PHYSICAL REVIEW B **68**, 140201(R) (2003)

X-ray diffraction of a disordered charge density wave

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(Received 9 May 2003; published 2 October 2003)

We study the x-ray-diffraction spectrum produced by a collectively pinned charge density wave, 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.

DOI: 10.1103/PhysRevB.68.140201 PACS number(s): 71.45.Lr, 74.25.Qt, 78.70.Ck, 79.60.Ht

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,[7–9] instead of the power-law growth as for interfaces. The positional order is only algebraically destroyed,[9,10] leading to divergent Bragg peaks and a nearly perfect crystal state. Quite remarkably, it was shown that for weak disorder this solution is *stable* to the proliferation of topological defects, and thus a thermodynamically stable phase having both glassy properties and quasilong range positional order exists.[10] This phase, nicknamed Bragg glass, has prompted many further analytical and experimental studies (see, e.g., Refs. 11,12 for reviews and further references). Although its existence can be tested indirectly by the consequences on the phase diagram of vortex flux lines, the most direct proof is to measure the predicted algebraic decay of the positional order. Such a measurement can be done by means of diffraction experiments, using either neutrons or x rays on the crystal. Neutron-diffraction experiments have recently provided unambiguous evidence[13] of the existence of the Bragg glass phase for vortex lattices.

Another periodic system in which one can expect a Bragg glass to occur are charge density waves,[2] where the electronic density is spatially modulated. Disorder leads to the pinning of the CDW.[14] In such systems very high-resolution x-ray experiments can be performed.[15] The resolution is, in principle, much higher than the one that can be achieved by neutrons for vortex lattices, consequently, CDW systems should be prime candidates to check for the existence of a Bragg glass state. However, compared to the case of vortex lattices, the interpretation of the spectrum is much more complicated for two main reasons: (i) the phase of the CDW is the object described by an elastic energy, whereas the x rays probe the displacements of the atoms in the crystal lattice (essentially, a cosine of the phase); (ii) since the impurities substitute some atoms of the crystal, the very presence of the impurities changes the x-ray spectrum. This generates nontrivial terms of interference between disorder and atomic displacements.[15,16] It is thus necessary to make a detailed theoretical analysis for the diffraction due to a pinned CDW. The study of the spectrum has been carried out so far either for strong pinning or at high temperatures.[15–18].

In this paper we focus on the low-temperature limit where a well-formed CDW exists, and on weak disorder, 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,[16] 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[19] for the total diffraction intensity in a crystal is given by

$$I(q)=\frac{1}{V}\sum_{i,j} e^{-iq(R_i-R_j)}\overline{\langle f_i f_j e^{-iq(u_i-u_j)}\rangle}, \tag{1}$$

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 $\overline{\langle \dots \rangle}$ 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

$$I(q)=\overline{f}^2\sum_K \delta(q-K)+\Delta f^2 N_I, \tag{2}$$

where $\Delta f=f_I-f$ is the difference between the impurity I and the host atom scattering factors, $N_I=n_I(1-n_I)$, where n_I is the impurity concentration and $\overline{f}$ is the average scattering factor. The usual Bragg peaks, in correspondence to the reciprocal lattice vectors K, arise from the first term in Eq. (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 deformation:[2]

$$\rho(x)=\rho_0\cos[Qx+\phi(x)]. \tag{3}$$

ϕ is the phase of the charge density wave and Q is the modulation vector. The associated Hamiltonian reads

$$H=\int d^dx\frac{c}{2}[\nabla\phi(x)]^2\pm V_0\int d^dx\Sigma(x)\rho(x), \tag{4}$$

where d is the dimension of the space. The first term in Hamiltonian (4) represents the elasticity. The second term 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 $\overline{\Sigma(x)\Sigma(y)}=N_I\delta(x-y)$, 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, ρ_0 is absorbed in V_0 and we define the disorder strength $D=V_0^2N_I$.

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

$$u(x)=\frac{u_0}{Q}\nabla\cos[Qx+\phi(x)]. \tag{5}$$

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 Eq. (1):

$$I(q)=\int_r\langle\overline{f_{r/2}f_{-r/2}e^{-i\delta q[u(r/2)-u(-r/2)]}}\rangle. \tag{6}$$

where $\int_r=(1/a^d)\int d^dre^{-i\delta qr}$ and $f_{r/2}=\overline{f}+\Delta fa^{d/2}\Sigma(r/2)$. In Eq. (6) we have applied the standard decomposition in center of mass R and relative r coordinates $[x=R+r/2$ and $y=R-(r/2)]$. 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 Eq. (6) as powers of Ku_0. Developing up to the second order we get[17]

$$I(q)=I_{\rm d}+I_{\rm a}+I_{\rm tripl}, \tag{7}$$

with

$$I_{\rm d}=\overline{f}^2q^2\int_r\overline{\left\langle u\left(\frac{r}{2}\right)u\left(-\frac{r}{2}\right)\right\rangle},$$

$$I_{\rm a}=-iq\Delta fa^{d/2}\overline{f}\int_r\left\langle\overline{\Sigma\left(-\frac{r}{2}\right)u\left(\frac{r}{2}\right)-\Sigma\left(\frac{r}{2}\right)u\left(-\frac{r}{2}\right)}\right\rangle,$$

$$I_{\rm tripl}=-iq\Delta f^2a^d\int_r\left\langle\overline{\Sigma\left(-\frac{r}{2}\right)\Sigma\left(\frac{r}{2}\right)\left[u\left(\frac{r}{2}\right)-u\left(-\frac{r}{2}\right)\right]}\right\rangle.$$

While the contribution $I_{\rm d}$ represents the intensity due to the atomic displacements alone, the contributions $I_{\rm a}$ and $I_{\rm tripl}$ 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 the absence of disorder ($D=0$ and $\phi\sim$const), the displacement term has the form $I_{\rm d}=f^2q^2u_0^2\Sigma_K\delta(q+K\pm Q)$ 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,[15,16,18] 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 $I_{\rm a}$ was evaluated by means of models[16–18] which describe the pinning by imposing a constant value ϕ_0 to the phase in Eq. (3) in proximity of each impurity, and $I_{\rm tripl}$ 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, ϕ_0 is not constant and, for sufficiently large domains, one should have[16] $I_{\rm a}\propto\overline{\cos(\phi_0)}\sim 0$. 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.[10,23] We first perform the average over the disorder using the standard replica techniques. The replicated Hamiltonian corresponding to Eq. (4) is

$$H_{\rm eff}=\int d^dr\sum_a\frac{c}{2}(\nabla\phi_a)^2-\frac{D}{T}\sum_{a,b}\cos[\phi_a(r)-\phi_b(r)], \tag{8}$$

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 Hamiltonian (4) to its replicated version we also need to change the correlation functions containing explicitly the disorder: we have, for example, $\langle\overline{\Sigma(-r/2)u(r/2)}\rangle\to-(D/nTV_0)\Sigma_{a,b}\langle\rho_a(-r/2)u_b(r/2)\rangle_{\rm eff}$. After some manipulations and using Eq. (3), we obtain

$$I_{\rm d}=\overline{f}^2q^2u^2{}_0\int_r[e^{-iQr}+\text{c.c.}]C_{\rm d}(r), \tag{9}$$

$$I_{\rm a}=-\overline{f}\Delta fqu_0\sqrt{N_Ia^dD}\int_r[e^{-iQr}-\text{c.c.}]C_{\rm a}(r),$$

where $C_{\rm d}(r)=1/n\Sigma_a\langle e^{i[\phi_a(r/2)-\phi_a(-r/2)]}\rangle_{\rm eff}$ and $C_{\rm a}(r)=(1/Tn)\Sigma_{a,b}^n\langle e^{i[\phi_a(r/2)-\phi_b(-r/2)]}\rangle_{\rm eff}$ 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 $I_{\rm d}$, but it is antisymmetric for $I_{\rm a}$. The sum of these two terms leads to an asymmetry of the peaks. Figure 1 show the behavior of the different contributions.

Following the method used in Ref. 10 for flux lines in the presence of weak disorder, we can calculate the various terms in Eq. (9). We look for the best trial Gaussian Hamiltonian $H_0=\int_qG_{ab}(q)\phi_a(q)\phi_b(-q)$ in replica space, which approximates Eq. (8). Defining

$$B_{ab}(r)=\langle[\phi_a(r)-\phi_b(0)]^2\rangle_0$$
$$=2T\int_q[\tilde{G}(q)-G_{ab}(q)\cos qr], \tag{10}$$

where $\tilde{G}$ is the diagonal element of G_{ab}, and using the Gaussian approximation, the positional correlation functions become $C_{\rm a}(r)=(1/nT)\Sigma_{a,b}^ne^{-B_{ab}(r)/2}$ and $C_{\rm d}(r)=e^{-\tilde{B}/(r)2}$,

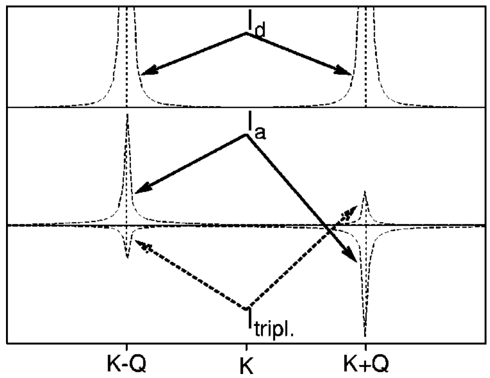

FIG. 1. Intensities of the different contributions to satellite peaks. The more divergent term I_d, is symmetric. I_a and I_{tripl} are antisymmetric, with $I_a \gg I_{tripl}$.

where $\tilde{B}$ 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 distances. 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

$$\chi(r)=\frac{1}{T}\left[1-\exp\left[-T\int_q G_c(q)\cos qr\right]\right], \tag{11}$$

where $G_c=1/cq^2$ is the connected part of G_{ab}. In $d=3$ we estimate

$$C_a^{RS}(r)\sim\frac{2\pi^2}{cr}e^{-\tilde{B}(r)/2}. \tag{12}$$

The triplet term can be evaluated in an analogous way, but it gives nonzero contributions only considering higher-order harmonic terms in the electron density. Equation (3) 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 $\propto\Delta f^2 q u_0 N_i a^d D$ and a correlation $C_{tripl}=(1/2nT^2)\Sigma_{a,b,c}^{n}\langle(e^{-i[\phi_c(r/2)-2\phi_a(r/2)+\phi_b(-r/2)]}\rangle_{eff}$. In $d=3$ and at low temperatures we finally obtain

$$C_{tripl}^{RS}(r)\sim\frac{2\pi}{c^2ar}e^{-\tilde{B}/2}. \tag{13}$$

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-Ovchinikov length)[14,24] $R_a=(c^2/D)^{1/4-d}$ (for $d=3$, $R_a=c^2/D$) such that ϕ varies on scale given by the length R_a. The ratio of the two intensity peaks is

$$\frac{I_{tripl}}{I_a}=-\frac{\Delta f}{\pi \bar{f}}\sqrt{N_I}\sqrt{\frac{a}{R_a}}. \tag{14}$$

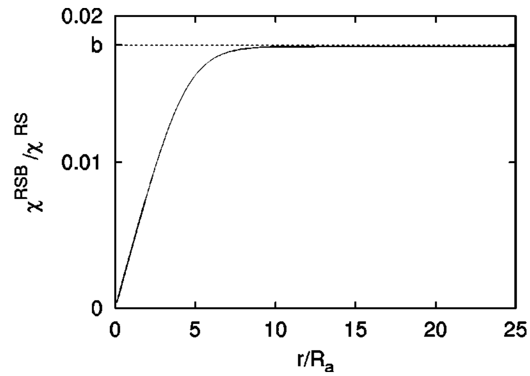

FIG. 2. Ratio between the RSB and RS solutions for $\chi(r)$. At large distance this ratio tends towards a constant value b, where $b \sim 0.018$. This means that the RSB solution affects $\chi(r)$ only by a multiplicative factor.

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

Since for $d=3$ the RS solution is unstable, so 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 parametrized 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 Ref. 10:

$$C_d(r)\sim e^{-\phi_T^2/2}\left(\frac{l}{r}\right)^{\eta}, \tag{15}$$

where $\phi_T^2 \simeq 2T/\pi ca$ 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 Eq. (15) is controlled by small v $(v<v_c)$. Values of v above the breaking point $(v>v_c)$ give the small distance contribution. Finally, one finds $v_c=\frac{1}{8}\phi_T^2(a/l)$.

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

$$\chi(r)=\frac{1}{T}\left[1-\int_0^1 dv\exp\left\{-T\int_q[\tilde{G}(q)-G(q,v)]\cos qr\right\}\right]. \tag{16}$$

Restricting to the case $d=3$, we write

$$\tilde{G}(q)-G(q,v)=\frac{1}{c}\left[\frac{1}{q^2+l^{-2}}+\frac{2}{l^2}\int_{v/v_c}^{1}dt\frac{1}{\left[q^2+\left(\frac{t}{l}\right)^2\right]^2}\right]. \tag{17}$$

By integrating Eq. (17) over q and with some manipulations, Eq. (16) becomes:

$$\chi(r)=\frac{v_c}{T}\left[1-\int_0^1 dz\exp\left(-8\pi^3\int_z^1\frac{dt}{t}e^{-rt/l}\right)\right]. \tag{18}$$

The low-temperature behavior $(l\sim R_a)$ of this term is shown in Fig. 2. As for the replica symmetry case, we have $C_a(r)\propto(1/r)e^{-\tilde{B}/(r)2}$. We can now compare the two terms:

$$I_{\mathrm{d}}(K+Q)=\overline{f}^2K^2u_0^2\int_r\left(\frac{R_a}{r}\right)^{\eta} \tag{19}$$

$$I_{\mathrm{a}}(K+Q)=-2\pi^2\overline{f}\Delta f\sqrt{N_I}\sqrt{\frac{a}{R_a}}Ku_0\int_r\left(\frac{R_a}{r}\right)^{\eta}\frac{ba}{r}.$$

After executing the *d*-dimensional Fourier transforms, we conclude that both terms are divergent: in particular, $I_{\mathrm{d}}\propto 1/q^{d-\eta}$ and $I_{\mathrm{a}}\propto 1/q^{d-\eta-1}$. 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 $C_{\mathrm{d}}(r)$ decays to zero less rapidly than $C_{\mathrm{a}}(r)$. However, if the divergence in Eq. (19) 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 Ku_0 whereas I_a is only linear.

The power-law line shape is obtained for a short-range isotropic elasticity. The elasticity is actually anisotropic[20] along the Q direction and has the form

$$H_{\mathrm{el}}=\int dxd^{d-1}y\frac{c_1}{2}(\partial_x\phi)^2+\frac{c_2}{2}(\partial_y\phi)^2, \tag{20}$$

where $x\|Q$ 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 Eq. (4) by redefining the spatial variables $x'=x/\sqrt{c_1}$ and $y'=y/\sqrt{c_2}$, with $c=(c_1c_2^{d-1})^{1/2}$. 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 (20) 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 wants to examine the short-range regime, one should keep the dispersion of $c_1(q)\sim 1/q^2$. This leads to a different behavior of the peaks with respect to Eq. (19). In particular we have found $I_{\mathrm{d}}\propto 1/q^3$ and $I_{\mathrm{a}}\propto 1/q$. This means that, in this case, the peaks should be even more divergent than in Eq. (19), but the asymmetric term becomes negligible. All details concerning this calculation will be examined elsewhere.[21]

On the experimental side few detailed diffraction spectra are available at the moment. One case is doped blue bronzes where the line shape corresponding to the CDW has been obtained after subtraction of a Friedel oscillation contribution.[15] 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 in the strong pinning regime, whereas our calculations concern the weak pinning limit. It would thus be highly desirable to have more detailed analysis of the line shapes either in this compound, for different impurity concentrations, or in less disordered systems, where one can expect a Bragg glass behavior.

We thank J.-P. Pouget and S. Ravy for stimulating discussions.

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