\eta = 0.005$  are shown on the inset. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this Letter.)

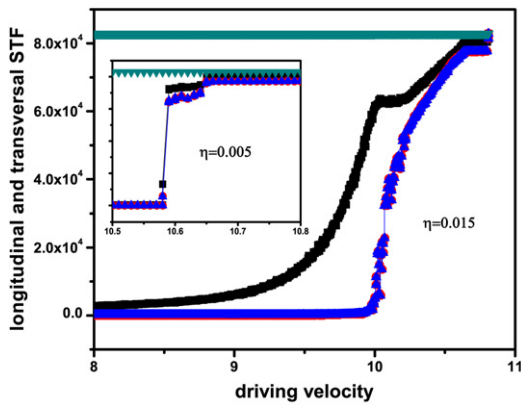


Fig. 2. The same as in Fig. 1, but for the system with  $32 \times 36$  lubricant atoms.

sult in different scenarios of the redistribution of the energy of driving across different degrees of freedom in the lubricant.

Figs. 1 and 2 show that the two-stage melting does occur at the damping value of  $\eta = 0.015$ . Here we plot the structure factor peak heights, that correspond to the “longitudinal” and “transverse” order parameters in the lubricant film, versus the decreasing  $v_d$  (i.e., versus increasing effective driving  $g_{ls}^{(d)}$ ). One can see that the disordering along the direction of driving occurs earlier and, furthermore, both peaks decrease smoothly with  $v_d$ , which indicates continuous second-order phase transitions. At  $v_d \approx 10$ , when the lubricant film becomes completely disordered along the  $z$ -direction, the slope of the transverse order parameter versus  $v_d$  increases, which indicates that the driving energy is not spent anymore on the longitudinal disordering, but goes completely to disorder the system in the transverse direction. By contrast, at a lower value of the damping constant  $\eta = 0.005$ , we observe *sharp and simultaneous* melting both along the  $z$ -direction and transversally along the  $y$ -direction. Noticeably, this sharp transition occur for both sys-

tems within the same very small range, that is an indication of size independence of the transition concept. The effect of the finite size only shows in the value of driving velocity at the transition point.

In conclusion, we propose here a possible scenario of the order–disorder transition in a thin lubricant film subjected to sliding friction. We demonstrate a theoretical possibility for the novel two-stage melting mechanism in such a system. Furthermore, we argue that under certain conditions, the quasi-1D melting appears to be a continuous second-order transition, instead of first order as occurs in 3D systems. To illustrate our point, we have carried out Langevin MD simulation of a simple system, which support our conclusions. However, we have to emphasize that our results must be treated prudently. Namely, (i) in a strict sense, we cannot speak about the phase transition in its usual sense because the frictional systems are out of equilibrium; and (ii) the systems we study are simplistic models, so that the further investigations with more realistic systems are definitely needed.

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