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A possible mechanism for photoinduced structural phase transitions in low-dimensional electron–lattice systems

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Abstract

We study low-dimensional electron–lattice (el–l) models in order to know fundamental features of photoinduced structural phase transitions (PSPT). To avoid entering into unnecessarily specific details, the model is chosen to be as simple as possible, namely, includes only nearest-neighbor hopping of electrons and a site-diagonal el–l coupling. Our special concern is in the geometrical and dimensional effects and hence single- and coupled-chain systems as well as a two-dimensional system are investigated. We prepare two states, that are, stable and metastable charge–density–wave (CDW) ground states and discuss metastable-to-stable and stable-to-metastable transitions. For both the cases, we particularly focus on two important features. One is the nonlinearity in the converted fraction as a function of the light intensity, while the other is the aggregation of excitations. Performing adiabatic-potential-surface analyses and dynamical calculations, we demonstrate that our model changes its nature drastically when the geometry and the dimensionality are altered, and then manifests both the features simultaneously with suitably adjusted conditions.

Keywords: Photoinduced structural phase transition; Electron–lattice interaction; Charge–density-wave (CDW)

1. Introduction

The phenomena of photoinduced structural phase transitions (PSPTs) are attracting more and more attentions in recent years [1–7]. The number of examples is increasing year by year, ranging over conjugate polymers, neutral-ionic-transition systems, manganese oxides, and so on. However, it is still a situation that only the existence of each PSPT has been just identified. Although the experimental quests for detailed mechanisms have been started very recently, we have no decisive result yet.

In such a situation, we think that it will be meaningful to investigate the mechanism from a theoretical point of view. Although studies related to specific cases will be important, we judge that the clues are still too few to develop them. Thus we do not require a strong connection with the real materials, but focus on a charge–density–wave (CDW) model with a half-filled electronic band, as an example. In particular, our CDW model is very simple, having only the electron itineracy and the electron-lattice (el–l) coupling. We therefore believe that it will be one of the best models for the investigation of PSPT from a general point of view. Although the PSPTs in three-dimensional systems are also discovered, we here confine ourselves mainly in one-dimensional (1-D) cases. This comes from a theoretical advantage, that is, a domains wall (DW) separating phases is regarded as a point in 1-D cases. Hence the dynamics, at least, that after an early time during which a DW picture is not applicable, can be described using only the coordinates of gravitational centers of DWs. Of course, we must discuss the electronic excitations for PSPT. In the framework of DW concepts, they are interpreted as the internal structures of DWs. Namely, the DWs in the ground state and those in the excited ones are different. This is the aspect completely different from the conventional theories based on the traditional Ginzburg–Landau equations. Moreover,

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we extend 1-D cases to coupled-chain systems. This adds a useful variety to the internal structure of a DW, and also makes it possible to discuss generally the nonlinearity seen in the converted fraction as a function of the light intensity, and the aggregation of excitations, with the help of additional knowledge in a corresponding 2-D case.

2. Model

We utilize the following model with a site-diagonal el–l coupling:

$$H = -\sum_{(l,l')\sigma} t_0(C^{\dagger}_{l\sigma}C_{l'\sigma} + \text{h.c.}) + \frac{\Delta}{2}\sum_l (1-(-1)^l)n_l - \alpha\sum_l Q_l(n_l-1) + \frac{m}{2}\sum_l \dot{Q}_l^2 + \frac{K}{2}\sum_l Q_l^2, \quad (1)$$

where $C^{\dagger}_{l\sigma}$ and $C_{l\sigma}$ are creation and annihilation operators, respectively, of an electron with σ spin at the lth site in a system of general dimension, and Q_l is the lth lattice displacement with mass m. Note that the factor $(-1)^l$ means $(-1)^{l_x}(-1)^{l_y}$ with $l=(l_x,l_y)$, in the cases with two directions. The first term means the nearest-neighbor hopping with transfer energy t_0. Without the second and fourth terms, the model with a half-filled band describes static CDW states, which are symbolically represented as …0202… and …2020… in electron densities along a spatial direction [8]. Here the site energy Δ in the second term lifts up the degeneracy of these two-fold ground states and then yields stable and metastable phases. A DW hence separates these phases along the chain-parallel direction. In the following, we use a dimensionless displacement q_l and the el–l coupling energy $S \equiv \alpha^2/K$, instead of Q_l and α, respectively. In the case of coupled-chain systems, periodic and open boundary conditions (BC) are imposed for chain-perpendicular and parallel directions, respectively. Thus our system can be considered as a cylinder with a circumference M that is nothing but the number of chains. Gradually increasing M from zero (a single chain) to infinity (2-D cases), we can investigate continuous changes in the fundamental properties related to PSPT. To treat this model, we use an adiabatic approximation for the lattice, focusing on classical aspects of the lattice motions. The electronic degrees of freedom are, on the other hand, calculated full quantum-mechanically, using time-dependent Schrödinger equations.

3. Metastable-to-stable transitions

Here we discuss the cases with intermediate el–l couplings, because efficient PSPTs are rather difficult in strong-coupling cases and, while, they are too easy and almost trivial in weak ones. Fixing the S/t_0 ratio so as to give a CDW gap around 1.0 (here and hereafter all the energies are scaled by t_0), we observe the following general tendencies [9]. Firstly, in a single-chain system, only one photon is enough to drive the system into a stable phase. This comes from the basic property that all the excited states have no energy barrier for the transition, even in the presence of a finite barrier in the ground state. While, in a 2-D system, the same type of barriers are so dominant, namely, continue to exist in higher excitations, for example, up to as many as ten-electron excitations. Then we expect that a moderate amount of energy barriers, that is, the nonlinearity, will be obtained in coupled-chain systems. In fact, we find that the nonlinearity is continuously enhanced in increasing M. In Fig. 1, we show a case with $M=4$, in which the energy barrier survives up to three electron excitations, while it disappears with four and more than four electron excitations. Since an attractive force due to a multi-bond effect is also confirmed among these four electron excitations, this result means that both the conditions, namely, those for the nonlinearity and the aggregation, are satisfied simultaneously. Moreover, our dynamical calculations also support this scenario [9]. It is already confirmed that two excitations merge into a larger unit, and a pair of larger units also merge to trigger a PSPT. This mechanism is quite new compared with the conventional theory of nucleation, in the sense that the electron excitation by the light is a key factor.

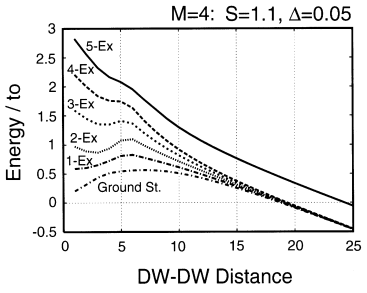

Fig. 1. The adiabatic potential curves as functions of the DW–DW distance in a four-chain system. The background and domain phases are metastable and stable, respectively. The lowest curve is for the ground state, and the other ones for the excited states. The abbreviation 'n-Ex' means the *lowest* n-electron excitation state (this is used here and in Fig. 2). The parameters are $S=1.1$ and $\Delta=0.05$.

4. Stable-to-metastable transitions

These situations are subtler than those in the previous section, and our study is still in progress. Thus we present our latest result that is still preliminary, but expected to have an important meaning. Fig. 2 also shows the result

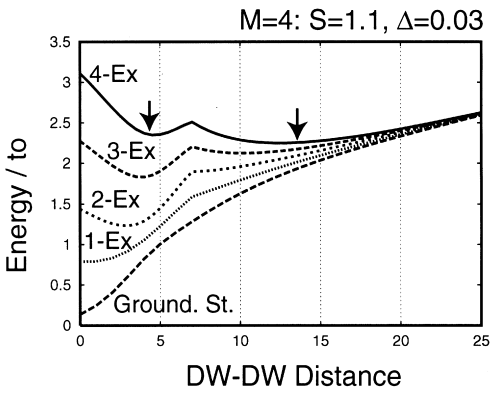

Fig. 2. Same as Fig. 1, but in the background of the stable phase. The parameters are $S = 1.1$ and $\Delta = 0.03$. The two vertical arrows specify the sizes of possible metastable domains.

in a four-chain ($M = 4$) system, but here the metastable domain is formed in the background of the stable phase. As is seen clearly in the shapes of potential surfaces, only the formation of a local domain is possible up to three-electron excitations. It is natural because such a small domain is confined in a width of coupled chains. While, in the surface of four-electron excitations, a new structure, of which domain size is as large as 13 sites, is found in addition to the local domain. From the lattice configuration, we see that this structure is extended in the chain-parallel direction, and hence can be regarded as a DW-domain-DW structure. As for the aggregation of excitations, almost the same behavior is expected also in this case, because the excitations do not identify in which states they are, when they are still local. We have also performed dynamical calculations with a very small local deformation with four electron excitations, as the initial state. Then the formation of the aforementioned extended domain is confirmed within ten lattice periods.

Lastly, we mention the stabilities. Due to its characteristic structure, the DW-domain-DW is stable against the radiative decays, because the associated electron transitions occur between the levels localized at each DW. In fact, we see that the decay probability decreases exponentially with the DW–DW distance. As for the non-radiative decay, on the other hand, its probability is sensitive to the energy difference between the levels associated with the electronic transitions. Unfortunately, these energy differences converge to zero in increasing the DW–DW distance in our model. There is therefore a tendency that the probability is first enhanced with the increase in the DW–DW distance and then decreases due to the reduction of the matrix element. In that sense we might need another suitable model in which energy levels do not become degenerate in the limit of a large DW–DW distance. Nevertheless, even in the present model, the absolute magnitude of the decay probability is not substantial when the lattice frequency is small. We estimate by the dynamical calculations that the life time of the extended domain is about 7 ns for $\hbar\omega = 0.01$.

5. Summary

We have seen what type of transitions are expected in the systems with various geometrical conditions. Even when we confine ourselves in the intermediate el–l couplings, the transitions drastically change their nature. Common to both the two cases (metastable to stable and vice versa), the aggregation of excitations is possible in the coupled-chain systems, because of the multi-bond effect. Moreover, the nonlinearity also manifests itself in a more exaggerated way in the same systems with larger circumferences. As a conclusion, a feasible scenario for PSPT is proposed using a four-chain system, where four excitations are expected to aggregate into a seed for the transitions.

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