Coexistence of short- and large-scale phase variations in a charge-density wave weakly coupled to impurities

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We reexamine the problem of phase pinning of a three-dimensional charge-density wave (CDW) weakly coupled to impurities. Within an analytical approach, we find that the phase is adjusted both at the short scale, around individual impurity sites, and at the large scale, over extended domains containing many impurities. Both phase adjustments are important for the physical behavior and contribute to the understanding of experimental data as well as numerical simulation results in CDW systems. The former give the main contribution to energy and are responsible for the white-line effect observed in x-ray patterns. The latter determine the functional dependence of the threshold field $E_{\rm th}$ for nonlinear conduction. Unlike what was widely accepted, we find that $E_{\rm th}$ is not related to the total energy gained by phase spatial variations. This picture is consistent with that emerging from the recent experimental investigations of doped $NbSe_3$ and could contribute to clarifying the controversy on the pinning type in this material.

I. INTRODUCTION

Impurity pinning is a subject of continuing interest, because of its profound role for understanding the unusual properties of the low dimensional materials undergoing a charge-density wave (CDW) transition.[1] Extensive studies, from both theoretical and experimental sides, are especially devoted to the fascinating but difficult problem of CDW dynamics, in the interesting regime near the threshold field for nonlinear conduction. The understanding of the static properties is a prerequisite for understanding the more complex CDW dynamics. The conventional theory of the static CDW containing a random distribution of quenched impurities considers this system as a frustrated one, whose properties are governed by two antagonistic conditions: the intrinsic CDW *periodicity* tends to impose a *uniform* phase (usually modeled by an elastic energy term, proportional to the square of the phase gradient[2]), while the impurity potential prefers, due to the *random* impurity location, a *nonuniform* phase.[4,2,3] The spatial variation of the CDW phase $\varphi(\mathbf{r})$ is controlled by the Fukuyama-Lee-Rice (FLR) parameter ε.[2,3] It is defined as the ratio of the energy gained by adjusting the phase at the $j^{\rm th}$ impurity site to the value preferred by the impurity potential to the associated elastic energy cost. While the nature of pinning for large ε is clear, — the phase being (almost) perfectly adjusted to the values preferred by the impurity potential (strong pinning limit[2,3]) — the small ε limit turned out to be more delicate. By supposing that the phase variations around individual impurity sites are large ($\sim \pi$), the small energy gain from the impurity potential, insufficient to overcome the corresponding elastic energy cost for $\varepsilon < 1$, simply shows that the state with the uniform phase (whose energy is taken throughout zero) has a lower energy. Therefore, within the conventional approach,[2,3] the role of the phase variations at short scale, around individual impurities was ignored, and the attention was solely focused on the phase variations occurring at large scale, within extended regions containing many impurities. A scaling argument demonstrated that, for space dimensions less than four, one can diminish the energy with respect to that of uniform phase if the system breaks in phase coherent (Fukuyama-Lee) domains containing a large number of impurities.[5,2,3] For a three-dimensional system, — to which the present study is devoted —, this collective mechanism of pinning leads to an energy gain $|W_{\rm FLR}| \propto \varepsilon^4$, an average phase gradient $\overline{(\nabla\varphi)^2}_{\rm FLR}^{1/2} \propto 1/L_{\rm FLR} \propto \varepsilon^2$ ($L_{\rm FLR}$ — length of Fukuyama-Lee domain) and a ratio $r \equiv -W_{\rm imp}/W_{\rm el}$ of the energy gain from impurity potential to the elastic energy having the value $r_{\rm FLR} = 4/3$.

In contrast to these analytical results, numerical simulation studies on the ground state revealed a different dependence for the energy gain, $|W_{\rm NS}| \propto \varepsilon^2$, and the average phase gradient, $\overline{(\nabla\varphi)^2}_{\rm NS}^{1/2} \propto \varepsilon$,[6] while the parameter r tends to saturate at the value $r_{\rm NS} = 2$ for small ε.[7] Because $|W_{\rm FLR}|/|W_{\rm NS}| \propto \varepsilon^2$ ($\ll 1$ for $\varepsilon \ll 1$), one immediately arrives to the conclusion[8] that the state, where (only) large-scale phase variations[2,3] occur, cannot be the ground state of the interacting CDW-impurity system. An approach based on a simple variational ansatz proposed recently[8] succeeded to reproduce all aforementioned features found by the previous numerical simulations.[6,7] Unlike in the conventional FLR approach,[2,3] the approach developed in Ref. 8 demonstrated that, even for $\varepsilon \ll 1$, small phase adjustments at individual impurity sites are more efficient than large-scale phase variations in diminishing the total energy. Besides the good agreement found for the ε dependence of

energy and average phase gradient (reproducing the numerical simulation results within factors of order unity) and the absolute value of the parameter $r(=2)$, direct evidence on the role played by short-scale phase variations was given by the dependence of the CDW properties on impurity concentration in the artificial lattice used in Ref. 7 to minimize the energy, as revealed by the critical analysis[8] of the numerical simulation findings.

Despite the success of the analytical approach of Ref. 8 to account for the ground state properties (energy, phase gradient, r) found by previous numerical simulations[6,7] and the unambiguous demonstration that the FLR solution corresponds to a considerable higher energy, it failed to explain why the values of the threshold field found by numerical simulation in Ref. 7 obey the law $E_{\rm th} \propto \varepsilon^4$, in accord with the FLR predictions (cf. Table I of Ref. 8). On the other side, we shall refer to the recent reports on the doped $NbSe_3$, one of the CDW materials most extensively investigated experimentally. A detailed analysis showed that the threshold field values found in both $Ta_x Nb_{1-x}Se_3$ and $Ti_x Nb_{1-x}Se_3$ samples are proportional to the square of the concentration, $E_{\rm th} \propto x^2$, again consistent to the FLR prediction for weak pinning.[9] Subsequent high-resolution x-ray scattering experiments[10] gave further support to this interpretation: large-scale phase variations were observed and their characteristic length could be identified with the length of Fukuyama-Lee domains. However, a more recent analysis revealed that only very fast phase variations, with wavelength shorter than the amplitude correlation length ξ, are excluded.[11] Summarizing the foregoing discussion, one can conclude that, including either the short- or the long-scale phase variations, neither the approach of Ref. 8, nor the conventional FLR approach[2,3] can account for all numerical simulation and experimental results listed above.

Basically, both the FLR approach[2,3] and the recently proposed one[8] attempted to find analytically a state whose energy is smaller than that of the nonpinned CDW within certain simplifying assumptions on the manner in which the phase adjustments in the presence of impurities occur. Instead of solving the complicated problem of finding the function $\varphi(\mathbf{r})$ that minimizes the free energy functional, both assumptions introduced a variational parameter — the domain length $L_{\rm FLR}$ in Refs. 2, 3, the degree of local phase adjustment denoted by φ_0 in Ref. 8 — that can easily be determined analytically. While the solution obtained in Ref. 8, based on a simple variational ansatz that allows for phase adjustments at individual impurity sites (even) in the small ε limit, corresponding to a CDW whose energy is substantially lower than found by Lee and Rice,[3] unambiguously demonstrated that the latter cannot be the ground state, it can obviously rule out neither a state of lower energy, nor the occurrence of large-scale phase adjustments.

We shall present in this paper analytical results, obtained by minimizing the energy functional of the three-dimensional CDW weakly coupled to impurities with respect to the phase $\varphi(\mathbf{r})$, which reconfirm many results derived in Ref. 8. The method used here is not new; it has been used in a number of previous investigations,[4,12–14] where, nevertheless, different aspects have been emphasized. On the other side, numerical simulations[6,7] posterior to these analytical studies did not attempt to establish any connection between the two methods. The present study demonstrates that, contrary to those claimed previously,[3] short- and large-scale variations of the phase of a three-dimensional CDW weakly coupled to impurities coexist, but their roles are different. In addition to the good agreement with numerical simulation results already found in Ref. 8, the present findings are in accord to the dependences $E_{\rm th} \propto \varepsilon^4$ and $E_{\rm th} \propto n_i^2$ (n_i — impurity concentration) found in the numerical simulation[7] and in experiments on doped $NbSe_3$,[9] respectively, as well as with the formation of large phase coherent domains[10] and the exclusion of very fast phase variations[11] in the latter system. However, contrary to what was claimed previously,[2,3,15] no simple relationship is found here between the threshold field $E_{\rm th}$ for depinning and the gain in the total energy due to the phase adjustments produced by the CDW-impurity interaction. As already noted,[8] the occurrence of phase adjustments at individual impurities is consistent with the variety of CDW materials in which the "white-line" effect was observed experimentally,[16] a finding incompatible with the FLR model of weak pinning.[16,8] Therefore, although recovering results derived previously, the present investigation shows that the physical picture of the three-dimensional CDW weakly coupled to impurities is more complex than that emerging from the previous studies.

The remaining part of this paper is organized as follows. The model of the deformable CDW containing impurities is described in Sec. II, where the effect of short-scale phase variations is also investigated. Sec. III is devoted to the large-scale phase adjustments. Some discussions and conclusions are made in the last Sec. IV.

II. EFFECT OF SHORT-SCALE PHASE ADJUSTMENTS

In the presence of impurities, both the phase φ and amplitude of the CDW are distorted. Over large distances, — within the mean-field theory, larger than the amplitude (BCS) correlation length ξ— the amplitude fluctuations are energetically more expensive than the phase fluctuations. Therefore, many investigators[2,4,12–14,7] disregarded the variations within the correlation length ξ and described the CDW by a free energy functional W expressed solely in terms of $\varphi(\mathbf{r})$ (phase-only theory), obtained by making a Ginzburg-Landau gradient expansion. In the static limit, zero temperature and in the absence of an applied electric field, one can write

$$W \equiv W_{\rm el} + W_{\rm imp} = \frac{\kappa}{2}\int_V \mathbf{d}^3\mathbf{r}\,|\nabla\varphi(\mathbf{r})|^2 + E_i\int_V \mathbf{d}^3\mathbf{r}\,n(\mathbf{r})\cos\left[\mathbf{Q}\mathbf{r}+\varphi(\mathbf{r})\right]. \quad (1)$$

Here, κ is the CDW elasticity, $\mathbf{Q}$ is the CDW wave vector, and E_i (>0) is the pinning strength, proportional to the density of CDW condensate and $\mathbf{Q}$-Fourier component of electron-impurity potential. Because of impu-

rity randomness, the fluctuation of the impurity concentration $n(\mathbf{r}) \equiv \sum_{j \le N_i} \delta(\mathbf{r} - \mathbf{R}_j) - n_i$ has the correlator $\langle n(\mathbf{r}) n(\mathbf{r}') \rangle_i = n_i \delta(\mathbf{r} - \mathbf{r}')$, $\langle \rangle_i$ denoting throughout impurity averaging and $n_i \equiv N_i/V$ is the mean concentration (V — total volume).[4] To express the gradient term of Eq. (1) in an isotropic form, we have assumed that the transverse correlation lengths are larger than the interchain spacings and performed an appropriate length scaling.[3] To find the ground state of the CDW with quenched random impurities, one has to determine the function $\varphi(\mathbf{r})$, which minimizes the sum of the two competing energies $W_{\rm el}$ and $W_{\rm imp}$ entering the functional (1).

Following Refs. 12–14, we shall look for a minimum of Eq. (1) corresponding to a system in which (almost) independent phase coherent domains, containing many impurities ($n_i \Omega \gg 1$, Ω — domain volume), are formed. For each domain $\mathcal{D}$, one can define a collective phase coordinate $\psi_{\mathcal{D}} = \langle \varphi(\mathbf{r}) \rangle_{\mathcal{D}} = \int_\Omega \mathbf{d}^3\mathbf{r} \varphi(\mathbf{r})/\Omega$ — $\langle \rangle_{\mathcal{D}}$ standing throughout for the spatial average over the domain $\mathcal{D}$ — and split $\varphi(\mathbf{r}) = \psi_{\mathcal{D}} + \theta(\mathbf{r})$.

By supposing that the CDW within a domain $\mathcal{D}$ does not experience elastic forces from other domains (independent domain approximation),[12,13] the boundary terms give a vanishing contribution and the stationarity of the functional (1) leads to

$$\nabla^2 \theta(\mathbf{r}) = -n_i^{-1/3} \varepsilon \, n(\mathbf{r}) \sin[\mathbf{Q}\mathbf{r} + \psi_{\mathcal{D}} + \theta(\mathbf{r})], \qquad (2)$$

where the FLR parameter $\varepsilon = n_i^{1/3} E_i/\kappa$ has been introduced.[17] The above equation can be rewritten in integral form by means of a Green function.[4,12–14] To solve this integral equation diagrammatically,[4,13] particularly convenient is to use a Green function, whose average over domains vanishes, $\langle G(\mathbf{r}) \rangle_{\mathcal{D}} = 0$. Straightforward analysis shows that, rather than spherical domains,[4,12,13] cubic domains (of volume $\Omega = L^3$)[14] are more convenient for calculations and will, therefore, be used below. Imposing, furthermore, periodic boundary conditions, one gets the following Green function:[14]

$$G(\mathbf{r}) = L^{-3} \sum_{\mathbf{k} \neq 0} \mathbf{k}^{-2} e^{-i\mathbf{k}\mathbf{r}}. \qquad (3)$$

The integral version of Eq. (2) reads

$$\theta(\mathbf{r}) = n_i^{-1/3} \varepsilon \int_\Omega \mathbf{d}^3\mathbf{r}' G(\mathbf{r} - \mathbf{r}') \, n(\mathbf{r}') \sin[\mathbf{Q}\mathbf{r}' + \psi_{\mathcal{D}} + \theta(\mathbf{r}')]. \qquad (4)$$

A diagrammatic technique was previously developed to solve Eq. (4) by iteration.[4,13] Simple expressions can be obtained for the correlation function of the CDW order parameter and the mean square value of θ, because only zeroth order terms give nonvanishing contributions:[14]

$$\langle \exp\{i[\theta(\mathbf{r}) - \theta(\mathbf{r}')]\} \rangle_i = \exp(-|\mathbf{r} - \mathbf{r}'|/L_c),$$
$$\overline{\theta^2} \equiv \langle \theta^2 \rangle_{i,\mathcal{D}} \simeq (2\pi)^2 \frac{L}{148 L_c},$$
$$L_c = \frac{16\pi}{n_i^{1/3} \varepsilon^2}. \qquad (5)$$

Here and hereafter, the bar will be used to denote the double average over space and impurity distribution. Let us proceed with the evaluation of the various contributions to the total energy. It is convenient to use dimensionless energy densities $w = W/(V \kappa n_i^{2/3})$. By combining Eqs. (2) and (4) and applying the diagrammatical method,[4,13] the elastic energy per domain associated with the fluctuations of θ can also be obtained in a closed form:

$$w_{{\rm el},\theta} = \frac{n_i^{-2/3}}{2} \overline{|\nabla\theta|^2} = \frac{\varepsilon^2}{4} n_i^{-1/3} G(\mathbf{r})|_{\mathbf{r}=0}. \qquad (6)$$

As already mentioned, a phase-only description is meaningful only if too fast spatial phase variations are explicitly excluded. This is in accord with recent experimental findings on $NbSe_3$.[11] To this aim, the summation over $\mathbf{k}$ in Eq. (6) has to be cut off at large wave vectors ($|\mathbf{k}| > k_c$). This yields a finite value of $G(\mathbf{r})|_{\mathbf{r}=0} \sim k_c$. Restricting the summation over $\mathbf{k}$ in each direction to the range $(-k_c, +k_c)$ in Eq. (3), one gets $G(\mathbf{r})|_{\mathbf{r}=0} = 0.0613\, k_c$. Within the mean-field theory of CDW, k_c is of the order ξ^{-1}.

Unlike the elastic term, the CDW-impurity interaction energy cannot be expressed in closed analytical form. If we set $\theta = 0$ in the right-hand side of Eq. (4) and insert this expression in Eq. (1), we easily get the following expression for the energy gain from the impurity potential:

$$w_{{\rm imp},\theta} = -\frac{\varepsilon^2}{2} n_i^{-1/3} G(\mathbf{r})|_{\mathbf{r}=0}. \qquad (7)$$

Notice that this contribution comes from the adjustment of the local phase $\theta(\mathbf{r})$ and not from the tendency of the average phase $\psi_{\mathcal{D}}$ to conform to the fluctuations of the impurity potential in the domain $\mathcal{D}$. However, as suggested by Eq. (5), this zeroth order approximation is meaningful only if the domain length L is sufficiently small and the relevant quantities can be expressed as convergent expansions in powers of $\overline{\theta^2}$.

III. EFFECT OF LARGE-SCALE PHASE ADJUSTMENTS

Using the explicit form (3) of the Green function in Eq. (4), one can check by straightforward calculations that the average over domain boundary of the phase gradient vanishes, $\oint d\mathbf{S} \cdot \nabla\theta = 0$. Integrated over the domain volume, Eq. (2) then gives

$$\Omega F(\psi_{\mathcal{D}}) \equiv n_i^{-1/3} \varepsilon \int_\Omega \mathbf{d}^3\mathbf{r} \, n(\mathbf{r}) \sin[\mathbf{Q}\mathbf{r} + \psi_{\mathcal{D}} + \theta(\mathbf{r})] = 0. \qquad (8)$$

In accord to the independent domain approximation used in the derivation of Eq. (2), the above equation shows that the force F exerted on the CDW by impurities in the domain $\mathcal{D}$ vanishes (in the absence of an applied field[18]). Notice, however, that Eqs. (4) and (8) must be solved self consistently. By inserting the former into the latter, one gets an equation determining $\psi_{\mathcal{D}}$; then Eq. (4) will give

the optimum value of the local phase $\theta(\mathbf{r})$. Therefore, the phase $\varphi(\mathbf{r}) = \psi_{\mathcal{D}} + \theta(\mathbf{r})$ at each point will be fixed (pinned CDW).

Klemm and Schrieffer[13] showed by iterating Eq. (4) that the force per domain $F(\psi)$, defined by Eq. (8), computed for an arbitrary value of ψ, can be expanded in Fourier series:

$$F(\psi) = \sum_{m\geq 1} F_m \cos(m\,\psi + \vartheta_m), \tag{9}$$

where both amplitudes F_m and phases ϑ_m strongly dependent upon the particular impurity configuration. Nevertheless, for sufficiently large domains ($n_i\Omega \gg 1$), the Fourier coefficients F_m can be estimated, within uncertainty factors of order unity, by replacing them with their root mean square values, and the latter can be computed from the force-force correlations $\langle F(\psi + \psi_1)\,F(\psi_1)\rangle_i$.[13] The force-force correlation can be calculated either perturbatively, by expanding in powers of ε^2, or iterating Eq. (4). It can be shown that in the nth order in ε^2, each diagram contributing to $\langle F(\psi + \psi_1)\,F(\psi_1)\rangle_i$ behaves as $\Omega^{-1} n_i^{1/3}\,\varepsilon^2\,\overline{\theta^2}^{n-1}$;[13] so, the perturbation series converges for sufficiently small values of $\overline{\theta^2}$. Retaining only the contributions with $n = 1, 2$ to the force-force correlation, the expressions of the two Fourier coefficients in Eq. (9) can be obtained by performing calculations similar to those of Ref. 13. Using the fact that, for arbitrary ψ, the force acting on the CDW in a single domain is related to (the derivative with respect to ψ of) the energy gained from the impurity potential in the domain,[13] the lowest order contributions to the energy density can be expressed as

$$w_{\rm imp} - w_{{\rm imp},\theta} = w_{{\rm imp},\psi} = n_i^{-2/3}\left[F_1\,\cos(\psi_{\mathcal{D}} + \vartheta_{1,\mathcal{D}}) + \frac{1}{2}F_2\,\cos(2\psi_{\mathcal{D}} + \vartheta_{2,\mathcal{D}})\right], \tag{10}$$

where $F_1^2 = n_i^{1/3}\varepsilon^2(1-\overline{\theta^2})/\Omega$ and $F_2^2 = n_i^{1/3}\varepsilon^2\overline{\theta^2}/\Omega$, $\overline{\theta^2}$ and $w_{{\rm imp},\theta}$ are given by Eqs. (5) and (7), respectively. The above equation shows that, besides the energy gain $w_{{\rm imp},\theta}$ [Eq. (7)] due to the adjustments of the local phase θ, the energy can be further diminished if the average phase value $\psi_{\mathcal{D}}$ is properly adjusted to take advantage of the fluctuations in the impurity potential in the domain $\mathcal{D}$: the optimum value of $\psi_{\mathcal{D}}$, that minimizing $w_{{\rm imp},\psi}$, is just the value that ensures a vanishing force [Eq. (8)]. Setting $\overline{\theta^2} = 0$ in Eq. (10), the contribution to energy $w_{{\rm imp},\psi}$ coming from optimizing the value of the average phase $\psi_{\mathcal{D}}$ to the fluctuations of the impurity potential in the domain $\mathcal{D}$, $\psi_{\mathcal{D}} + \vartheta_{1,\mathcal{D}} = \pi$ ($F_1 > 0$) recovers the FLR expression, derived from simple considerations on a random walk, neglecting phase adjustments at individual impurities.[2,3] While the expression (10) (eventually extended to include also higher order harmonics) — basically, a result of perturbation theory — allows us to determine an optimum value of the average phase $\psi_{\mathcal{D}}$ within the domain $\mathcal{D}$, it cannot be used to find the domain size L. If the sample is sufficiently large and comprises many domains, the above argument indicates the tendency of $\psi_{\mathcal{D}}$'s to be adjusted to values depending on the impurity configuration in the various domains, fluctuating, therefore, uncorrelated from one domain to another. In accord with Eq. (1), these fluctuations in $\psi_{\mathcal{D}}$'s yield an extra elastic energy $w_{{\rm el},\psi}$ that cannot be determined within the independent domain approximation used above; nevertheless, it must be included in the total energy for samples consisting of many domains. Employing the FLR method,[2,3] one obtains

$$w_{{\rm el},\psi} = \frac{n_i^{-2/3}}{2\,V}\int_V \mathbf{d}^3\mathbf{r}\,\langle|\nabla\psi|^2\rangle_i = \frac{n_i^{-2/3}}{2}\,\overline{|\nabla\psi|^2} = \frac{3}{2\alpha}\left(n_i^{1/3}L\right)^{-2}, \tag{11}$$

where α is a numerical factor of order unity. Equations (6), (7), (10), and (11) give the various contributions to the total energy density associated to both short- and large-scale variations of the CDW phase deformed by impurities. The inspection of these equations shows that the terms related to the short-scale phase fluctuations, $w_{{\rm imp},\theta}$ and $w_{{\rm el},\theta}$, do not depend on the domain size L; nevertheless, $\overline{\theta^2}$ [and thence L, cf. Eq. (5)] must be small enough to ensure a convergent perturbation series and to justify thereby the truncation used in Eq. (10). To check this approximation, we shall determine the value of the domain length L_0 that minimizes the total energy, by furthermore assuming that the second harmonic gives a negligible contribution to Eq. (10). Using the value $\alpha = 3/\pi^2$ estimated by Fukuyama and Lee,[2] one easily gets $\overline{\theta^2} = 0.248$, corresponding to a domain length $L_0 = 1.079\,L_{\rm FLR} = 0.929\,L_c$, very close to the FLR length $L_{\rm FLR} = (2/\alpha)^2 n_i^{-1/3}\varepsilon^{-2}$, and a ratio between the amplitude of the second harmonic and that of the first harmonic $\rho \equiv F_2/(2F_1) = 0.287$. These values could be taken as indicating that the approximation used for evaluating the contribution to energy coming from adjusting the average phase ψ gives results reliable within factors of the order of unity.[19] Accepting this approximation, the various contributions to energy and average phase gradient, obtained from Eqs. (6), (7), (10), and (11), read

$$w_{{\rm el},\theta} = -\frac{1}{2}w_{{\rm imp},\theta} = \frac{1}{2}\,n_i^{-2/3}\,\overline{|\nabla\theta|^2} = 0.0153\,n_i^{-1/3}\,k_c\,\varepsilon^2, \tag{12}$$

$$w_{{\rm el},\psi} = -0.833\,w_{{\rm imp},\psi} = \frac{1}{2}\,n_i^{-2/3}\,\overline{|\nabla\psi|^2} = 0.002\,26\,\varepsilon^4. \tag{13}$$

Because of the nonvanishing $\overline{\theta^2}$, the value found from Eq. (13) for the ratio of the $|w_{{\rm imp},\psi}|/w_{{\rm el},\psi} = 1.20$ is slightly smaller than the value 4/3 obtained within the Lee-Rice approach.[7,8] It is useful to compare the energy gain and the average phase gradient, due to short- and large-scale variations. Using Eqs. (12) and (13), one easily gets $w_\theta/w_\psi = 0.68\,k_cL_0$ ($w_\zeta \equiv w_{{\rm imp},\zeta} + w_{{\rm el},\zeta}$, $\zeta = \theta,\,\psi$) and $\overline{|\nabla\theta|^2}/\overline{|\nabla\psi|^2} = 0.14\,k_cL_0$. For small ε, L_0

is much larger than the average spacing between impurities $n_i^{-1/3}$. On the other side, a phase-only description is justified for $k_c \sim 1/\xi$ and $n_i^{-1/3} > \xi$. The large value of $k_c L_0$ thus obtained demonstrates that the short-scale phase adjustments give the largest contribution to both energy and phase gradient.

In the presence of an applied field, the collective coordinate becomes time dependent, $\psi_{\mathcal{D}}(t)$. Klemm and Schrieffer investigated the dynamics of $\psi_{\mathcal{D}}(t)$ by assuming that $\partial\psi_{\mathcal{D}}(t)/\partial t$ is small with respect to the natural frequencies of internal degrees of freedom within the domain. Then, the internal deformations can be integrated out and an effective potential [whose truncated expression is given by Eq. (10)] is obtained; it depends only on $\psi_{\mathcal{D}}(t)$ and governs the dynamics of $\psi_{\mathcal{D}}$ (adiabatic approximation). Within the same approximation used above, a threshold field for nonlinear conduction can be found, behaving as[13]

$$E_{\rm th} \propto n_i^2 (E_i/\kappa)^4 = n_i^{2/3} \varepsilon^4. \tag{14}$$

This functional dependence is of the type found by Lee and Rice,[3] although they ignored the short-scale phase variations.

IV. DISCUSSIONS AND CONCLUSIONS

We have presented the above results obtained analytically for the three-dimensional CDW weakly coupled to a random distribution of quenched impurities. They will be compared below with those obtained from numerical simulation studies on the same system,[6,7] as well as with experimental data reported in CDW materials.

Abe found by numerical simulation on a related model that the pinning of the CDW phase produces, for small ε, an energy gain w proportional to ε^2 and an average phase variation among neighboring impurity sites proportional to ε.[6] This is exactly the behavior $w \simeq w_\theta \propto \varepsilon^2$ and $n_i^{-1/3} \overline{|\nabla\varphi|^2}^{1/2} \simeq n_i^{-1/3} \overline{|\nabla\theta|^2}^{1/2} \propto \varepsilon$ expressed by Eqs. (12) and (13) and demonstrates that the dominant contributions come from the short-scale phase adjustments. This ε dependence is indeed at variance with that expected within the FLR approach, but it does not rule out the occurrence of large-scale phase adjustments. However, because of the smallness of the contributions originating from the large-scale adjustments of phase to energy, the formation of well defined phase coherent domains appears unlikely to be put into evidence by numerical simulation, whose results are presumably also affected by finite size effects. This explains the puzzling results found by Abe:[6] it would be quite unrealistic to extract a characteristic length (to be identified with L_0) from the extremely broad distribution of the domain size obtained by numerical simulation. To a large extent, this also applies to the broad distribution for the threshold field $E_{\rm th}$ found by Abe; the latter was extracted in Ref. 6 by employing an indirect method, examining the dependence of the lowest energy state found numerically on the average phase, to which $E_{\rm th}$ is canonically conjugated (see also below).

To find the phase configuration that minimizes Eq. (1) by numerical simulation, Matsukawa[7] replaced the continuous function $\varphi(\mathbf{r})$ by a discrete set $\{\varphi_\lambda\}_{\lambda \le N_L}$, introducing a cubic lattice with N_L sites among which N_i impurities are randomly distributed with the probability (concentration) c. As previously pointed out,[8] the introduction of the artificial lattice prevents phase variations on a length shorter than the artificial lattice constant, $d = (c/n_i)^{1/3}$, or alternatively, Fourier components with wave vectors larger than $2\pi/d$, to be identified with the present cutoff parameter k_c. Making use of Eqs. (12) and (13), the following expression of the parameter $r \equiv -w_{\rm imp}/w_{\rm el}$ studied by Matsukawa is obtained: $r = (2 + 0.0443\, c^{1/3}\varepsilon^2)/(1 + 0.0368\, c^{1/3}\varepsilon^2)$. For the samples with $\varepsilon = 1$ and $c = 1$ studied by Matsukawa — corresponding to a domain size $n_i^{1/3} L_0 \simeq 46.7$ — this formula yields a value $r = 1.97$, in good agreement with the value $r_{\rm NS} \simeq 2.05$ found numerically. For $\varepsilon = 2$ ($n_i^{1/3} L_0 \simeq 11.7$), a value $r_{\rm NS} \simeq 1.95$ was obtained in Ref. 7 by taking the average over seven samples with $c = 0.5$ and one sample with $c = 1$. It is again very close to the value $r = 1.91$ deduced from the present analytical formula. For $\varepsilon = 3$, a small domain size $n_i^{1/3} L_0 \simeq 5.2$ is obtained, indicating that the present approach marginally applies, because the statistical fluctuations come into play. This is reflected in the difference between the values found analytically and by numerical simulation, $r = 1.80$ and $r_{\rm NS} = 2.0$, respectively. Besides the parameter r, which saturates for small ε at the value 2 and not at 4/3, expected if only large-scale phase variations were present, it would be interesting to study the average phase variation, for which we expect a dependence $n_i^{-1/3} \overline{|\nabla\varphi|^2}^{1/2} \simeq n_i^{-1/3} \overline{|\nabla\theta|^2}^{1/2} \propto \varepsilon c^{-1/6}$. While the c dependence could be observed with greater difficulty, because of the small exponent of c, the linear dependence on ε of this quantity, giving a more direct confirmation of the role played by short-scale phase adjustments, is expected to be easily observed. Unfortunately, this prediction of the present analytical approach cannot be compared with numerical simulation, because results on the average phase variation among adjacent impurity sites were not reported in Ref. 7.

Agreement with numerical simulation results for the quantities mentioned above was also found in Ref. 8. More interesting is therefore that, unlike the approach of Ref. 8, the present one can also explain [cf. Eq. (14)] why the threshold field for the onset of collective conduction — extracted in Ref. 7 (unlike in Ref. 6) by directly computing the dc current — is proportional to ε^4. The inspection of Eqs. (12), (13), (14) reveals that, contrary to the usual claim,[3,15] the total energy gain, due to the CDW pinning, is not related to the threshold field. To initiate the CDW sliding, the latter has to supply only an energy of the order of the energy gain $|w_\psi|$ resulting from the adjustment of the average phase conforming to the fluctuations of the impurity potential in Fukuyama-Lee domains. An important consequence of this fact is that, not very far above the threshold, the depinned CDW has a nonuniform phase. The sliding of a nonuniform CDW,

dragging short-scale phase deformations — a result derived here within an analytical treatment — should have important implications for understanding the behavior found in experiments on CDW materials for $E \gtrsim E_{\rm th}$. Owing to the fact that the sliding CDW cannot relax instantaneously at experimental scale, combined with the spatial inhomogeneity inherently present in real samples, a nonuniform CDW phase is very likely associated to a variety of phenomena (hysteretical behavior, metastability) reported in many CDW materials in the regime near threshold.[1] Obviously, for samples consisting of many domains, this mechanism fully supports their bulk generation. Because of the large energy involved [$|w_\theta| \gg |w_\psi|$, cf. Eqs. (12) and (10)], the sliding of a uniform CDW can be expected only far above the threshold ($E \gg E_{\rm th}$). The threshold field was obtained in Refs. 6 and 8 from the point where the local minimum of the total energy as a function of the average phase (computed for the whole sample) disappears. In this case, as shown in Ref. 8, the field has to supply an energy of the order of the total energy gain obtained by deforming the phase, $|w| \simeq |w_\theta| \propto \varepsilon^2$. This is why the result $E_{\rm th} \propto |w| \propto \varepsilon^2$ obtained in Ref. 8 was found in accord to that of Ref. 6, but at variance to that of Ref. 7.

On the other side, the good agreement with all the findings of numerical simulations enumerated above, along with the experimental findings on narrow band noise — usually displaying a strong fundamental frequency peak and higher harmonic peaks of diminished amplitude,[1] associated to Eq. (10) and the terms neglected there[13] — could be taken as justifying the method employed to compute the contribution to energy of the large-scale phase adjustments by truncating the corresponding Fourier series [Eq. (10)]. This agreement could also suggest that the coupling between phase coherent domains (ignored in the present analytical treatment) do not qualitatively alter the static CDW properties.

One should remark that the randomness of impurity distribution is essential in determining this physical behavior. A long time ago and in a different context, a model similar to the present one was investigated, in which the elastic term in Eq. (1) has the form $W_{\rm el} = (\kappa/2) \sum_{(j,l)} (\varphi_j - \varphi_l)^2$, the summation being over the nearest neighboring sites, $\varphi_j = \varphi(\mathbf{R}_j)$, and the centers responsible for the phase distortions are supposed to occupy the sites of a lattice .[20] The various quantities computed below for this case will be specified by the index "reg." By taking the continuum limit, justified in the weak coupling limit, the solution of that model can be obtained by minimizing with respect to k the following expression — which is exact in thermodynamical limit — of $\omega_{\rm reg} \equiv w_{\rm reg}/(n_i Q^2 a^2)$:[20]

$$\omega_{\rm reg} = \tilde{\epsilon}\left(1 - \frac{2}{k^2}\right) + \frac{1}{2} - \pi\frac{\sqrt{\tilde{\epsilon}}}{k}\frac{1}{K(k)} + 4\frac{\tilde{\epsilon}}{k^2}\frac{E(k)}{K(k)},$$

where a is the lattice constant, $\tilde{\epsilon} \equiv E_i/(\kappa Q^2 a^2)$, K and E are the complete elliptic integrals of the first and second kind.[21] The phase at site j is expressed by $\varphi_j = \Phi_0 + {\rm am}(j\sqrt{\tilde{\epsilon}}/(kQa); k) - Qaj$, where ϕ_0 is an *arbitrary* constant and am is the elliptic function amplitude.[21] The minimization of $\omega_{\rm reg}$, with respect to k, yields an equation determining k as a function of $\tilde{\epsilon}$, but because ϕ_0 is arbitrary, all phase φ_j's remain undetermined (nonpinned). In the weak coupling limit ($\tilde{\epsilon} \ll 1$), the following leading contributions can be obtained: $\omega_{\rm reg} = -\tilde{\epsilon}^2/4$, $r_{\rm reg} \equiv -W_{\rm imp}^{\rm reg}/W_{\rm el}^{\rm reg} = 2$, $\overline{|\nabla\varphi_{\rm reg}|^2}^{1/2} = Q\tilde{\epsilon}/\sqrt{2}$; that is, results similar to those expressed by Eqs. (12) for the case of random centers. They demonstrate, once more, that the leading contribution to energy, phase gradient and the parameter r is brought about by the local adjustments of phase and not by slow phase variations. Because of the *regular* distribution assumed in deriving these expressions, there is no mechanism to stabilize large-scale variations of phase, which remains, therefore, nonpinned; this contrasts to the case of random impurities, where the *fluctuations* of impurity potential can sustain large-scale phase variations, that are responsible for the existence of optimum values of the average phase in extended domains.

A series of experiments was recently carried out in order to understand the nature of impurity pinning in doped niobium triselenide.[9–11] This material is of special interest because the band structure resulting from hybridized electronic orbitals of Nb and Se atoms determines the formation of two independent CDW's.[1] The detailed analysis of the threshold field data[9] led to a quadratic concentration dependence $E_{\rm th} \propto x^2$ for the two CDW's, in both $Ta_xNb_{1-x}Se_3$ and $Ti_xNb_{1-x}Se_3$ samples, in accord to Eq. (14). A subsequent high-resolution x-ray study[10] on Ta-doped $NbSe_3$ found that the real-space correlation function obtained from the measured scattering intensities decays exponentially with large characteristic lengths in all directions, in agreement with the theoretical result [Eq. (5)]. These lengths could be identified with those of the Fukuyama-Lee domains: they are much greater than the impurity spacing and scale with the inverse of the Ta concentration and with temperature as the square of the CDW gap. The largest ε value for both Ta- and Ti-doped samples, for the two CDWs, deduced by fitting the experimental data[9] is $\varepsilon \approx 1$, so that it falls in the range where the present results apply.

Despite the fact that this behavior found experimentally agrees with the Lee-Rice prediction for $E_{\rm th}$ and L_0,[3] the phase adjustment found here is more complex than described by Lee and Rice. Lee and Rice[3] studied the deformation of the CDW order parameter around a single impurity and evaluated both the energy gain from impurity potential and elastic energy cost accompanying this deformation. By implicitly assuming a large phase adjustment ($\sim \pi$) at the impurity site, they correctly observed that, for weak coupling (corresponding to $\varepsilon n_i^{-1/3} k_c \ll 1$), this process involves an elastic energy that exceeds the accompanying energy gain, rendering thereby the strong pinning impossible. Based on this reason, they claimed that "the phase assumes its value at infinity everywhere" (cf. Sec. II of Ref. 3) and considered only slow coherent variations of phase over large regions; this claim turns out to be in conflict to the present

analysis, revealing that short- and large-scale phase adjustments coexist.

A recent study of the high-resolution x-ray scattering intensities in $NbSe_3$ samples lightly doped with tantalum[11] demonstrated that two lengths — identified with the Fukuyama-Lee domain size and the amplitude correlation length — are necessary to analyze the x-ray line shape. The large-scale phase variations over Fukuyama-Lee domains are not the only ones occurring in the system. More rapid phase variations also exist; only phase fluctuations within a length that could be identified with the amplitude correlation length are suppressed. This experimental finding is fully consistent to the picture emerging from the theoretical analysis presented in Sec. II. Apart from the cutoff $\sim k_c^{-1}$ ($\sim \xi$) imposed within the framework of the phase-only theory, the spatial variation of θ has *no* characteristic length.

The nature of CDW pinning in $NbSe_3$ doped with both tantalum and titanum was a subject of controversy.[22,23] Briefly, the theoretical estimations[23] showed that the energy gained by short-scale phase adjustments is substantially larger than that gained by large-scale phase adjustments within FLR domains, whereas the experimental threshold field data scale with the square of impurity concentration.[9] The former was interpreted by Tucker[23] as evidence for strong pinning, while the latter was taken by Thorne and McCarten[22] as a confirmation of the Lee-Rice mechanism of weak pinning. The present results could clarify this controversy. As shown here, even in the weak-coupling limit, the largest contribution to energy comes indeed from short-scale phase adjustments ($|w_\theta|/|w_\psi| \approx k_c L_0 \gg 1$). However, in the same limit, the threshold field obeys Eq. (14) (i.e., of Lee-Rice type); $E_{\rm th}$ needs not supply an energy equal to the difference between the energy of the pinned and uniform CDW states ($\approx |w_\theta|$), but a much smaller amount, of the order of the energy gain ($|w_\psi|$), obtained by optimizing the average phase.

Further support for the presently proposed interpretation comes from the recently observed $+\mathbf{Q}/-\mathbf{Q}$ asymmetry of the diffuse satellite lines in the x-ray patterns ("white-line" effect).[16] As discussed elsewhere,[16,8] the contribution to the scattering intensity of the white-line effect is proportional to the quantity $\langle\cos(\mathbf{Q}\mathbf{R}_j+\theta_j)\rangle_i$ (denoted by y^{-1} in Sec. VII of Ref. 8), whose value plays, therefore, a key role in its observability. If *only* large-scale phase variations were present, $\langle\cos(\mathbf{Q}\mathbf{R}_j+\theta_j)\rangle_i = N_0^{-1/2}$ would necessarily be a small quantity and the white-line effect could not be observed ($N_0 = n_i L_0^3 \gg 1$ — number of impurities per phase coherent domain). For small ε, Eqs. (12) and (13) yield a considerably larger value $\langle\cos(\mathbf{Q}\mathbf{R}_j+\theta_j)\rangle_i = 0.031 k_c n_i^{-1/3}\varepsilon \approx k_c n_i^{-1/3} N_0^{-1/6}$, such that the white-line effect could be observed even for small ε. Although no microscopic estimations of the parameter ε were reported, it is likely that small values of this parameter characterize (at least) some of materials from the large class of the quasi-one-dimensional CDW materials, where the white-line effect was observed.[16] Therefore, in accord to the previous suggestions,[16,8] this effect should be interpreted as giving evidence for the occurrence of local deformations of CDW phase correlated with impurity positions. However, the observability of this effect does not necessarily rule out the occurrence of large-scale phase adjustments.

At the present phenomenological level, no notable difference exists between a charge and a spin-density wave (SDW). Therefore, the present analysis can also be extended without difficulty to the latter. Subtracting the possible effect of residual defects, a dependence of the threshold field for SDW depinning proportional to the square of the concentration of Sb substituting As atoms in the Bechgaard salt $(TMTSF)_2 As F_6$ was found experimentally,[24] in accord to Eq. (14), expected to apply because of the weakness[25] of the SDW-impurity coupling. Unfortunately, a direct experimental evidence of local adjustments of the SDW phase could hardly be obtained.

In conclusion, the present analytical study has demonstrated that both short- and large-scale adjustments of the phase exist in a CDW weakly coupled to impurities and are important to understand the statical CDW behavior revealed in experiments performed on CDW systems as well as the results — usually less transparent physically — obtained by numerical simulations. A quantitative description of the CDW dynamics requires the inclusion of the effects, neglected here, of the coupling among phase coherent domains as well as the contribution of metastable states. It would be interesting to investigate to what extent the present ideas could be extended to other related phenomena, e.g., the pinning of the vortex structure of type-II superconductors.[26]

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