Coulomb Disorder in Periodic Systems

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We study the effect of unscreened charged impurities on periodic systems. We show that the long wavelength component of the disorder becomes long ranged and dominates static correlation functions. On the other hand, because of the statistical tilt symmetry, dynamical properties such as pinning remain unaffected. As a concrete example, we focus on the effect of Coulomb disorder generated by charged impurities, on 3D charge density waves with non local elasticity. We calculate the x-ray intensity and find that it is identical to the one produced by thermal fluctuations in a disorder-free smectic-A. We discuss the consequences of these results for experiments.

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I. INTRODUCTION

The effect of quenched disorder on various condensed elastic systems is one of the fascinating problems in statistical mechanics. Examples of physical systems range from domain walls in magnetic and ferroelectric materials, contact lines of of a liquid meniscus on a rough substrate, crack propagation, to vortex lattices in type II superconductors, charge density waves (CDW) and Wigner crystals. In these systems, the competition between elastic interactions which tend to impose some long range order in the system and quenched disorder, leads to the formation of glassy phases. Two broad classes of elastic systems can be distinguished: random manifold systems such as domain walls, contact lines and cracks, and periodic systems such as vortex lattices, charge density waves, and Wigner crystals. The latter are characterized by a long range crystalline order in the absence of disorder and thermal fluctuations. For these systems, a crucial question is whether a weak disorder destroys entirely the crystalline order, or whether some remnants of the underlying periodic structure remain observable.

One of the earliest attempts to answer this question, was the pioneering work by Larkin on vortex lattices. Using a random-force model, he showed that due to the relevance of disorder in the renormalization group sense, long range order was entirely destroyed below four dimensions. Above four dimensions, long range order persists as disorder becomes irrelevant. A similar conclusion was reached by Sham and Patton for the case of a CDW with short range elasticity, where, using an Imry-Ma approach, they concluded that long range order was impossible in the presence of disorder below four dimensions. The problem of short range disorder in periodic systems with short ranged elasticity was revisited in Refs. It was argued that the periodicity present in systems like CDW and vortex lattices plays a pivotal role in determining the physics of the system in the presence of disorder. More precisely, it was shown that, though the disorder is relevant below four dimensions, due to the underlying periodicity of the system a quasi long-range order persisted for dimensions between two and four. This is in stark contrast to the earlier results which predicted a total destruction of order. The resulting phase, nicknamed Bragg glass phase, possesses both quasi long range order and metastability and glassy properties. It was further shown that the Bragg glass phase is stable to the formation of defects. Recent neutron scattering experiments on vortex lattices have furnished clear evidence for the existence of such a phase.

A complication arises in charged periodic systems due to the Coulomb repulsion, which renders the elasticity non-local. This non local elasticity tends to rigidify the system, so that short range correlated disorder could be irrelevant in dimension smaller than four. For instance, within the random force model, the correlation function of the displacement in three dimensions displays a logarithmic growth indicating quasi-long range order. In fact, when the periodic structure of the CDW is properly taken into account, the growth of the displacement correlation function is even weaker, increasing only as \( \log(\log(r)) \) with the distance \( r \). A second complication arising from Coulomb interaction is that the disorder induced by charged impurities has long-range correlations. This type of disorder can exist in certain doped CDW materials such as \( \text{KMo}_{1-x}\text{V}_x\text{O}_3 \).

In this paper, we study the effect of the competition of the non-local elasticity produced by the Coulomb interaction with the long-range random potential resulting from the presence of charged impurities on the statics. The paper is organized as follows: in Sec. we introduce a decomposition of the Coulomb potential on the Fourier modes of the periodic structure. With this decomposition, we show that only the long-wavelength component...
of the random potential i.e., forward scattering disorder, possesses long range correlations. Using statistical tilt symmetry, we deduce that due to the short ranged nature of the backward scattering terms engendered by the disorder, the dynamical properties in the presence of charged impurities are not qualitatively different from those in the presence of neutral short ranged impurities. In Sec. III, we consider the problem of the CDW system and we derive the non-local elastic Hamiltonian. In Sec. IV we derive the static displacement correlation functions and x-ray intensity of the CDW with charged impurities and we highlight the similarity of the latter to the x-ray intensity of smectic-A liquid crystals subjected to thermal fluctuations. In Sec. V we discuss the existence of charged impurities, the original charge density on the scale of a lattice spacing. The low energy physics of the periodic structure can be described directly by solving the Poisson equation with a Gaussian integration: $\rho_0(r) = \rho_0(1 - \nabla \cdot u)$ describes the density fluctuation arising from the long wavelength deformation of the periodic structure and $\rho_0$ is the average density. In the second term, the vectors $G$ belong to the reciprocal lattice of the perfect periodic structure, and $u(r)$ represents a slowly varying elastic deformation of the structure. The quantities $e^{iG \cdot (r - u(r))}$ describe fluctuations of the density on the scale of a lattice spacing. The low energy physics of the periodic structure can be described in terms of a purely elastic Hamiltonian which has the generic form for isotropic systems

$$H_0 = \int \frac{\alpha}{2} (\nabla u)^2$$

where $\alpha$ is the elastic coefficient and $\int d^3r$ is a shorthand for $\int d^3r$. This form can easily be generalized to anisotropic systems. Well known examples of charged periodic structures are the Wigner crystal of charged colloidal crystals and the charge density waves. In many charged systems, unscreened Coulomb interactions are present:

$$H_C = \frac{e^2}{2} \int_{r,r'} \frac{\rho(r)\rho(r')}{4\pi\epsilon|r - r'|}$$

and strongly affect the elasticity and dispersion of the compression modes of the system. Moreover, in the presence of charged impurities, the original charge density on the lattice interacts with the charge impurity yielding:

$$H_{\text{dis}} = e^2 \int_{r,r'} \frac{\rho(r)\rho_{\text{imp}}(r')}{4\pi\epsilon|r - r'|}$$

where $\rho_{\text{imp}}$ denote the impurity density. Using the decomposition of the density, we now show that the Coulomb interactions fundamentally modify only the long wavelength components of the elasticity and of the disorder energy.

To better handle the periodicity of the elastic structure, it is convenient to use the decomposition of the Coulomb interaction in terms of the reciprocal lattice vectors $G$. In three dimensions, this decomposition reads

$$\frac{1}{4\pi|\mathbf{r}|} = \int \frac{d^3q}{(2\pi)^3} \frac{e^{i\mathbf{q} \cdot \mathbf{r}}}{q^2}$$

where

$$V_G(r) = \int_{BZ} \frac{d^3q}{(2\pi)^3} \frac{e^{i\mathbf{q} \cdot \mathbf{r}}}{(q + G)^2},$$

and $\int_{BZ}$ indicates that the integral is restricted to the first Brillouin zone. It is straightforward to check that $V_{-G}(r) = V_G(r)$. Using Eq. (5), the interaction term $H_C$ can be rewritten as:

$$H_C = \frac{e^2}{2\epsilon} \sum_{G \neq 0} \int_{r,r'} V_G(r - r') e^{iG \cdot (u(r) - u(r'))} + \frac{e^2}{2\epsilon} \int_{r,r'} \rho_0(r)\rho_0(r')$$

Note that due to the slow variation of $u(r)$, terms involving the oscillatory factors $e^{i(G - G') \cdot r}$ can be dropped from the interaction. Let us first consider the term involving long wavelength fluctuations of the density. Since we are interested only in the long wavelength properties, we can replace the integration over the Brillouin zone in $V_0(r)$, by a Gaussian integration:

$$\int_{BZ} \frac{d^3q}{(2\pi)^3} \rightarrow \int \frac{d^3q}{(2\pi)^3} e^{-q^2a^2},$$

with the parameter $a$ chosen so that $\pi/a \sim |G_{\text{min}}, G_{\text{min}}$ being the reciprocal lattice vector having the shortest length. In this case, $V_0(r)$ can be obtained indirectly by solving the Poisson equation with a Gaussian charge density and is found to be

$$V_0(r) = \frac{1}{4\pi\epsilon} \text{erf} \left( \frac{r}{2a} \right),$$

In the limit $r \gg a$, we recover the known result $V_0 \sim 1/(4\pi r)$. Clearly, the non-oscillating component of the
Coulomb potential remains long-ranged and tends to rigidify the system.

It now remains to be seen whether the oscillating parts of the Coulomb interaction specified by \( V_G \) for \( G \) are long-ranged or not. We first note that the above trick of replacing the integration over the Brillouin zone by a Gaussian integral over the entire space is not applicable anymore, as it would introduce a spurious integration over a region where \( G + q = 0 \). This would result in an (incorrect) \( 1/r \) behavior of \( V_{G\neq 0}(r) \). To obtain a correct estimate for \( V_G \) we replace the integral over the Brillouin zone by

\[
\int_{BZ} \frac{d^3q}{(2\pi)^3} \to \int \frac{d^3q}{(2\pi)^3} F_{BZ}(q, \epsilon),
\]

where \( F_{BZ}(q, \epsilon) \) is an indefinitely derivable function with a compact support contained in the first Brillouin zone (see App. A for an explicit form of \( F_{BZ}(q, \epsilon) \)). Obviously, for \( G \neq 0 \), \( F_{BZ}(q, \epsilon)/|q + G|^2 \) is also an indefinitely differentiable function of compact support. A well-known theorem\(^{39,40}\) then shows that the Fourier transform of \( F_{BZ}(q, \epsilon)/|q + G|^2 \) is indefinitely differentiable and for \( r \to \infty \) is \( o(1/r^n) \) for any \( n > 0 \). This implies that the function \( V_G(r) \) is short ranged. Incorporating the above results in Eq. (7), we see that while the non oscillating terms containing \( U \) of the Coulomb interaction modifies the long wave-length behavior of the elasticity, rendering it non-local, the short ranged nature of the oscillatory terms merely renormalizes the elastic coefficients. This is explicitly shown in Eq. (10) for the particular case of a CDW. The resulting non-local character of the elastic interactions modifies strongly the static and dynamic properties of the system.\(^{27,29,41,42}\)

To understand the nature of the interaction with the charged impurities, we use the above procedure to rewrite the random potential generated by the impurities as:

\[
U_{\text{imp.}}(r) = \int_{BZ} \frac{d^3q}{(2\pi)^3} e^{iG \cdot r} \rho_{\text{imp.}}(q),
\]

\[
= \sum_G e^{iG \cdot r} U_G(r).
\]

Using this in Eq. (4), the interaction of the system with the random potential is given by

\[
H_{\text{dis.}} = \frac{e^2}{\epsilon} \sum_G \int r, r' U_G(r)e^{iG \cdot u(r)}
\]

\[
+ \frac{e^2}{\epsilon} \int r, r' U_0(r)\rho_0 (r),
\]

In Eq. (11), the interaction of \( \rho_0 \) with the random potential \( U_0 \) is called forward scattering, and the terms containing \( e^{iG \cdot u(r)} \) are called backward scattering. This nomenclature originates in the theory of electrons in 1D random potential.\(^{43,44}\) To calculate the disorder correlation functions, we consider the case of Gaussian distributed impurities where\(^{45,46}\)

\[
\rho_{\text{imp.}}(G + q)\rho_{\text{imp.}}(G' + q') = (2\pi)^3 D_\delta G \cdot G' \cdot \delta(q + q'),
\]

the parameter \( D \) measuring the disorder strength. Consequently, we find that for \( G \neq 0 \):

\[
U_G(r)U_{-G}(r') = D \int_{BZ} \frac{d^3q}{(2\pi)^3} e^{iG \cdot r} (G + q)^3 (G' + q)^3,
\]

Using the same arguments as before, we infer that the correlations of \( U_G(r) \) are short ranged, as in the case of neutral impurities, for all \( G \) except \( G = 0 \). This implies that the backward scattering terms induced by disorder are short-ranged and the treatment of these terms within the replica or the Martin-Siggia-Rose methods is identical to the case of neutral or screened impurities. However, the \( G = 0 \) component

\[
U_0(r) = \int_{BZ} \frac{d^3q}{(2\pi)^3} e^{iG \cdot r} \rho_{\text{imp.}}(q),
\]

manifests power law decay of the forward scattering correlations. This term however can be gauged out by the statistical tilt symmetry, and affects mainly the static properties of the periodic system. Typically, in periodic systems with both short range disorder and local elasticity, the contribution of the forward scattering disorder can be neglected and it is the backward scattering that induces collective pinning and Bragg glass features like a quasi order in the static correlation functions. Here, we have shown that even in the case of long range disorder, the backward scattering terms behave essentially like their short ranged (neutral impurities) counterparts. However, the effect of the forward scattering terms on the correlation functions has to be studied carefully. In the next section, we show that in the case of charged impurities in a charge density wave system, the forward scattering term strongly modifies the static correlation.

Finally, we remark that our decomposition of the elastic energy and the impurity potential is not exclusive to the Coulomb potential and is applicable to other long-range potentials. As a result, the conclusions of the present sections are expected to be valid for more general long range potentials.

### III. Charge Density Waves

In this section, we re-derive the elastic Hamiltonian for a \( d \) dimensional CDW with screened Coulomb interactions between the density fluctuations at zero temperature. We consider an incommensurate CDW, in which the electron density is modulated by a modulation vector \( Q \) incommensurate with the underlying crystal lattice. In this phase, the electron density has the following form:

\[
\rho(r) = \rho_0 + \frac{\rho_0}{Q^2} Q \cdot \nabla \phi(r) + \rho_1 \cos(Q \cdot r + \phi(r)),
\]

where \( \rho_0 \) is the average electronic density (see App. C for details). The second term in Eq. (12) is the long
wavelength density and corresponds to variations of the density over scales larger than $Q^{-1}$. The last oscillating term describes the sinusoidal deformation of the density at a scale of the order of $Q^{-1}$ induced by the formation of the CDW with amplitude $\rho_1$ and phase $\phi$.

In the absence of Coulomb interactions, the low energy properties of the CDW can be described by an effective Hamiltonian for phase fluctuations. For CDW aligned along the $x$ axis, i.e., $Q = Q\hat{x}$, this phase only Hamiltonian reads\cite{47,48,49,50}.

\[
H_0 = \frac{\hbar v_F n_e}{4\pi} \int d^3 r \left[ (\partial_x \phi)^2 + \frac{v_y^2}{v_z^2} (\partial_y \phi)^2 + \frac{v_y^2}{v_z^2} (\partial_z \phi)^2 \right],
\]

where $v_F$ is the Fermi velocity and $n_e$ is the number of chains per unit surface that crosses a plane orthogonal to $Q$. The velocity of the phason excitations parallel to $Q$ is $v_x = (m_e/m_x)^{1/2} v_F$ with $m^*$ the effective mass of the CDW and $m_e$ the mass of an electron. $v_y$ and $v_z$ denote the phason velocities in the transverse directions. A crucial observation is that a deformation of $\phi(r)$ along $Q$ produces an imbalance of the electronic charge density which then augments the electrostatic energy due to Coulomb repulsion between density fluctuations. We evaluate the contribution of Coulomb interactions screened beyond the characteristic length $\lambda$ which accounts for the presence of free carriers. This length diverges in the limit $T \to 0$.\cite{51,52} The electrostatic energy takes the form:

\[
H_C = \frac{e^2}{8\pi \epsilon} \int d^3 r d^3 r' e^{-|r-r'|/\lambda} \frac{\rho(r) \rho(r')}{|r-r'|},
\]

where we have assumed for simplicity an isotropic dielectric permittivity $\epsilon$ of the host medium.\cite{24,25,51,53,54} Due to the periodicity of the CDW system, we can use the decomposition of the Coulomb potential derived in Sec.\textbf{III} obtaining:

\[
H_C = \frac{e^2 \rho_0^2}{8\pi \epsilon Q^2} \int_{r,r'} \partial_x \phi(r) e^{-|r-r'|/\lambda} \frac{\rho(r) \rho(r')}{|r-r'|} + \frac{e^2 \rho_1^2}{2\epsilon} \int_{r,r'} \left[ V_{\perp Q}(r-r') e^{i(\phi(r)-\phi(r'))} + c.c. \right],
\]

In Eq.\textbf{ (18)}, we have neglected the contribution of the higher harmonics of the CDW. Note that the oscillating terms, as discussed in App.\textbf{B} only contribute to a renormalization of the coefficients in the short range elastic Hamiltonian\textbf{ (10)} and thus can be neglected. However, the contribution of the long-wavelength term has more dramatic effects and reads

\[
H_C = \frac{e^2 \rho_0^2}{2\pi Q^2} \int_{BZ} \frac{q_x^2}{\lambda^2 + q^2} |\phi(q)|^2,
\]

in the three dimensional case. It is interesting to note that Coulomb interactions generate a non local elasticity i.e., a $q$-dispersion in the elastic constant.

The total Hamiltonian in $d = 3$ now reads,

\[
H_{el.} = H_0 + H_C = \frac{1}{2} \int G^{-1}(q)|\phi(q)|^2,
\]

\[
G^{-1}(q) = \frac{n_e \hbar v_F}{2\pi} \left[ \frac{q_x^2}{q^2 + (\lambda - 2)^2 \xi^2} + q_y^2 + \frac{v_y^2}{v_z^2} q_z^2 + \frac{v_z^2}{v_x^2} q_x^2 \right]
\]

where the lengthscale $\xi$ is defined by:

\[
\xi^2 = \frac{n_e \hbar v_F}{2\pi e^2 \rho_0^2} Q^2 c.
\]

Depending on the ratio $\lambda/\xi$, two regimes of behavior can be identified: (i) Short ranged elasticity: when $\lambda/\xi \ll 1$ the Coulomb correction to the short range elasticity is small even in the limit $q \to 0$ and hence can be neglected.

(ii) Long range elasticity: for $\lambda/\xi \gg 1$, the Coulomb correction to the short-range elasticity cannot be neglected. This regime is relevant at low temperatures, when the number of free carriers available to screen the Coulomb interaction is suppressed by the CDW gap.\cite{21,52} Mean field calculations show that this regime is obtained for temperatures $T < 0.2 T_c$, where $T_c$ is the Peierls transition temperature.\cite{22} In the following, we focus on regime (ii), and accordingly, we take $\lambda^{-1} = 0$ in Eq.\textbf{ (20)}.

\section{IV. FORWARD SCATTERING}

As discussed in Sec.\textbf{ II} the case of the short ranged elasticity has been studied by various authors. For charged periodic systems with short range disorder and a nonlocal elasticity generated by Coulomb interactions, it is known that $d = 3$ becomes the upper critical dimension for disorder and the displacement correlations grow as $B(r) = \log log \Lambda r$.\cite{28,29} Here, we study the effect of the long range disorder on the static correlations of a charged periodic system. Since, the backward scattering terms generated by such a disorder are short ranged, they lead to the same physics as that of short ranged disorder with the corresponding nonlocal elasticity. These terms contribute a log log $r$ term to the displacement correlations. However, in this case a simple dimensional analysis shows that the forward scattering terms generate the leading contribution to the correlation functions. In the following, we calculate the contribution of the forward scattering disorder to the displacement correlation function in the $d = 3$ CDW.

\subsection{A. Displacement correlation functions}

The displacement correlation function is defined by:

\[
B(r) = \langle (\phi(r) - \phi(0))^2 \rangle = \frac{2}{L^d} \sum_q \langle \phi(q) \bar{\phi}(-q) \rangle [1 - \cos q \cdot r].
\]
The calculation of the correlation induced by the forward scattering disorder is analogous to the calculation of Larkin for the random force model. Assuming an infinite screening length \( \lambda \), the Hamiltonian reads:

\[
H = H_{el} + \frac{e^2}{4\pi \varepsilon} \int \frac{d^3q}{(2\pi)^3} \rho_{\text{imp}}(q) \rho(q') r_{\text{imp}}(r) \frac{\rho_{\text{imp}}(q) \rho(q')}{|r - r'|}.
\] (23)

Using Eq. (15) in Eq. (23), we obtain an expression of the form Eq. (12). Keeping only the forward scattering term we get:

\[
H = \int \frac{d^3q}{(2\pi)^3} \left[ \frac{G^{-1}(q)}{2} |\phi(q)|^2 + i \rho e^2 q_x \rho_{\text{imp}}(q) \phi(q) \right].
\] (24)

Shifting the field \( \phi \)

\[
\phi(q) = \tilde{\phi}(q) + \frac{e^2}{\varepsilon} q_x G(q) \rho_{\text{imp}}(q),
\] (25)

brings the Hamiltonian back to the form of Eq. (20). The average over disorder now yields:

\[
\langle (\phi(q) \phi(-q)) \rangle = \langle \tilde{\phi}(q) \tilde{\phi}(-q) \rangle + \frac{e^2}{\varepsilon} DQ^2 G(q)^2 \rho_{\text{imp}}(q) \rho_{\text{imp}}(-q)
\]

\[
= \kappa \left[ T \left( G(q) + \frac{e^2}{\varepsilon} q_x^2 \right) \right],
\] (26)

where \( \langle \ldots \rangle \) and \( \overrightarrow{\ldots} \) denote thermal average and disorder average respectively. Eq. (26) shows that even in the presence of Coulombian disorder, the statistical tilt symmetry is preserved. This implies that in the presence of backward scattering disorder, the forward scattering term can be gauged out by \( Q \), and the contribution of the forward scattering disorder \( B^{FS} \) is simply added to the one obtained from the backward scattering disorder, \( B^{BS} \). We conclude

\[
B^{FS}(r) = 2D \left( \frac{e^2}{\varepsilon} \frac{\rho_0}{Q^2} \int \frac{d^3q}{(2\pi)^3} \frac{q_x^2 G(q)^2}{q^4} [1 - \cos(q \cdot r)] \right). (27)
\]

We want to evaluate this integral for the case of \( v_y = v_z = v_\perp \). In the following we will use \( q_x^2 = q_y^2 + q_z^2 \). To obtain the asymptotic behavior of \( B(r) \) for \( r \rightarrow \infty \) we need to consider the \( q \rightarrow 0 \) limit of the integrand. The form of \( G(q) \) suggests a scaling \( q_x \sim q_\perp^4 \), which then allows us to consider the integral:

\[
F(r) = \int \frac{d^3q}{(2\pi)^3} \frac{q_x^2}{q^2 + (q_\perp^2)^2} (1 - \cos(q \cdot r)) (28)
\]

where \( \xi' = \xi_{\perp}/v_x, r_\perp^2 = y^2 + z^2 \) and \( \Lambda \) is a momentum cut-off. A study of the limits of this function for \( r_\perp \rightarrow \infty \) and \( |x| \rightarrow \infty \) shows that its asymptotic behavior is well described by:

\[
F(r) \sim \frac{v_x}{16\pi v_{\perp} \xi} \ln \left( \frac{r_\perp^2 + 4(v_{\perp}|x|\xi/v_x)}{\Lambda^2} \right). (29)
\]

Therefore, we have for \( r \rightarrow \infty \):

\[
B^{FS}(r) = \kappa \ln \left( \frac{r_\perp^2 + 4(v_{\perp}|x|\xi/v_x)}{\Lambda^2} \right). (30)
\]

where:

\[
\kappa = \frac{DQ^2 v_x}{16\pi \xi \rho_0 v_{\perp}}.
\] (31)

The full asymptotic correlation function is given by the sum of the forward scattering contribution, Eq. (30), and the backward scattering contribution given in Eq. (51) of Ref. 29 for the case of a short-range disorder and non-local elasticity:

\[
B^{BS}(r) = \log(\log(\max(\Lambda|x|, (\Lambda r_{\perp})^2))). (32)
\]

Obviously, the contribution of the backward scattering terms is subdominant and can be neglected.

\section{Analogy with smectics-A}

We note that the result Eq. (29) can be obtained in the entirely different context of liquid crystals. If we consider a smectic-A liquid crystal, its elastic free energy reads:

\[
\mathcal{F}_{\text{el}} = \int d^3r \left[ \frac{1}{2} B(\partial_{\perp} u)^2 + \frac{1}{2} k_{11}(\Delta_{\perp} u)^2 \right],
\] (33)

where \( u \) represents the displacement of the smectic layers, \( B \) is the compressibility, and \( k_{11} \) measures the bending energy of the smectic layers. If we now assume a random compression force given by:

\[
\mathcal{F}_{\text{dis}} = \int d^3r \eta(r) \partial_{\perp} u(r),
\]

\[
\eta(r) \eta(r') = D \delta(r - r'),
\] (34)

\[
\eta(r) \eta(r') = D \delta(r - r'),
\] (35)

a straightforward calculation shows that the displacement correlation function \( (u(r) - u(0))^2 \) is given by Eq. (29). Smectics-A with disorder have been considered previously in Ref. 51 albeit with a different type of disorder coupling to \( \nabla_{\perp} u \). This yields a displacement correlation function superficially similar to \( F(r) \) with \( q_{\perp}^2 \) replacing \( q_x^2 \) in the numerator. The random compression force, which is not natural in the smectic-A context, is thus easily realized with charge density wave systems.
V. EXPERIMENTAL IMPLICATIONS

In the preceding sections, we have shown that the forward scattering terms generated by charged impurities lead to smectic-like order in a charge density wave material. A frequently used technique to characterize positional correlations in CDW systems is x-ray diffraction. In the present section, we provide a calculation of the x-ray intensity resulting from such a smectic-like order, and we provide a quantitative estimate of the exponent $\kappa$.

A. x-ray intensity

The intensity of the x-ray spectrum is given by

$$I(q) = \frac{1}{L^3} \sum_{i,j} e^{-i(q(R_i-R_j))} \left( f_i f_j e^{-i(q(u_i-u_j))} \right) .$$

(36)

where $u_i$ is the atom displacement from the equilibrium position $R_i$, $f_i$ represents the total amplitude scattered by the atom at the position $i$ and depends exclusively on the atom type. We consider the simple case of a disordered crystal, made of one kind of atoms, characterized by the atom type. We consider the simple case of a disordered crystal, made of one kind of atoms, characterized by the atom type. We consider the simple case of a disordered crystal, made of one kind of atoms, characterized by the atom type.

By expanding Eq. (36) for low $q(u_i-u_j)$, one finds an expression of the x-ray satellite intensity comprising a part $I_d$, which is symmetric under inversion around the Bragg vector $K$ and a part $I_a$, which is antisymmetric under the same transformation. The symmetric part is given by the following correlation function:

$$I_d(q) = \frac{f^2}{2} \int d^3 r e^{-i q \cdot r} \langle u(r/2) u(-r/2) \rangle .$$

(38)

and the antisymmetric part by:

$$\frac{I_a(q)}{a \cdot (b \times c)} = 2 q \Delta f \text{Im} \int d^3 r e^{-i q \cdot r} \langle \rho_{\text{imp}}(r/2) u(-r/2) \rangle ,$$

(39)

where: $\delta q = (q-K) \sim Q$, and $a \cdot (b \times c)$ is the volume of the unit cell of the crystal. After some manipulations, Eq. (68) can be rewritten as:

$$I_d(K + Q + k) = u_0^2 f^2 K^2 \int d^3 r e^{-i k \cdot r} C_d(r) ,$$

(40)

$$I_a(K + Q + k) = -J K u_0 \Delta f \sqrt{NB} \int d^3 r e^{-i k \cdot r} C_a(r) ,$$

(41)

where $N$ is the number of impurities in the unit cell, and:

$$C_d(r) = \left\langle e^{i(\phi(r/2) - \phi(-r/2))} \right\rangle ,$$

(42)

$$C_a(r) = \chi(r) C_d(r) ,$$

(43)

where $\chi(r)$ is defined by Eq. (33) of Ref. 29. It is easy to show, using this definition and the statistical tilt symmetry that $\chi(r)$ is independent of the forward scattering disorder. To Eq. (41), one sees that the term $C_d^{B,S}$ gives only a logarithmic correction to $C_d$. As a result, the symmetric structure factor $C_d^{B,S}$ is the backward scattering contribution which has been obtained in Ref. 29 and $C_d^{F,S}$ is the forward scattering contribution, given by:

$$C_d^{F,S}(r) = \left( \frac{\Lambda^{-2}_{1}}{r_{\perp}^2 + 4(v_{\perp}^2|x|/v_x)} \right)^{\kappa} ,$$

(44)

where we have used Eq. (69), assuming a Gaussian disorder. Using Eq. (32), one sees that the term $C_d^{B,S}$ gives only a logarithmic correction to $C_d$. As a result, the symmetric structure factor $I_d$ is dominated by the contribution of the forward scattering disorder. To obtain the structure factor, we Fourier transform Eq. (44) to obtain

$$I_d(q) = \int d^3 r e^{i q \cdot r} \left( \frac{\Lambda^{-2}_{1}}{r_{\perp}^2} \right)^{2\kappa} \frac{r_{\perp}^2 v_x}{2\xi v_{\perp}}$$

$$\times \int_0^{+\infty} du \frac{\cos(q v_{\perp}^2 u v_x)}{(1 + u)^\gamma} ,$$

(45)

Using the following relation,

$$\int_0^{+\infty} du \frac{\cos(q \lambda u)}{u} = \frac{\lambda^{-1}}{\Gamma(\gamma)} \int_0^{+\infty} dv e^{-v\lambda} v^{\gamma-1}$$

(46)

we finally obtain

$$I_d(q) = \pi(q x_s^{2})^{-\kappa-2} \frac{\Lambda^{-2}_{1}}{2^{2(\kappa-1)} \Gamma(\kappa)}$$

$$\times \int_0^{+\infty} dw \frac{u_{\perp}^{1-\kappa}}{w^{2} + 1} e^{-w v_{\perp} x_s^2 \xi/\nu_s^2} ,$$

(47)

so that $I_d(q) \sim (|q_x|)^{\kappa-2}$ for $q_x^2 (\xi v_{\perp} / x_s) \ll |q_x|$ and $I_d(q) \sim (|q_x|)^{2(\kappa-2)}$ otherwise. The intensity $I_d(q = 0)$ is divergent for $\kappa \leq 2$ but is finite for $\kappa > 2$, i.e. for strong disorder. Next, we turn to the evaluation of $I_a$. From Ref. 29 we know that $\chi(r) \sim 1/x$ when $x_s \gg v_x$ and $\chi(r) \sim 1/r_{\perp}^2$ when $x_s \ll v_x$. This implies that $I_a$ is subdominant in comparison with $I_d$. In particular, $I_a(q) \sim (|q_x|)^{\kappa-1}$ for $q_x^2 (\xi v_{\perp} / x_s) \ll |q_x|$ and $I_a(q) \sim (|q_x|)^{2(\kappa-1)}$ for $q_x^2 (\xi v_{\perp} / x_s) \gg |q_x|$.
We note that these intensities are remarkably similar to those of a disorder-free smectic-A liquid crystal at positive temperature. In fact, the expression of the exponent $\kappa$ Eq. (31) is analogous to the expression (5.3.12) in Ref. 58, with the disorder strength $D$ playing the role of the temperature $k_B T$ in the smectic-A liquid crystal.

![Image of the x-ray intensity in a CDW with Coulombic disorder and charged impurities for $K, Q$ parallel to the chain direction.](image)

**FIG. 1:** A sketch of the x-ray intensity in a CDW with Coulombic disorder and charged impurities for $K, Q$ parallel to the chain direction. We have taken $\kappa = 0.5$ in the expressions of $I_a$ and $I_d$.

**B. Estimate of the exponent $\kappa$**

Let us turn to an estimate of the exponent $\kappa$ appearing in the intensities to determine whether such smectic-like intensities are indeed observable in experiments. To do this, we first need to determine whether Coulomb interactions are unscreened by comparing the screening length with $\xi$ given by Eq. (24). This question is relevant only to a material with a full gap, in which free uncondensed electrons cannot screen charged impurities. A good candidate is the blue bronze material $K_{0.3}MoO_3$ which has a full gap, and is well characterized experimentally. We now evaluate the quantity $\xi$ for this material. Using the parameters of Ref. 67:

$$n_e = 10^{20} \text{chains/m}^2, \quad v_F = 1.3 \times 10^5 \text{m/s}, \quad \rho_0 = 3 \times 10^{27} \text{e}^-/\text{m}^3, \quad Q = 6 \times 10^9 \text{m}^{-1}, \quad (47)$$

and a relative permittivity of $\varepsilon_{K_{0.3}MoO_3} = 1$, so that $\kappa$ in Eq. (24) is equal to the permittivity of the vacuum, we obtain: $\xi \simeq 5 \text{Å}$. Therefore, the screening length can be large compared to $\xi$ at low temperature, and we expect that Coulombian effects will play an important role in this material. We can use this value of $\xi$ to evaluate the exponent $\kappa$ in Eq. (31). For the doped material $K_{0.3}Mo_{1-x}V_xO_3$, we find that the disorder strength can be expressed as a function of the doping and obtain:

$$D = x(1-x) \frac{\#(\text{Mo atoms/unit cell})}{a \cdot (b \times c)}.$$  

(51)

This formula is derived in App. E. For the crystal parameters, $a = 18.25 \text{Å}, b = 7.56 \text{Å}, c = 9.86 \text{Å}, \beta = 117.53^\circ$, with 20 Molybdenum atoms per unit cell, and a doping $x = 3\%$, the disorder strength $D = 4.8 \times 10^{26} \text{m}^{-3}$. Moreover, using the experimental bounds of the velocities: $3.6 \times 10^2 \text{m/s} < v_\perp < 1.6 \times 10^3 \text{m/s}$ and $v_\parallel = 3.7 \times 10^3 \text{m/s}$, we find that $\kappa$ is in the range $[0.16 - 0.8]$. Therefore, the smectic-like order should be observable in x-ray diffraction measurements on this material.

**VI. CONCLUSION**

In this paper, we have introduced a decomposition of the disorder induced by charged impurities in terms of the reciprocal lattice vectors of a periodic charged elastic system. Using this decomposition, we have shown that only the long wavelength (forward scattering) component of the disorder was long-range correlated. Components with wavevectors commensurate with the reciprocal lattice of the periodic elastic system remain short ranged. The latter can thus be treated with the standard techniques developed for impurities producing short range forces. We find that only the forward scattering is affected by the long-range character of the forces created by charged impurities. Due to the statistical tilt symmetry, this implies that only the statics of the periodic elastic system is modified by Coulombian disorder. This has allowed us to obtain a full picture of the statics of charge density wave systems in $d = 3$ in the presence of charged and neutral impurities. The results are summarized in Table I. A remarkable result is that in the case of charged impurities in a system with unscreened Coulomb interactions, the x-ray intensity turns out to be identical to that produced by thermal fluctuations in a smectic-A liquid crystal, with the disorder strength playing the role of an effective temperature. This behavior of the scattering intensity should be observable in the blue bronze material $K_{0.3}MoO_3$ doped with charged impurities such as Vanadium.
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APPENDIX A: DECOMPOSITION OF THE COULOMB POTENTIAL

In this appendix, we give more details on the decomposition of the Coulomb potential using infinitely differentiable functions of compact support. First, let us discuss a simple decomposition in 1D. We consider the function $F_\epsilon(x)$ such that:

\begin{align}
F_\epsilon(x) &= F_\epsilon(-x) \\
F_\epsilon(x) &= 1, \quad 0 \leq x \geq 1 \\
F_\epsilon(x) &= \frac{1}{2} \left\{ 1 - \tanh \left[ \frac{2(1-x)}{(x-1)^2 + \epsilon^2} \right] \right\}, \quad |x-1| < \epsilon \\
F_\epsilon(x) &= 0, \quad x > 1 + \epsilon
\end{align}

(A1)

It is easy to check that $F_\epsilon$ is continuous, infinitely differentiable, and that:

\begin{equation}
\sum_{n=-\infty}^{\infty} F_\epsilon(x - 2n) = 1.
\end{equation}

(A2)

Applying this formula to a one dimensional reciprocal space, we obtain:

\begin{equation}
\sum_{n_x=-\infty}^{\infty} F_\epsilon \left( \frac{a}{\pi} \left( q - \frac{2\pi n_x}{a} \right) \right) = 1,
\end{equation}

(A3)

i.e. we have constructed explicitly a partition of the unity. The generalization to a cubic lattice in a three dimensional space is obvious:

\begin{equation}
\sum_{n_x,n_y,n_z=-\infty}^{\infty} F_\epsilon \left( \frac{a}{\pi} \left( q_x - \frac{2\pi n_x}{a} \right) \right) F_\epsilon \left( \frac{a}{\pi} \left( q_y - \frac{2\pi n_y}{a} \right) \right) \times F_\epsilon \left( \frac{a}{\pi} \left( q_z - \frac{2\pi n_z}{a} \right) \right) = \sum_G F^{3D}_\epsilon(q-G) = 1.
\end{equation}

(A4)

The function $F^{3D}_\epsilon$ has a compact support, and vanishes rapidly outside the Brillouin zone.

APPENDIX B: CONTRIBUTION OF THE OSCILLATING COMPONENTS OF THE DENSITY TO THE ELASTIC HAMILTONIAN IN THE PRESENCE OF COULOMB INTERACTION

In this appendix, we calculate the contribution of the oscillating terms to the Hamiltonian of the charge density wave with unscreened Coulomb interactions, Eq. (17), and show that they only induce corrections to the short range elasticity. Inserting the expression of the density, Eq. (15), in Eq. (7), the contribution of the oscillating component of wavevector $Q$ is given by,

\begin{equation}
H^\text{osc.}_C = \frac{\epsilon^2}{2e} \sum_{n,n'} \int dx dx' V_Q(x-x',n-n') \times \cos [\phi(x,n) - \phi(x',n')],
\end{equation}

(B1)

where we have reestablished the discrete character of the transverse dimension $y$. Both the intrachain $(n = n')$ and the interchain contributions $(n \neq n')$ are short ranged. Let us first consider the case of $n \neq n'$. We have to compute integrals:

\begin{equation}
V_Q(x,n) = \int \frac{d^d q}{(2\pi)^d} F^{3D}_\epsilon(q) \frac{e^{i(q_x x + q_z z - n_z)}}{(Q + q_z)^2 + q^2_\perp},
\end{equation}

(B2)
To evaluate the above integral in closed form, we need to make some approximations. Since \( F_r \) vanishes for \( Q + q_x = 0 \), we can neglect \( q_x \) compared to \( Q \) in this integral. Then, we can extend the integration over the whole reciprocal space without encountering any singularity. The \( q_x \) integration produces a \( \delta(x) \) function, and the \( q_y \) integration gives an exponential in 2D and a modified Bessel function \( K_0 \) in 3D. In the two-dimensional case, we find that:

\[
V_Q(x, n) = \frac{\exp(-Q\ell_n)}{2Q} \delta(x), \quad (B3)
\]

and in the three-dimensional case:

\[
V_Q(x, n) = \frac{K_0(Q\sqrt{(n_0\ell_y)^2 + (n_z\ell_z)^2})}{2\pi} \delta(x). \quad (B4)
\]

where, \( \ell_y \) and \( \ell_z \) are interchain spacings. Due to the exponential decay of the interchain interaction with the distance, it is justified to neglect interchain interactions beyond nearest neighbors. The logarithmic divergence in Eq. \( (B4) \) for \( n = 0 \) is an artefact of the approximation we make when we integrate over the entire reciprocal space instead of the first Brillouin zone. A more refined estimate yields a finite, short ranged intrachain contribution.

The short range contribution in the electrostatic energy thus reads:

\[
H_{\text{osc}}^{\text{sec.}} = \sum_{a=y,z} J_a \int dx \sum_n \cos[\phi(x, n + e_a) - \phi(x, n)] + \sum_n \int V_Q(x - x', 0) dx dx' \cos[\phi(x, n) - \phi(x, n')], \quad (B5)
\]

where:

\[
J_a = \frac{e^2\rho_a^2}{2\pi e} K_0(Q\ell_a). \quad (B6)
\]

In Eq. \((B5)\), we expand \( \cos(\phi(x, n) - \phi(x', n)) \sim 1 - (x - x')^2 / 2(\partial_x \phi(x, n))^2 \) to show that the backscattering term reduces to short ranged elastic forces. Making \( \phi(x, n) = \phi(x, n) + n_y\pi + n_z\pi \), we can make the sign in front of \( J_a \) negative, and obtain the ground state for \( \phi = 0 \). In its ground state, the CDW is out of phase on two nearest neighbor chains. This ground state is represented on Fig. 3. The excitations above this ground state are described by the Lagrangian \((D3)\) derived in App. D.

**APPENDIX C: ZERO TEMPERATURE LIMIT OF THE CHARGE DENSITY IN A CDW**

In the present appendix, we discuss the zero temperature limit of the expression of the charge density in a CDW. Let us consider the form of the charge density in the presence of a non-uniform \( \phi \). It is given\(^{25, 47, 70, 71}\) by the expression:

\[
\rho(r) = \rho_0 + \frac{\rho_0 \bar{\rho}_e}{Q} \partial_x \phi(r) + \rho_1 |\psi| \cos(Qx + \phi(r)), \quad (C1)
\]

where \( \rho_0 \) is the average electron density, \( \rho_1 \) is the condensate amplitude at \( T = 0 \), \( |\psi| \) takes into account the reduction of CDW order by thermal fluctuations (\( |\psi| = 1 \) at \( T = 0 \)), the factor \( \bar{\rho}_e \) takes into account the presence of non-condensed electrons\(^{21}\) at finite temperature (at \( T = 0 \), \( \bar{\rho}_e = 1 \) and \( Q = 2k_F \)). Using the relation \( k_F = \frac{\pi}{2} \rho_0 \), valid in a one-dimensional system, one can see that this relation simplifies (at \( T = 0 \)) to:

\[
\delta \rho = \frac{\partial_x \phi}{\pi}. \quad (C2)
\]

In the paper, we consider temperatures very low compared to the Peierls transition temperature, and we take \(|\psi| = 1\), \( \bar{\rho}_e = 1 \). This yields Eq. \((14)\).

**APPENDIX D: DERIVATION OF THE HAMILTONIAN OF A THREE-DIMENSIONAL CHARGE DENSITY WAVE**

In this appendix, we provide a derivation of the Hamiltonian of a three dimensional CDW starting from the original Fukuyama one dimensional description. The Lagrangian of a CDW in a single chain is given by\(^{47}\)

\[
\mathcal{L} = \frac{\hbar v_F}{4\pi} \int dx \left[ \left( \frac{\partial_t \phi}{v^2} - (\partial_x \phi)^2 \right) \right] (D1)
\]

Obviously, this Lagrangian describes phason waves propagating with the velocity \( v_x \). \( v_F \) is the Fermi velocity of the electrons forming the CDW and \( \phi \) is the phase of the CDW. We define an effective mass \( m^* \) by:

\[
\frac{v^2_F}{v^2} = \frac{m^*}{m_e}, \quad (D2)
\]

where \( m_e \) is the electron mass.

In a three-dimensional CDW with screened Coulomb interactions, the chains are coupled by a backscattering interaction. The resulting Lagrangian reads:

\[
\mathcal{L} = \frac{\hbar v_F}{4\pi} \sum_n \int dx \left[ \left( \frac{\partial_t \phi}{v^2} - (\partial_x \phi)^2 \right) \right] (D3)
\]

\[
\frac{1}{2} \sum_{n, n'} J(n, n') \int dx \cos[\phi(x, n) - \phi(x, n')],
\]

where \( J(n, n') \) is short-ranged and is given by Eq. \((B6)\). Expanding the cosines,

\[
\cos[\phi(x, n) - \phi(x, n + e_y)] = 1 - \frac{(\phi(x, n) - \phi(x, n + e_y))^2}{2} + o(\nabla_y^2)
\]

\[
\cos[\phi(x, n) - \phi(x, n + e_y)] = 1 - \frac{\ell_y^2}{2} (\partial_y \phi)^2 + o(\partial_y \phi)^2, \quad (D4)
\]

and defining

\[
\frac{\hbar v_F}{4\pi} \frac{v^2_{y,z}}{v^2} = J_{y,z}, \quad (D5)
\]
the Lagrangian in Eq. (D3) can be rewritten as

\[
\mathcal{L} = \sum_{n} \frac{h v_F}{4 \pi} \int dx \left[ \frac{(\partial_x \phi)^2}{v_\phi^2} - (\partial_x \phi)^2 \right] - \frac{v_y^2}{v_\phi^2} (\partial_y \phi)^2 - \frac{v_z^2}{v_\phi^2} (\partial_z \phi)^2
\]

(D6)

The sum over lattice sites in the transverse direction can be replaced by an integral, by writing:

\[
\mathcal{L} = \frac{h v_F}{4 \pi l_\perp} \int d^2r \left[ \frac{(\partial_x \phi)^2}{v_\phi^2} - (\partial_x \phi)^2 \right] - \frac{v_y^2}{v_\phi^2} (\partial_y \phi)^2 - \frac{v_z^2}{v_\phi^2} (\partial_z \phi)^2
\]

(D7)

The phason dispersion is now \( \omega(q)^2 = v_x^2 k_x^2 + v_y^2 k_y^2 + v_z^2 k_z^2 \). The momentum conjugate to \( \phi \) is obtained by the usual relation:

\[
\Pi = \frac{\delta \mathcal{L}}{\delta (\partial_x \phi)} = \frac{h v_F}{2 \pi v_\phi^2} \partial_t \phi,
\]

(D8)

yielding the Hamiltonian:

\[
H = \frac{h v_F}{4 \pi l_\perp} \int d^3r \left[ \frac{4 \pi^2 v_\phi^2 (l_\perp l_z)^2}{h^2} \Pi^2 + (\partial_x \phi)^2 \right] - \frac{v_y^2}{v_\phi^2} (\partial_y \phi)^2 + \frac{v_z^2}{v_\phi^2} (\partial_z \phi)^2.
\]

(D9)

From the Hamiltonian Eq. (D9), it is straightforward to obtain the Debye-Waller factor associated with the zero point fluctuations of the phase \( \phi \). In the isotropic case, \( v_y = v_z = v_\phi \), one finds that \( \langle \cos \phi(x) \rangle \tau = -\propto v \sim e^{-C(m_\phi/m^*)^2} \), where \( C \sim \pi/4 \) is a dimensionless constant of order 1. Due to the smallness of the ratio \( m_\phi/m^* \sim 10^{-2} \), the zero point motion can be neglected, and the kinetic term \( \Pi^2 \) in Eq. (D9) can be dropped. This leads to the Hamiltonian (16).

APPENDIX E: ESTIMATION OF THE DISORDER STRENGTH

Here, we give an estimation of the disorder strength \( D \) in doped KMo\(_{1-x}\)V\(_x\)O\(_3\). We assume a binomial distribution of Vanadium impurities on the Molybdenum sites. The Vanadium impurities carry an extra electron compared to the Molybdenum ions. The resulting charge density fluctuation reads:

\[
\delta \rho(r) = \sum_{i,\alpha} (x - \sigma_{i,\alpha}) \delta(r - R_{i,\alpha}),
\]

(E1)

where \( i \) is the index of the cell and \( \alpha \) is the index of the Molybdenum site in a given cell. \( \sigma_{i,\alpha} = 0 \) if the site is occupied by a Molybdenum ion, and \( \sigma_{i,\alpha} = 1 \) if it is occupied by a Vanadium impurity. By construction, the expectation value of \( \delta \rho(r) \) is zero. We estimate the second moment of \( \delta \rho(r) \) as:

\[
\langle \delta \rho(r) \delta \rho(r') \rangle = \sum_{i,j,\alpha,\beta} \delta(r - R_{i,\alpha}) \delta(r' - R_{j,\beta})
\]

(E2)

\[
= x(1-x) \sum_{i,\alpha} \delta(r - R_{i,\alpha}) \delta(r' - r),
\]

where we have used the property that \( \delta(r - R_{i,\alpha}) \delta(r' - R_{j,\beta}) = \delta_{i,j} \delta_{\alpha,\beta} \delta(x - \sigma_{j,\beta})^2 \). The expectation value of \( \sum_{i,\alpha} \delta(r - R_{i,\alpha}) \) is simply the number of Molybdenum ions per unit volume, leading to the formula Eq. (61).

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The exponent η is universal and the best prediction is given by FRG calculation η ≈ 1.1. However also in the case where the elastic object has many components (i.e. for the vortex lattice) the possible variation of this exponent is still less than a percent. [S. Bogner, T. Emig and T. Nattermann Phys. Rev. B 63 174501 (2001)].

by slowly varying, we mean that the Fourier components of u are different from zero only for wavevectors much smaller than |Gmin|. A more straightforward approach would be to keep the hard cutoff at the edge of the Brillouin zone. Then, the function V_G would decay as 1/r^2 with an oscillating prefactor. The same oscillation would be also obtained for a hard cutoff.