X-ray diffraction of a disordered charge density wave

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We study the X-ray diffraction spectrum produced by a collectively pinned charge density wave (CDW), for which one can expect a Bragg glass phase. The spectrum consists of two asymmetric divergent peaks. We compute the shape of the peaks, and discuss the experimental consequences.

The statics and dynamics of disordered elastic objects govern the physics of a wide range of systems, either periodic, such as vortex flux lines [1] and charge density waves (CDW) [2], or involving propagating interfaces, such as domain walls in magnetic [3] or ferroelectric [4] systems, contact lines of liquid menisci on rough substrates [5] and propagation of cracks in solids [6]. It was recently shown that periodic systems have unique properties, quite different from the ones of the interfaces. If topological defects (i.e. dislocations etc.) in the crystal are excluded, displacements grow only logarithmically [7, 8, 9], instead of the power-law growth as for interfaces. The positional order is only algebraically destroyed [9, 10] leading to divergent CDW peaks for which one expects to be in the Bragg glass regime. We show that the diffraction spectrum consists in two asymmetric peaks. In contrast to previous assumptions [10], we show that the asymmetry is present also in the weak pinning limit. The peaks are power-law divergent, with an anisotropy in shape. This form is consistent with the Bragg glass behavior [10]. The asymmetry is a subdominant power-law too, with an exponent that we determine. We also briefly discuss the role of unscreened Coulomb interaction for the CDW on the diffraction spectrum.

The general expression [12] for the total diffraction intensity in a crystal is given by

\[ \mathcal{I}(q) = \frac{1}{V} \sum_{i,j} e^{-i q (R_i - R_j)} \left( f_i f_j e^{-i q (u_i - u_j)} \right), \]  

where \( u_i \) is the atom displacement from the equilibrium position \( R_j = ja \), with \( a \) indicating the lattice constant, \( f_i \) the atomic scattering factor and \( \langle \cdots \rangle \) denotes the double average over the disorder and over the thermal fluctuations. As an example let us first consider the case of fixed atoms \( u_i = 0 \). We obtain:

\[ \mathcal{I}(q) = f^2 \sum_K \delta(q - K) + \Delta f^2 N_I, \]  

where \( \Delta f = f_I - f \) is the difference between the impurity and the host atom scattering factors, \( N_I = n_I (1 - n_I) \), with \( n_I \) is the impurity concentration and \( f_I \) is the average scattering factor. The usual Bragg peaks, in correspondence to the reciprocal lattice vectors \( K \), arise from the first term in (2), the second term is responsible for a background intensity, called Laue scattering, due to the disorder.

In a second stage we take into account displacements of the atoms related to the presence of a CDW. To this purpose, we consider an electron density characterized by a sinusoidal modulation:

\[ \rho(x) = \rho_0 \cos(Qx + \phi(x)). \]
\( \phi \) is the phase of the charge density wave and \( Q = 2k_F \), where \( k_F \) is the Fermi wave vector. The associated Hamiltonian writes:

\[
H = \int d^d x \frac{c}{2} (\nabla \phi(x))^2 + V_0 \int d^d x \Sigma(x) \rho(x),
\]

where \( d \) is the dimension of the space. The first term in the Hamiltonian \( \Sigma \) represents the elasticity. The elasticity in fact anisotropic \( 2\) along the \( Q \)-direction:

\[
H_{\text{el}} = \int dx d^{d-1} y \left( \frac{c_1}{2} (\partial_2 \phi)^2 + \frac{c_2}{2} (\partial_y \phi)^2 \right),
\]

where \( x \parallel Q \), and \( c_1 \gg c_2 \). The compression along \( x \) corresponds to an increase of electric charge density and thus pays the price of Coulomb repulsion, while distortions along the remaining \( d-1 \) directions are much easier. We are led back to \( 4 \) by redefining the spatial variables \( x' = x/\sqrt{c_1} \) and \( y' = y/\sqrt{c_2} \), with \( c = (c_1 d^{-1})^2 \). The main effect in the diffraction spectrum is thus to make the shape of the peaks anisotropic, but this will not change the overall divergence. The local, but anisotropic, elasticity \( 5 \) is valid beyond the distance at which the Coulomb interaction between various parts of the CDW is screened. If this length is very large, or if one want to examine short range regime one should keep the \( q \)-dependence in the elastic constants. This leads to a more complicated behavior that we will only briefly discuss here and will be examined in details elsewhere \( 21 \). The second term in the Hamiltonian \( 4 \) reflects the effect of the disorder on the electron density. The Gaussian random function \( \Sigma(x) \) describes the impurity distribution and is characterized by the correlator \( \Sigma(x)\Sigma(y) = N_I \delta(x-y) \), \( V_0 \) is a positive constant which measures the impurity potential and finally the sign \( +(-) \) is related to the repulsive (attractive) interaction between the electrons and the local impurity. In the following, we restrict our analysis to the repulsive case, \( \rho_0 \) is absorbed in \( V_0 \) and we define the disorder strength \( D = V_0^2 N_I \).

A density modulation is accompanied by a lattice distortion \( u \) given at low temperature by

\[
u(x) = \frac{u_0}{Q} \cos(Q x + \phi(x)).
\]

We are interested in the behavior of the scattering intensity \( I(q) \) near a Bragg peak \( (q \sim K) \). Since \( |\delta q| = |q-K| \ll K \), we can take the continuum limit \( i \to x \) and we obtain from \( 10 \):

\[
I(q) = \int \left\langle J_T \int d^3 r e^{-iq \cdot r} - \frac{D}{T} \sum_{a,b} \cos(\phi_a(r) - \phi_b(r)) \right\rangle,
\]

where \( D = T \int d^3 r e^{-iq \cdot r} + \int d^3 r \Sigma(r) \) and \( f_T = T + \Delta f_d/2 \Sigma(r) \). In \( 7 \) we have applied the standard decomposition in center of mass \( R \) and relative \( r \) coordinates \( (x = R + \frac{r}{2} \) and \( y = R - \frac{r}{2} \)). The integration over \( R \) has already been performed because \( u \) vary slowly at the scale of the lattice spacing. Assuming that in the elastic approximation displacements remain small \( (u_i \ll R_i) \), one can expand \( 4 \) as powers of \( Ku_0 \). Developing up to the second order we get \( 17 \):

\[
I(q) = I_d + I_a + I_{\text{tripl}},
\]

\[
I_d = T^2 q^2 \int \langle u^2 \rangle u(-\frac{q}{2}),
\]

\[
I_a = -iq \Delta f_d/2 T \int \langle \Sigma(-\frac{q}{2}) u(-\frac{q}{2}) - \Sigma(\frac{q}{2}) u(-\frac{q}{2}) \rangle,
\]

\[
I_{\text{tripl}} = -iq \Delta f^2/2 T \int \langle \Sigma(-\frac{q}{2}) \Sigma(\frac{q}{2}) (u(\frac{q}{2}) - u(-\frac{q}{2})) \rangle.
\]

While the contribution \( I_d \) represents the intensity due to the atomic displacements alone, the contributions \( I_a \) and \( I_{\text{tripl}} \) are generated by the coupling between the disorder and the displacement. The presence of a CDW is signaled by the formation, around each Bragg peak, of two satellites at reciprocal vectors \( K \pm Q \). In absence of disorder \( (D = 0) \) the displacement term has the form \( I_d = f^2 q^2 u_0^2 \sum K \delta(q + K \pm Q) \) and the other terms are vanishing: in this case the two satellites have the same intensity and the broadening is absent. To interpret the experimental findings \( 15 \), in particular to explain the measured strong asymmetry \( 25 \) between the peaks at \( K + Q \) and at \( K - Q \), we need to account for the effect of impurities.

In the literature the term \( I_a \) was evaluated by means of models \( 16, 17, 18 \) which describe the pinning by imposing a constant value \( \phi_0 \) to the phase in \( 4 \) in proximity of each impurity, and \( I_{\text{tripl}} \) was conjectured to be negligible \( 17, 22 \). In that approach, the observed satellite asymmetry is seen as a clear sign of the strong disorder; in fact, \( \phi_0 \) is not constant and, for sufficiently large domains, one should have \( 16 \) \( I_a \propto \cos(\phi_0) \sim 0 \). To go beyond this phenomenological approach and also deal with the weak disorder limit, in which one expects the Bragg glass, we use a Gaussian variational approach \( 14, 22 \). We first perform the average over the disorder using the standard replica techniques. The replicated Hamiltonian corresponding to \( 4 \)

\[
H_{\text{eff}} = \int d^4 r \sum_a \frac{c}{2} (\nabla \phi_a)^2 - \frac{D}{T} \sum_{a,b} \cos(\phi_a(r) - \phi_b(r)),
\]

where \( T \) is the temperature and the sum over the \( n \) replica has to be considered in the limit \( n \to 0 \). We stress that, moving from the Hamiltonian \( 4 \) to its replicated version we also need to change the correlation functions containing explicitly the disorder: we have, for example, \( \langle \Sigma(-\frac{q}{2}) u(\frac{q}{2}) \rangle \to -\frac{D}{T V_0} \sum_a \langle \rho_a(-\frac{q}{2}) u_1(\frac{q}{2}) \rangle_{\text{eff}} \). After
some manipulations and using (9), we obtain:

\[
I_d = \int d^2 q x^2 a^2_0 \int_r \left[ e^{-i q r} + c.c. \right] C_d(r),
\]

\[
I_a = -\int \Delta f q u_0 \sqrt{N_j} a^4 D \int_r \left[ e^{-i q r} - c.c. \right] C_a(r),
\]

where \(C_d(r) = \langle e^{i(\phi_a(\mathbf{r})-\phi_b(\mathbf{r}))} \rangle_{\text{eff}}\) and \(C_a(r) = \frac{1}{\pi n} \sum_{a,b} \langle e^{i(\phi_a(\mathbf{r})-\phi_b(\mathbf{r}))} \rangle_{\text{eff}}\) are the positional correlation functions controlling the behavior of each contribution.

We notice that the intensity of the peaks at \(q = Q + K\) and \(q = K - Q\) is symmetric, as in the case of a pure system, for the displacement term \(I_d\), but it is antisymmetric for \(I_a\). The sum of these two terms leads to an asymmetry of the peaks. Fig. 1 shows the behavior of the different contributions.

Following the method used in (10) for flux lines in presence of weak disorder, we can calculate the various terms in (10). We look for the best trial Gaussian Hamiltonian \(H_0 = \int \text{d}x G_a(q) \phi_a(q) \bar{\phi}_b(-q)\) in replica space, which approximates (9). Defining

\[
B_{ab}(r) = \langle (\phi_a(r) - \phi_b(0))^2 \rangle \eta
\]

\[
= 2T \int_q \left[ \tilde{G}(q) - G_{ab}(q) \cos qr \right],
\]

where \(\tilde{G}\) is the diagonal element of \(G_{ab}\), and using the Gaussian approximation, the positional correlation functions become \(C_d(r) = \frac{1}{\pi T} \sum_{a,b} e^{-\frac{\tilde{B}_{ba}(r)}{2}}\) and \(C_a(r) = e^{-\frac{\tilde{B}_{aa}(r)}{2}}\), where \(\hat{B}\) is the diagonal element of \(B_{ab}\). Two general classes of solutions exist for this problem: while the first class preserves the permutation symmetry of the replica (RS), the second class (RSB) breaks the replica symmetry. It has been shown (10) that the stable solution for \(d > 2\) corresponds to the RSB class, while the RS solution remains valid at short distance. \(C_d\) is similar to the correlation calculated for flux lines (10) and will be discussed later. To evaluate the contribution of the interference between disorder and displacement we factorize the antisymmetric term \(C_a(r) = \chi(r) C_d(r)\). We first consider the RS approximation:

\[
\chi(r) = \frac{1}{T} \left[ 1 - e^{-T f \eta G_c(q) \cos qr} \right],
\]

where \(G_c = \frac{1}{\eta T}\) is the connected part of \(G_{ab}\). In \(d = 3\) we estimate

\[
C_{a,\text{RS}}^R(r) \sim \frac{2\pi^2}{c^2 T} e^{-\frac{\phi_x^2}{2}}.
\]

The triplet term can be evaluated in an analogous way, but it gives non-zero contributions only considering higher order harmonic terms in the electron density. Equation (13) becomes: \(\rho(x) = \rho_0 \cos(n(Qx + \phi(x)))\), with \(n = 1, 2\). As we have already found for \(I_a\), we get an antisymmetric term with a prefactor \(\Delta f^2 q u_0 N_j a^4 D\) and a correlation \(C_{\text{tripl.}} = \frac{2\pi^2}{c^2} \sum_{a,b,c} \langle e^{i(\phi_a(\mathbf{r})-\phi_b(\mathbf{r}))}\rangle_{\text{eff}}\) in \(d = 3\) and at low temperature we finally obtain

\[
C_{\text{tripl.},\text{RS}}^R(r) \sim \frac{2\pi}{c^2 T} e^{-\frac{\phi_x^2}{2}}.
\]

It is interesting to evaluate, at this stage, the relative weight of the two antisymmetric terms in a satellite peak. We introduce the Fukuyama-Lee length (or Larkin-Ovchinnikov length) (14,23) \(R_a = (c^2 / D)^{1/4 - d}\) (for \(d = 3\) \(R_a = c^2 / D\)) such that \(\phi\) varies on scale given by the length \(R_a\). The ratio of the two intensity peaks is:

\[
\frac{I_{\text{tripl.}}}{I_a} = -\frac{\Delta f^2}{\pi T} \sqrt{N_j} \frac{\sqrt{a}}{R_a}.
\]

For weak disorder \(R_a \gg a\) it follows that \(I_a \gg I_{\text{tripl.}}\) and we thus need only to consider \(I_a\) and \(I_d\).

Since for \(d = 3\) the RS solution is unstable, to obtain the correct physics one has to look for the RSB method. Within this scheme (10), the off diagonal elements of \(G_{ab}(q)\) are parameterized by \(G(q,v)\) where \(0 < v < 1\) and the solution is characterized by a variational breakpoint \(v_c\). The form of the symmetric part is given in (10):

\[
\tilde{C}_d(r) \sim e^{-\frac{\phi_x^2}{2}} \left( \frac{1}{r} \right)^\eta
\]

where \(\phi_x^2 \sim \frac{2T}{\eta}\) measures the strength of thermal fluctuations, and \(\eta \sim 1\) is the Bragg glass exponent in \(d = 3\). At low temperature one has \(l \sim R_a\). The algebraic behavior of (14) is controlled by small \(v\) \((v < v_c)\). Values of \(v\) above the breaking point \((v > v_c)\) give small distance contribution. Finally one finds \(v_c = \frac{1}{8} \frac{\phi_x^2 a}{T}\).

To fully characterize the spectrum it still remains to evaluate \(\chi(r)\) in the RSB scenario:

\[
\chi(r) = \frac{1}{T} \left[ 1 - \int_0^1 dv e^{-T \int_q \langle \Delta \tilde{G}(q) - G_q(q) \rangle \cos qr} \right].
\]
we have is sketched in Fig. 2. As for the Replica Symmetry case, terms: $K_{\delta}(17)$ becomes:

$$\tilde{G}(q) - G(q, v) = \frac{1}{c} \left[ \frac{1}{q^2 + l^2} + \frac{2}{l^2} \int_{l/v_c}^1 dt \frac{1}{(q^2 + (\frac{t}{4})^2)^{\frac{3}{2}}} \right].$$

(18)

By integrating (18) over $q$ and with some manipulations, [14] becomes:

$$\chi(r) = \frac{\nu_c}{T} \left[ 1 - \int_0^1 dz \exp \left( -8\pi^3 \int_z^1 \frac{dt}{l} e^{-rt/l} \right) \right].$$

(19)

The low temperature behavior ($l \sim R_a$) of this term is sketched in Fig. 2. As for the Replica Symmetry case, we have $C_a(r) \propto \frac{1}{r} e^{-\frac{a(r)}{r}}$. We can now compare the two terms:

$$I_d(K + Q) = \frac{\pi}{\nu_c} K^2 u_0^2 \int_r \frac{(R_a)}{r}^y$$

$$I_a(K + Q) = -2\pi^2 \Delta f \sqrt{N_f} \sqrt{\frac{a}{R_a}} K u_0 \int_r \frac{(R_a)}{r}^y \frac{b a}{r}.$$

(20)

After executing the $d$-dimensional Fourier Transforms, we conclude that both terms are divergent: in particular, $I_d \propto \frac{1}{q^2 + l^2}$ and $I_a \propto \frac{1}{q^2 + l^2}$. This effect, shown in Fig. 1, is a clear sign of a quasi-long range positional ordered phase. We have found that the peak at $K + Q$ is smaller than the $K - Q$ one, as the potential between the impurity and CDW is repulsive (we would have the opposite asymmetry in case of an attractive potential). We observe that for an ideal infinite resolution experiment, the symmetric term would be dominant, since $C_d(r)$ decays to zero less rapidly than $C_a(r)$. However, if the divergence in (20) is cut by the finite resolution of the experiment both terms should be taken into account because $I_d$ is quadratic in the small parameter $K u_0$ whereas $I_a$ is only linear.

The powerlaw lineshape is obtained for a short range elasticity. If the Coulomb interaction is unscreened, as might be the case in fully gapped systems such as the blue bronzes, the dispersion of $c_1$ should be kept in (5).

In that case $c_1(q) \sim \frac{q^2}{l^2}$, which leads to peaks diverging even faster than $[20] [21]$. On the experimental side few detailed diffraction spectra are available at the moment. One case is doped blue bronzes where the lineshape corresponding to the CDW has been obtained after substraction of a Friedel oscillation contribution [17]. The observed asymmetry of the peaks would be compatible with both strong and weak pinning. However given the short correlation length extracted from the data, this particular experiment is most likely still in the strong pinning regime. It would thus be highly desirable to have more detailed analysis of the lineshapes either in this compound, for different impurity concentrations, or in less disordered systems, where one can expect a Bragg glass behavior.

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[25] The asymmetry of the two peaks can be very strong: it has been observed that the intensity of the lowest peak can even be smaller than the Laue scattering intensity. In this case one talks of white line in the spectrum.