Comment on “Freezing by heating in a driven mesoscopic system”

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In a recent Letter Helbing et al. [1] found a fascinating fluctuation-induced ordering, which they dubbed “freezing by heating”, in a nonequilibrium model system of particles confined to a 2d narrow strip and driven to move in opposite directions: counter flowing chains of particles jam up and crystallize when sufficiently large noise is introduced into the system. In this Comment we point out that this kind of fluctuation-induced ordering is not an intrinsically nonequilibrium effect, and that it can be expected to occur in a variety of equilibrium systems, with reentrant order appearing as a function of temperature. We argue that this is a consequence of a familiar Helfrich-like interaction that had been used to understand a variety of phenomena ranging from the lamellar phase of fluctuating membranes to magnetization curves of a vortex liquid state of type II superconductors.

We demonstrate the mechanism behind reentrant melting with an equilibrium model of 2d colloids subjected to a 1d periodic potential, that received considerable attention recently due to the experimental discovery of isothermal melting that is reentrant as a function of strength of the periodic potential (produced by two interfering laser beams) [2], the so-called reentrant “laser-induced melting” (RLIM).

Since at all $T$ the orientational symmetry and translational symmetry along $y$ are explicitly broken by the 1d periodic potential with troughs running along $x$ and spacing $a = 2\pi/q$, the only remaining feature distinguishing a crystal from the modulated liquid is the spontaneous translational order along the troughs, characterized by quasi-long ranged $u_x$ phonon correlations and a finite shear modulus $\mu_{xy}$.

The mechanism behind RLIM can be understood as follows. Just above melting temperature $T_m(V_q)$, the dominant effect of increasing a weak periodic potential $V_q$ is to suppress thermal fluctuations, thereby leading to Lifshitz freezing [3]. In contrast, for large $V_q$, the suppression of the out-of-trough $u_y$ fluctuations has the dominant effect of reducing the inter-row interactions, thereby decoupling the particles in the neighboring rows, reducing the effective shear modulus $\mu_{xy}(T, V_q)$, and leading to RLIM [4].

Since it is clearly the ratio $T/V_q$ that determines the extent of the fluctuation-driven inter-row interaction, it follows that at large fixed $V_q$ increasing $T$ will increase the inter-row interaction and the modulus $\mu_{xy}(T, V_q)$. Because $T_m$ is typically a monotonically increasing function of the shear modulus, thermal enhancement of shear modulus implies an equilibrium analog of the “freezing by heating” transition discovered by Helbing, et al. [1].

Above qualitative argument can be formalized by an explicit calculation. Although $T$ dependence of both the bulk and the shear modulus maybe important and can be studied in full detail [5], in order to illustrate the effect, it is sufficient to focus on the effective shear modulus $\mu_{xy}(T, V_q)$. Clearly this modulus is proportional to the interaction potential between nearest particles in adjacent rows, typically of Debye-screened form $U_D \approx e^{-(a+u_x)/\xi}$ (though only its short-range nature is important), where $\xi$ is the screening length. Since phonon fluctuations $u_y$ transverse to the troughs are “optical” (i.e., “massive”, as opposed to the acoustic $u_x$ phonon) we can average over fast $u_y$ fluctuations obtaining an effective shear modulus $\mu_{xy}(T, V_q) \approx \mu_0 e^{(u_x^2)/2\xi^2} \approx \mu_0 e^{T/V_q}$ that clearly increases with $T$. We can then compute $\langle u_x^2 \rangle \approx T/\mu_{xy}(T, V_q) \approx T e^{-T/V_q}/\mu_0$ and use Lindemann criterion $\langle u_x^2 \rangle = L^2$ to estimate $T_m(V_q)$. This clearly gives two roots for $T_m$, with the lower corresponding to the conventional melting and the upper to the freezing by heating (see figure). At sufficiently high $T$, we expect above elastic calculation to become inaccurate and for the fluctuation enhancement of $\mu_{xy}(T, V_q)$ to saturate, leading to re-melting into a modulated liquid.

While extending above analysis to the nonequilibrium case studied by Helbing, et al. [1] remains an interesting and challenging problem, we believe that fluctuation-driven reentrant freezing mechanism described above is also at play in their system.

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References:

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[4] L. Radzihovsky, et al., Phys. Rev. E 63, 31503 (2001).
[5] Qualitative increase of $\mu_{xy}(T)$ with $T$ holds beyond this harmonic approximation.