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How to cite

CUGLIANDOLO, Leticia F., GIAMARCHI, Thierry, LE DOUSSAL, Pierre. Dynamic Compressibility and Aging in Wigner Crystals and Quantum Glasses. In: Physical review letters, 2006, vol. 96, n° 21. doi: 10.1103/PhysRevLett.96.217203

This publication URL: https://archive-ouverte.unige.ch/unige:36125

Publication DOI: 10.1103/PhysRevLett.96.217203

Dynamic Compressibility and Aging in Wigner Crystals and Quantum Glasses

Leticia F. Cugliandolo,[1,2] Thierry Giamarchi,[3] and Pierre Le Doussal[2]

[1]*Laboratoire de Physique Théorique et Hautes Energies, 4 Place Jussieu, 75252 Paris Cedex 05, France*
[2]*LPTENS CNRS UMR, 8549 24, Rue Lhomond 75231 Paris Cedex 05, France*
[3]*University of Geneva, DPMC, 24 Quai Ernest Ansermet, CH-1211 Geneva 4, Switzerland*

(Received 19 November 2005; published 1 June 2006)

We study the nonequilibrium linear response of quantum elastic systems pinned by quenched disorder with Schwinger-Keldysh real-time techniques complemented by a mean-field variational approach. We find (i) a quasiequilibrium regime in which the analytic continuation from the imaginary-time replica results holds provided the marginality condition is enforced, and (ii) an aging regime. The conductivity and compressibility are computed. The latter is found to cross over from its dynamic to static value on a scale set by the waiting time after a quench, an effect which can be probed in experiments in, e.g., Wigner glasses.

DOI: 10.1103/PhysRevLett.96.217203 PACS numbers: 75.10.Nr, 71.55.Jv, 72.20.−i

The dynamics and transport properties of glasses are the object of current theoretical and experimental interest [1–3]. The slow approach to the static limit and the accompanying aging phenomena [1] observed experimentally in classical glasses is captured by a variety of models [2]. A natural explanation for the slow dynamics is the existence of a special organization of an exponentially large number of metastable states.

Less is known when glasses evolve at very low temperatures and quantum fluctuations become important as occurs in Wigner [4,5] and Coulomb [6–10] glasses, as well as in spin [11] and other systems [12] at low temperature. Aging in the transport properties of Coulomb glasses was reported recently [6]. Theoretically, electronic glasses have been studied with numerical simulations in a classical limit [7] or with the imaginary-time [8] or static [9,10] methods combined with the replica trick applied to the isolated model. Strictly, the latter allows one to describe the statics and, via an analytical continuation to real time, the equilibrium dynamics with infinitesimal dissipative coupling to a bath. However, it is of a somehow broader use since it has also given access to dynamical quantities such as the finite frequency conductivity [13–15], and instanton calculations allowed us to relate, in the presence of a bath, the imaginary-time solution to the ultraslow dynamics (quantum creep) [16]. Still, a rigorous treatment of a full nonequilibrium relaxation requires special techniques devised to deal directly with the real-time dynamics of dissipative quantum systems [17,18]. Even in the infinitesimal coupling limit, these are needed to ascertain the validity of analytic continuations to real-time, especially for glassy systems.

The compressibility of electronic glasses has been measured recently [19–21]. Being of thermodynamic nature, in equilibrium it should only depend on the statics of the problem. Conversely, its time dependence reflects an out of equilibrium relaxation. Within an imaginary-time (Matsubara) variational calculation [13,14,22] the static, zero frequency, compressibility of a disordered Wigner crystal was found to be nonzero and identical to the one of the pure system. In contrast, the analytic continuation yielded a vanishing result even in the small real frequency limit. This hinted to the fact that aging effects could be present [5,22] though a firm conclusion was clearly beyond the imaginary-time calculation that assumes equilibrium at the outset.

In this Letter we study the out of equilibrium relaxation of a disordered Wigner crystal with the Schwinger-Keldysh (SK) technique [23,24]. We find two two-time regimes; one in which the equilibrium result is recovered and another one in which the compressibility is reduced and dominated by aging effects. Our calculation is performed within a mean-field-like variational approximation. We discuss its limits of validity as well as the relevance of our results for experimental systems.

We model a disordered quantum crystal as

$$H = \int_x \left\{ \frac{\hbar^2 \Pi^2(x)}{2m\rho_0} + \frac{c}{2}[\nabla u(x)]^2 \right\} - \int_x U(x)\rho_0 \cos\{Q[x - u(x)]\}, \quad (1)$$

where m is the mass of the particles, ρ_0 is the average density, $Q \equiv 2\pi/a$ with a the interparticle spacing, and $\int_x \equiv \int d^d x$. Π and u are conjugate operators $[u(x), \Pi(x')] = i\hbar\delta(x - x')$. $U(x)$ is a random potential with Gaussian statistics, $\overline{U(x)} = 0$ and $\overline{U(x)U(x')} = \Delta(x - x')$ with $\Delta(z)$ a function with finite range r_f. This model describes a large class of systems including charge density waves [25,26] (with a phase $\phi = 2\pi u/a$), Wigner crystals (upon generalization to a two component vector $\vec{u}$) [5,14], and Luttinger liquids in $d = 1$ [27].

The compressibility κ is defined as the change in density in response to a change in chemical potential μ. In linear response, it is given by the $q \to 0$ limit of the density-density correlator. For (1) the long wavelength part of the

density is $\rho(x) - \rho_0 \sim -\rho_0 \nabla u(x)$, and the equilibrium compressibility is $\kappa = \lim_{q\to 0}\kappa(q, \omega_n = 0)$ with $\kappa(q, \omega_n) = \rho_0^2 q^2 G_c(q, \omega_n)$, ω_n the Matsubara frequencies, and $G_c(q, \omega_n) = \overline{\langle u^*_{q,\omega_n} u_{q,\omega_n}\rangle - \langle u^*_{q,\omega_n}\rangle\langle u_{q,\omega_n}\rangle}$, where $\langle \ldots \rangle$ and $\overline{\cdots}$ denote the thermal and disorder average, respectively. Within the replica variational approach [13]

$$\kappa(q, \omega_n) = \frac{\rho_0^2 q^2}{\rho_m \omega_n^2 + cq^2 + \Sigma_1(1 - \delta_{n,0}) + I(\omega_n)}, \quad (2)$$

where $\rho_m = m\rho_0$, $\Sigma_1 \sim \rho_m \omega_p^2$ is a constant depending on disorder, ω_p is the pinning frequency [13,26], and $I(0) = 0$. κ is *independent of disorder* and simply given by

$$\kappa = \rho_0^2/c. \quad (3)$$

Alternatively, the real-time compressibility, i.e., the response to a time-dependent chemical potential, is given by the retarded linear response. Naively, this can be obtained from (2) by the standard analytical continuation $i\omega_n \to \omega + i\delta$ that leads to [13]

$$\kappa(q, \omega) = \rho_0^2 q^2 [-\rho_m \omega^2 + cq^2 + \Sigma_1 + \tilde{I}(\omega)]^{-1}, \quad (4)$$

where $\tilde{I}(\omega \to 0) \to 0$. Note that even performing the limit $\omega \to 0$ *first* while keeping q fixed one finds

$$\kappa = \lim_{q\to 0}\lim_{\omega\to 0} \kappa(q, \omega) = 0, \quad (5)$$

in disagreement with the result in (3).

The simplest example where such a difference arises is an isolated two level system (e.g., a spin). The static susceptibility in response to an external magnetic field is $\chi = 1/T$, where T is the temperature ($k_B = 1$). However, the response to a time-dependent field is always zero, leading to $\chi(\omega \to 0) = 0$ at variance with the static result. The difference is due to the existence of two degenerate ground states in the unperturbed system. In the static calculation one sums over both. The perturbation instead is unable to induce transitions leading to zero response. In a glass there may be no exact degeneracy between the multitude of metastable states. Still, if they are close enough in energy and their coupling is sufficiently weak one expects a similar phenomenon.

To analyze this issue, we study the real-time dynamics of model (1) using the SK technique. Let us sketch the main steps of the calculation. The observables are computed using a path integral on two fields, $u_\pm$, that live on the two sides of a closed time contour. The measure is given by e^{-S_K} where S_K is the (dimensionless) SK action $S_K = \frac{i}{\hbar} \times [S(u_+) - S(u_-)]$ and $S(u)$ is the standard action for a system described by (1). To take into account nonzero temperature and dissipation we couple the system to a thermal bath of independent harmonic oscillators [17]. By integrating them out we induce a coupling between the fields $u_\pm$. The average over disorder is easily done without replicas and has a similar (coupling) effect. In terms of the fields $u \equiv \frac{1}{2}(u_+ + u_-)$ and $\hat{u} \equiv \frac{1}{\hbar}(u_+ - u_-)$ the resulting action, $S_{\rm av} = S_0 + S_d$, depends on $\mathsf{R}(z) = \rho_0^2 \Delta_Q \cos(Qz)$ with Δ_Q the Fourier transform of the disorder correlation at wave vector Q, and reads

$$\begin{aligned} S_0 &= \frac{1}{2}\int_{q,\omega} (\, u^*_{q,\omega} \quad i\hat{u}^*_{q,\omega}\,) \\ &\quad \times \begin{pmatrix} 0 & -\rho_m\omega^2 + cq^2 + i\eta\omega \\ -\rho_m\omega^2 + cq^2 - i\eta\omega & -\eta\hbar\omega\coth(\frac{\hbar\omega}{2T}) \end{pmatrix} \\ &\quad \times \begin{pmatrix} u_{q,\omega} \\ i\hat{u}_{q,\omega} \end{pmatrix}, \\ S_d &= \frac{1}{2\hbar^2}\int_{xtt'} \sum_{\sigma=\pm1,\sigma'=\pm1} \sigma\sigma' \mathsf{R}\Big[u_{xt} - u_{xt'} \\ &\quad + \frac{\hbar}{2}(\sigma\hat{u}_{xt} - \sigma'\hat{u}_{xt'})\Big], \end{aligned} \quad (6)$$

with $\int_{q\omega} \equiv \int \frac{d^d q}{(2\pi)^d} \int \frac{d\omega}{2\pi}$. The terms proportional to η arise from the coupling to the bath and represent Ohmic dissipation. For η infinitesimal, one recovers the intrinsic dynamics of the quantum system. For the pure system ($\mathsf{R} = 0$) this action yields the equilibrium response obtained by the analytic continuation of the Matsubara representation. S_d can be rewritten as $S_d = \frac{2\rho_0^2\Delta_Q}{\hbar^2} \times \int_{xtt'} \sin(Q\hbar\hat{u}_{xt}/2)\sin(Q\hbar\hat{u}_{xt'}/2)\cos[Q(u_{xt} - u_{xt'})]$; it is clear that the quantum action crosses over to the dynamic Martin-Siggia-Rose action [28] when $\hbar \to 0$.

The disorder-averaged correlation and linear response are defined as $C_{x-x'}(t, t') = \frac{1}{2}\langle u^+_{xt}u^-_{x't'} + u^-_{xt}u^+_{x't'}\rangle = \langle u_{xt}u_{x't'}\rangle$ and $R_{x-x'}(t, t') = \frac{i}{\hbar}\langle u^+_{xt}(u^+_{x't'} - u^-_{x't'})\rangle = \langle u_{xt}i\hat{u}_{x't'}\rangle$, with $R_{x-x'}(t, t')$ for $t \le t'$ and $\langle \hat{u}_{xt}\hat{u}_{x't'}\rangle$ vanishing because of causality. The brackets represent here an average with the weight $e^{-S_{\rm av}}$. In the absence of disorder, C and R are stationary, i.e., depend only on $t - t'$; their Fourier expressions are

$$\begin{aligned} C^0_q(\omega) &= \frac{\eta\hbar\omega\coth(\frac{\hbar\omega}{2T})}{(\rho_m\omega^2 - cq^2)^2 + \eta^2\omega^2}, \\ R^0_q(\omega) &= [-\rho_m\omega^2 + cq^2 - i\eta\omega]^{-1}, \end{aligned} \quad (7)$$

and they obey the fluctuation-dissipation theorem (FDT)

$$R^0_q(\omega) = -\int_{\omega'} \frac{2\tanh(\frac{\beta\hbar\omega'}{2})}{\hbar(\omega - \omega' + i\epsilon)} C^0_q(\omega'). \quad (8)$$

In particular, $\hbar\,\mathrm{Im}R^0_q(\omega) = \tanh(\beta\hbar\omega/2)C^0_q(\omega')$. Since the long wavelength part of the density and the current are, respectively, $\rho(x) - \rho_0 = -\rho_0\nabla u(x, t)$ and $j(x) = \rho_0\partial_t u(x, t)$, in linear response the compressibility and conductivity are given by

$$\kappa(q, \omega) = q^2\rho_0^2 R_q(\omega), \qquad \sigma(\omega) = -i\omega\rho_0^2 R_{q=0}(\omega). \quad (9)$$

Inserting the free values in (7) one recovers (3) for the static compressibility and $\sigma(\omega) = \rho_0^2/(\eta - i\omega\rho_m)$ for the

static conductivity which, in the limit $\eta \to 0$, reproduces the Drude form, $\mathrm{Re}\sigma(\omega) = \frac{\rho_0 \pi}{m}\delta(\omega)$.

In the presence of disorder, one derives self-consistent mean-field equations for $C_q(t,t')$ and $R_q(t,t')$, where $t=0$ is the time when the system is quenched into the disordered state and set in contact with the bath. We show only the equation for the response (see [29] for details):

$$(\rho_m \partial_t^2 + \eta \partial_t + cq^2) R_q(t,t') = \delta(t-t') - \int_0^t ds \Sigma(t,s) \times [R_q(t,t') - R_q(s,t')], \tag{10}$$

with the self-energy $\Sigma(t,s) = -\frac{4}{\hbar}\mathrm{Im}V'[\tilde{B}(t,s) + i\hbar\tilde{R}(t,s)]$ and $V(z) = -\rho_0^2 \Delta_Q e^{-(1/2)Q^2 z}$. The tilde denotes an integration over q [e.g., $\tilde{R}(t,s) = \int_q R_q(t,s)$] and $B_q(t,t') = C_q(t,t) + C_q(t',t') - 2C_q(t,t')$.

Analysis of this equation in the long time limit $t, t' \to \infty$ shows that, as in the classical case [30] and the quantum dissipative p-spin model [17,18], the model exhibits two two-time regimes. First, for fixed $t - t'$ the two-time functions are stationary, $R_q(t,t') \to r_q(t-t')$ and $B_q(t,t') \to b_q(t-t')$, and the FDT (8) holds. For more separated times, there is an asymptotic aging solution, $R_q(t,t') \to R_q^A(t,t')$, as discussed below. The equation for $r_q(t-t')$ is obtained from (10) by a careful separation of time scales along the lines of [31]. Its solution reads

$$r_q(\omega) = [cq^2 + M - i\eta\omega - \rho_m\omega^2 - \Sigma(\omega)]^{-1}, \tag{11}$$

with the self-energy $\Sigma(\omega) = \int_0^\infty d\tau (e^{-i\omega\tau} - 1)\Sigma(\tau)$ and

$$\Sigma(\tau) = \frac{2}{\hbar}\sum_{\sigma=\pm 1} i\sigma V'[\tilde{b}(\tau) + i\hbar\sigma\tilde{r}(\tau)]. \tag{12}$$

The constant $M = \lim_{t\to\infty}\int_0^t ds\Sigma(t,s) - \int_0^{+\infty} d\tau'\Sigma(\tau')$, the so-called anomaly, arises from the contribution of the aging time scales to the FDT regime. Using FDT,

$$\tilde{b}(t) = \int_\omega 2(1 - e^{-i\omega t})\hbar[1 + 2f_B(\omega)]\mathrm{Im}\tilde{r}(\omega), \tag{13}$$

where $f_B(\omega) = 1/(e^{\beta\hbar\omega} - 1)$. The last self-consistency condition follows from matching the FDT regime with the aging one. Taking $t' \to t^-$ in (10) yields $(cq^2 + M)R_q^A(t,t^-) = \Sigma^A(t,t^-)r_q(\omega = 0)$, and the existence of a nonvanishing aging solution with $R_q^A(t,t') \to 0$ as $t' \to 0$ requires the "marginality condition"

$$1 = -4V''(b_\infty)\int_q (cq^2 + M)^{-2}, \tag{14}$$

where $b_\infty = \lim_{t\to\infty}\tilde{b}(t)$. One can explicitly check [29] that the solution in the FDT regime coincides with the analytical continuation of the saddle-point solution of the replica variational approach to the Matsubara action performed in appendix D of [13]. More precisely, $G_c(q,\omega_n)$ identifies (after analytic continuation) with $r_q(\omega)$. Similarly, $I(i\omega_n \to \omega + i\delta) \to \tilde{I}(\omega) = -\Sigma(\omega)$, $\Sigma_1 \to M$, $B \to b_\infty$, where B is defined in (31) in [13]. Importantly, this correspondence holds only if the replica symmetry breaking scheme is the one using the marginality condition (14). This choice, advocated in [13] for being the only one leading to a gapless conductivity, was often used since and is hereby fully justified within the SK formalism. Note that in $d = 1$ and for $\eta = 0$ model (1) has an additional stable 1 step replica symmetry breaking solution that is then discarded in the Matsubara treatment. The SK formalism also allows one to obtain the response for finite η. The low frequency behavior of the conductivity changes from $\mathrm{Re}\sigma(\omega) \sim \rho_0\omega^2/(m\omega_p^3)$ to $\mathrm{Re}\sigma(\omega) \sim \sqrt{\eta}\omega^{3/2}/\omega_p^3$ [32].

We now turn to the compressibility, i.e., the (linear) response of the density to a small change in chemical potential of amplitude $\delta\mu$ applied between times t_w and t (see Fig. 1). One finds

$$\kappa(q;t,t_w) \equiv \left.\frac{\delta\rho(t)}{\delta\mu}\right|_{\mu=0} = \frac{\rho_0^2 q^2}{cq^2 + M(t,t_w)}. \tag{15}$$

As $t - t_w$ increases one distinguishes two regimes. For $\omega_p^{-1} \ll t - t_w \ll t_w$ the response is dominated by the "FDT regime" and it is stationary; moreover $M(t,t_w) = M$ and the compressibility vanishes [$\kappa(q;t,t_w) \to 0$ when $q \to 0$], recovering (5). If, instead, $t - t_w \sim t_w$, the response is dominated by the aging regime and $M(t,t_w) = M - 4\int_{\tilde{B}^A(t,t_w)}^{\tilde{b}_\infty} V''(z)X[z]dz$, where $X(z)$ is the FDT violation ratio [17,30,31] as a function of $z = \tilde{B}$. For η infinitesimal, $X(z)$ coincides with $X \to u$, $z \to B(u)$ as found in the replica solution with (14) [13]. $\tilde{B}$ has an aging form and its detailed scaling depends on the model. In $d = 1, 2$ one finds $M(t,t_w) \simeq MF[h(t)/h(t_w)]$, where F and h are scaling functions. When times are very separated $t - t_w \gg t_w$, $M(t,t_w)$ tends to zero and, from (15), the compressibility is the constant (3). At intermediate time scales a mass is always present and the compressibility depends on the wave vector q. For a finite size system of size L one can estimate that the compressibility crosses over from being essentially zero to the thermodynamic one when

$$(L_c/L)^2 > M(t,t_w)/M, \tag{16}$$

[$L_c = \sqrt{c/\rho_m}\omega_p$ is the Larkin pinning length]. The char-

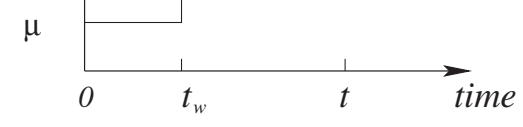

FIG. 1. Sketch of the variation of the chemical potential; dc (solid line) and ac (dashed line), for a system quenched at time $t = 0$. The density is measured at time t.

acteristic time scale to realize such evolution is the waiting time t_w. This potentially provides a direct experimental way to check for aging in these systems. Alternatively, one can apply an ac perturbation as typically done in experiments (see Fig. 1, and [1] for a similar discussion on the ac magnetic susceptibility of classical spin glasses) and obtain different results by tuning the period, τ, of the perturbation. Short τ's select the FDT contribution and thus a vanishing compressibility while very long τ's do not erase the contribution from the aging regime yielding a constant compressibility as in (3). The crossover in τ is typically of the order of t_w.

The above results are consistent with the picture that upon a change of chemical potential the system is first trapped in quasiequilibrium in a metastable state. Since these states are pinned the charge cannot fluctuate and the compressibility is essentially zero. On the other hand, if the change in chemical potential is maintained for a long time, the system explores other metastable states, the charge being allowed to change in the process, thereby leading to a finite response [33]. The mean-field solution presumably overestimates the separation between metastable states. Transitions can occur through activated processes due to quantum or thermal fluctuations (so-called creep). Although treating such processes is difficult, a possible modification of (15) is

$$\kappa(q; t, t_w) = \frac{\rho_0^2}{c} F[qL(t_w), h(t)/h(t_w)] \qquad (17)$$

with $F(0, y) = 0$, $F(x, \infty) = F(\infty, y) = 1$. Classical creep arguments [34] based on barriers growing as L^θ suggest $L(t) = L_c + h(t)$ with $h(t) = (T \ln t)^{2/\theta}$. Extensions incorporating quantum effects, as in [16], are needed to complete this picture.

In addition, the approach developed here allows one to generalize the mode-coupling theory to low-temperature glasses with quantum fluctuations and no quenched disorder (see [2] for a discussion of the classical limit).

We acknowledge interesting discussions with Z. Ovadyahu, A. K. Savchenko, D. Reichman, and A. Yacoby. This work was supported in part by the Swiss National Science Foundation under MANEP and Division II, and an ACI-France Grant. L. F. C. is a member of IUF.

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