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Collective transport of charges in charge density wave systems based on traveling soliton lattices

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Solitons are peculiar excitations that appear in a wide range of nonlinear systems such as in fluids or optics. We show here that the collective transport of charges observed in charge density wave (CDW) systems can be explained by using a similar theory based on a traveling soliton lattice. A coherent x-ray diffraction experiment performed in the sliding state of a CDW material reveals peculiar diffraction patterns in good agreement with this assumption. Therefore, the collective transport of charges in CDW systems may be due to a nonlinear interaction leading to a self-localized excitation, carrying charges without deformation through the sample, on top of the CDW ground state. This single theory explains why charges remain spatially correlated over very long distances and reconciles the main features of sliding CDW systems observed by transport measurements and diffraction. This approach highlights a new type of charge transport in CDW systems and opens perspectives in controlling correlated charges without dispersion over macroscopic distances.

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A soliton can take the form of a localized solitary wave which propagates in a medium while keeping a constant shape through nonlinear interactions. Once created, this localized wave, with particle-like properties, propagates without dispersion and with a remarkably long lifetime. Solitons are present in many systems such as fluids [1] and optical fibers [2] and also in more unexpected fields like traffic jams [3] and blood pressure [4]. Their involvement in electronic crystals is invoked in only a few systems, such as Josephson junctions [5] and conducting polymers [6], manifesting themselves in different forms.

Charge density wave (CDW) systems are another type of electronic crystal made of spatially correlated electrons. CDWs can coexist with spin density waves like in chromium [7,8] or compete with superconductivity like in cuprates [9]. Although the static CDW state is now well understood, the dynamical one is still debated. Indeed, the most spectacular property of a CDW system is its ability to carry correlated charges when submitting the sample to an external electric field. Above a threshold field E_{th}, a non-ohmic resistivity is observed, including voltage oscillations with a fundamental frequency f_0 proportional to the applied field, as well as several harmonics in the frequency spectrum [10,11]. Up to 23 harmonics have been observed in $NbSe_3$ [12]. This existence of collective transport through CDW compounds has received considerable interest for more than 35 years. However, the understanding of the type of charge carriers and their propagation mode remains incomplete.

Many theoretical approaches have been proposed to describe this phenomenon. The simplest one is based on translation invariance of the incommensurate CDW [13]: the whole sinusoidal density of condensed charges *slides* over the atomic lattice with a constant velocity. Although appealing, this explanation remains probably approximate since the CDW is described as an almost sinusoidal modulation from diffraction experiments [14] while transport measurements reveal a strong anharmonic signal [12]. A more realistic description of CDW dynamics assumes a slowly varying phase $\phi(x)$ of the CDW interacting weakly with impurities [15]. On the contrary, theories considering strong electron-phonon coupling neglect the role of impurities and treat CDW dynamics as only due to phonons [16]. The most accepted theory, developed by Ong and Maki [17] and Gor'kov [18–20], deals with the CDW-metal junction at electrical contacts. The conversion of normal electrons from the metallic electrode into condensed charges in the CDW is made possible by climbing CDW dislocations at the interface. These so-called *phase slippage* and current conversion phenomena are in agreement with local resistivity measurements close to contacts [21].

This conversion phenomenon is also accompanied by an elastic deformation of the CDW. Without an external field, the CDW $\rho = \rho_0 \cos[2k_F x + \phi(x,t)]$ is homogeneous along the sample and pinned at both ends at the two metal/CDW junctions. In the sliding state, however, the CDW is compressed at one electrode and stretched at the other as revealed by diffraction experiments [22,23]. A simple static elastic theory shows that the time-averaged phase ϕ of the elastic object has to obey a quadratic behavior: $\langle \frac{\partial^2\phi}{\partial x^2} \rangle_t =$ const. [24], leading to a parabolic phase between the two contacts, in agreement with the experiment.

The impressive number of studies focusing on sliding CDWs deserves, however, a few comments. While many studies have been devoted to the conversion process close to electrodes, the charge carrier propagation through macroscopic samples remains a subject that has been little studied to date. A pure quantum tunneling through the sample has been mentioned [25] and phase slippage mixed with quantum tunneling has also been considered at low temperature [26]. However, current oscillations are clearly observed in very long samples, up to several centimeters for $NbSe_3$ and a pure quantum tunneling over such large distances is probably unlikely. Note also that in the phase slippage theories [17], impurities play a minor role, hidden in the tunneling coefficient. The authors justify this absence by the increase of CDW correlation lengths ξ_l in the sliding regime. Nevertheless, recent diffraction experiments show that ξ_l is always shorter in the sliding state than in the pristine one [27], suggesting on the contrary that defects may still play a role in the sliding state.

Several ascertainments can also be made with respect to diffraction experiments. First, CDW systems can stabilize

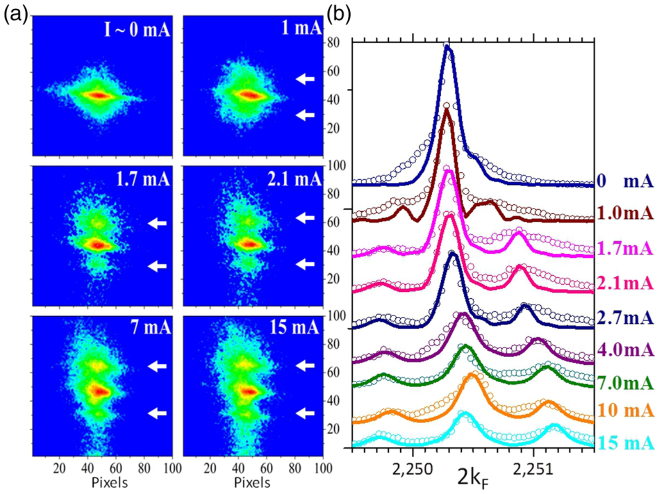

FIG. 1. Coherent diffraction patterns of the $2k_F$ satellite reflection associated with the CDW vs applied currents. (a) The $Q=(1,q_{2k_F},0.5)$ with $q_{2k_F}\approx 0.748\,b^*$ satellite reflection in the blue bronze $K_{0.3}MoO_3$ for several external currents ($T=70$ K). The two-dimensional patterns correspond to a sum over several θ angles through the maximum of intensity. The b^* chain axis is vertical and the additional satellites are indicated by white arrows. The scale is in pixels (logarithmic color scale from blue for lowest intensity to red for highest intensity). (b) Corresponding profiles along b^* after summation along the perpendicular axis (opened circles) and fits using the soliton lattice model (continuous lines). A convolution with a Lorentzian shape has been used to take into account the finite Q resolution as well as the projection along the chain axis b^*, the integration over several θ angles, and temporal fluctuations during data acquisition.

CDW dislocations, i.e., abrupt phase shifts of the CDW modulation, first observed by coherent x-ray diffraction [28]. Second, those CDW dislocations are mobile in the sliding regime, as proved by the disappearance of speckles [27], suggesting that moving phase shifts can play a role in the charge transport.

The starting point of this study is the reinterpretation of experimental data we have obtained in the archetype $K_{0.3}MoO_3$ blue bronze system under applied currents [29]. The most spectacular point of this experiment is the appearance of two secondary satellites located on both sides of the $2k_F$ satellite reflections associated with the CDW (see Fig. 1). The corresponding spatial frequencies leading to micron-size distances have been observed thanks to coherence properties of the x-ray beam [30]. In [31], we used a phenomenological static model to account for this observation. We develop here a fundamentally different model based on the existence of a moving 2π-soliton lattice that not only accounts for coherent diffraction experiments but also describes all the main features of sliding CDW observed by transport measurements and diffraction.

An interaction that couples impurity potential and the phase ϕ of the CDW is considered as in [32]. The corresponding phase-dependent Hamiltonian leads to the following equation of motion:

$$\frac{\partial^2\phi}{\partial t^2}-c_\phi^2\frac{\partial^2\phi}{\partial x^2}+\eta\frac{\partial\phi}{\partial t}+\omega_0^2\sin(\phi)=F, \qquad (1)$$

where $F=\frac{2c_\phi^2\,e\,E}{\hbar\,v_F}$ is proportional to the applied force, $c_\phi=\sqrt{m/m^*}\,v_F$ is the phason velocity, ω_0 the pinning frequency, m the free electron mass, m^* the electron band mass, and v_F the Fermi velocity. We also add an effective damping term $\eta\,\frac{\partial\phi}{\partial t}$ to mainly take into account the coupling between CDW and phonons. Contrary to [32], the $\sin(\phi)$ term is here not linearized, thus allowing abrupt phase variations. The usual nonperturbed sine-Gordon equation (for which $F=\eta=0$) is known to admit soliton solutions. However, soliton excitations are quite robust and survive the inclusion of a reasonable external force and dissipation keeping their topological properties, although the soliton shape is slightly modified [33].

Let us now solve Eq. (1) considering that the phase $\phi(x,t)$ contains two terms: a slowly varying phase $\phi_0(x)$ and a dynamical part $\phi_1(x,t)$ where ϕ_1 varies much more rapidly than the static one ($\langle|d^2\phi_1/dx^2|\rangle_t \gg \langle|d^2\phi_0/dx^2|\rangle_t$). The static part $\phi_0(x)$ can be calculated by averaging Eq. (1) in time:

$$\left\langle\frac{\partial^2\phi_0(x)}{\partial x^2}\right\rangle_t=\left(\frac{\eta\,\pi}{e}j-F\right)\Big/c_\phi^2, \qquad (2)$$

where the excess of current in the sliding mode $j=\frac{e}{\pi}\frac{\partial\phi}{\partial t}=\frac{e}{\pi}v_s$ is constant far from electrodes as observed by several transport measurements [34–38]. This leads to a quadratic variation of the phase $\phi_0(x)$ in perfect agreement with diffraction experiments [22,23]. The complete Eq. (1) now reads

$$\frac{\partial^2\phi_1}{\partial t^2}-c_\phi^2\frac{\partial^2\phi_1}{\partial x^2}+\omega_0^2\sin(\phi_1)=F+c_\phi^2\frac{\partial^2\phi_0}{\partial x^2}-\eta\frac{\partial\phi_1}{\partial t}. \qquad (3)$$

The dynamical part $\phi_1(x,t)$ obeys the sine-Gordon equation and is submitted to an effective force including the friction. Considering the periodic nucleation of CDW dislocations at the electrode [26], we obtain a train of solitons plus a negligible quantity δ [33]:

$$\phi_1(x,t)=\delta+\sum_{n=-\infty}^{\infty}4\arctan\left[\exp\left(\frac{x-v_st-ln}{l_S\,\gamma(v)}\right)\right], \qquad (4)$$

where l is the distance between successive solitons and $l_S=c_\phi/\omega_0$ their extension. Overlapping effects between solitons are neglected ($l/l_S>2$).

The soliton lattice model presented here leads to a singular diffraction pattern in good agreement with the experiment, especially for larger currents when the soliton lattice is well formed (see Fig. 1). Two additional satellite reflections appear on both sides of the main $2k_F$ peak located at $\delta q=\pm 2\pi/l$. Since Eq. (4) is not an even function, the two satellites at δq do not have the same intensity in agreement with the experiment. The soliton extension l_S mainly affects the intensity ratio between the central peak and the two satellites. Note that the central peak is not necessarily located at $2k_F$ but may be shifted with respect to l/l_S [39]. Note also that the soliton model used here is global in the sense that one soliton cannot be considered individually without considering the complete soliton lattice. The three main fitting parameters (l, l_S, and $2k_F$) are extremely sensitive to each other in this nonlinear model and the solution space is particularly narrow.

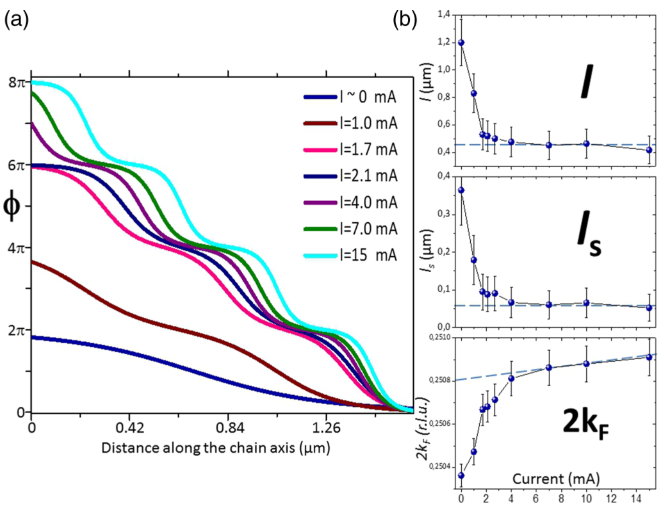

FIG. 2. (a) Profile of the CDW phase $\phi(x)$ obtained from the fits of Fig. 1(b) vs the distance along the chain axis for different external currents. (b) Distance l between solitons, soliton size l_S, and $2k_F$ vs currents.

The traveling soliton lattice correctly accounts for the diffraction patterns [see Fig. 1(a)]. The corresponding phase $\phi(x)$ and the behavior of l, l_S, and $2k_F$ versus the external current are shown in Fig. 2. The distance between solitons l reaches the micron size for small currents, decreases for $I < 2I_S$, and reaches a stationary value above $2I_S$ where I_S is the threshold current. The soliton extension l_S follows a similar behavior versus current. The distance l between solitons is always greater than the soliton extension l_S ($l/l_S > 3$) which justifies the assumption of noninteracting solitons.

A finite threshold field E_{th} appears in this nonlinear framework [for which $\frac{\partial^2\phi_1}{\partial t^2} = \frac{\partial\phi_1}{\partial t} = 0$ in Eq. (1)] in agreement with the experiment. For $E/E_{\text{th}} \gg 1$, the soliton reaches a stationary sliding velocity [33]

$$v_s = \frac{\pi e \tau^* v_F k_F}{2\omega_0 m^*}\sqrt{\frac{m}{m^*}}E, \qquad (5)$$

proportional to the applied field E, in agreement with the experiment. In this framework, a quantitative sliding velocity v_S can be given. Since the observed l saturates for large enough fields [see Fig. 2(b)] and the fundamental frequency ranged from $f_0 = 1$ to 100 MHz [40], $v_s = f_0 l$ ranges from $v_s = 0.5$ to 50 m/s.

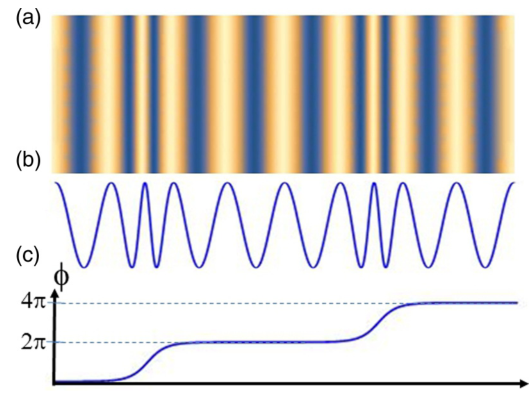

FIG. 3. (a) Soliton lattice in top the CDW modulation for $l/l_S = 20$. The lighter wave fronts correspond to an excess of electrons. (b) Corresponding electronic density profile and (c) corresponding phase ϕ_1. See movies showing the moving soliton lattice in the Supplemental Material [41].

Remarkably, the most impressive feature of a sliding CDW is the presence of many harmonics in the frequency spectrum as observed by transport measurements [12]. This signature is quite naturally explained by the abrupt shape of solitons when $l_S \ll l$ (see Fig. 2). Note that in the opposite case, when $l_S \sim l$, the electronic density is close to a harmonic modulation and the sliding process is more similar to a global CDW translation as in the Fröhlich approach [13].

In this semiclassical description, charges are carried by moving phase shifts of the CDW modulation (see Fig. 3 and Supplemental Material [41]) which can travel through macroscopic samples without deformation on top of the CDW ground state. The traveling 2π soliton lattice which is superimposed to a slowly varying static phase reconciles seemingly contradictory results: on the one hand, it explains results obtained by diffraction including the macroscopic elastic deformation along the sample and the singular coherent diffraction patterns in Fig. 1. On the other hand, this single theory also explains the main features observed in transport measurements such as the existence of a threshold field, the presence of correlated charges despite the large distances involved in this phenomenon, and finally the presence of several harmonics in the frequency spectrum. The model presented here highlights a type of charge transport based on traveling CDW phase shifts and opens new perspectives in controlling correlated charges over macroscopic distances.

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[41] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.94.201120 for movie of the traveling soliton lattice on top of the CDW state in 2D (movie 1) and in 1D (movie 2): on top, the moving phase $\Phi(x)$ corresponding to the traveling 2π soliton lattice and the corresponding CDW profile below.