光誘起構造相転移の理論

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1. Introduction

In recent years, there discovered many new insulating solids, which, being shined by only a few visible photons, become pregnant with a macroscopic excited domain that has new structural and electronic orders quite different from the starting ground state. This phenomenon is called "photoinduced structural phase transition"(PSPT).[1] According to recent development of laser spectroscopy, quite novel and noticeable properties of this PSPT are now discovered in various insulating solids.[1,2,3] 》

However, as for the real-time dynamics of these nonequilibrium PSPTs, there are still many unsolved problems. For this reason, in the present paper, we will be concerned with the real-time dynamics of domain formations, which finally results in the PSPT in a 2-D crystals. Taking a many-exciton system coupling strongly with Einstein-phonons in this 2-D insulating crystal, we numerically calculate spatio-temporal evolutions of photo-generated excitons, their proliferations and domain pattern formations, full-quantum mechanically.

As for the light excitation, we assume the case of pulse excitation composed of several successive ones with an equal time interval. We will show that there are two stages of exciton proliferation. One is the early stage, and is equal to the time region wherein the successive pulse excitation is going on. Another is the retarded stage, wherein the successive pulse excitation is turned off. In the former stage, the total number of excitons drastically increases by making use of large excess phonon energy just donated from the light pulse. While, in the retarded stage, the whole exciton system reaches some local adiabatic potential minimum. Hence the total number of excitons only gradually increases by the tunneling through various adiabatic potential barriers.

We will conclude that the spatial pattern of the exciton domain formed in the early stage, sensitively inherits the anisotropy of inter-exciton interactions. It will be also shown that this anisotropy makes the aforementioned tunneling quite efficient, and the proliferation quite successful even in the retarded stage, as compared with the cases wherein the inter-exciton interaction is isotropic.

2.Many-Exciton System Strongly Coupling with Phonons

Let us define our relevant system composed of many excitons coupling strongly with Einstein-phonons. The total Hamiltonian $(\equiv H_s)$ of our system is written as $(\eta = 1)$

$$H = H_s + H_r + H_{sr}\,, \qquad (1)$$

where H_s denotes the strongly coupled exciton-phonon system, which is given by

$$H_s = E\sum_l B_l^+ B_l + \sum_{l\neq l'} T(|l-l'|)\, B_l^+ B_{l'} + \omega\sum_l b_l^+ b_l - \sqrt{\omega S}\sum_l B_l^+ B_l\,(b_l^+ + b_l)$$

$$+ \sum_{l\neq l'} G(|l-l'|)\, B_l^+ B_l (B_{l'}^+ + B_{l'}) + \sum_{l>l'} V(|l-l'|)\, B_l^+ B_l B_{l'}^+ B_{l'}\,. \qquad (2)$$

Here E is the energy of an exciton, B_l^+ and b_l^+ are creation operators of an exciton and a phonon, respectively, at a lattice site specified by a position vector l in a 2-D square lattice. T is the

exciton transfer from site l' to l, ω is the energy of the Einstein-phonon, and S is an exciton-phonon coupling constant. G and V are the third- and the fourth-order anharmonic inter-exciton interactions, respectively, which come from a long range Coulomb interaction among electrons and holes constituting these excitons.

H_r represents a reservoir composed of the radiation field and the acoustic phonons, linearly coupling with the exciton and the Einstein phonon fields through H_{sr}. Consequently, various relaxations can occur in our relevant system, such as phonon relaxations, radiative and nonradiative decays of excitons.

In order to make our later discussions simple and clear, we focus only on two typical cases of the parameters T, V and G, that is, an anisotropic case and an isotropic one as shown in TABLE 2. All these parameters are listed in TABLE.1 and 2, and their spatial extensions are illustrated in Fig.1.

The isotropic case is the standard one since it includes the interactions only between neighboring two sites. In this model, two excitons at neighboring two sites attract each other, and it tends to make an exciton-cluster. Throughout the present paper, the occupation of a single site by more than one exciton is excluded from the beginning. From eq.(2), we can easily see that the photo-excited excitons proliferate through the third-order anharmonicity G. In the anisotropic case, on the other hand, the interactions between neighboring sites and that between next neighboring sites are assumed to be opposite in their signs.

In the present paper, we will not be concerned with their microscopic origins. We will treat them only phenomenologically, and compare aforementioned two typical cases in connection with the domain pattern formation and the proliferation. In order to perform practical calculations, we also have to derive the master equation under Markov approximation for the reservoir.

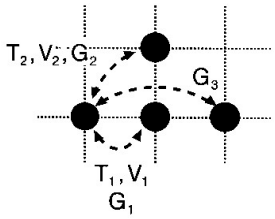

FIGURE 1. Spatial extensions of T, V and G in the 2-D square lattice. Parameter values are listed in TABLE.2.

TABLE 1. Common parameters in eq. (2)

ω	0.10 eV
E/ω	7.80
S/ω	6.45

TABLE 2. Parameters for inter-exciton interactions used in eq. (2)

	Anisotropic case	Isotropic case
$(V_1, V_2)/\omega$	(-1.45, 0.63)	(-0.9, 0.0)
$(T_1, T_2)/\omega$	(-1.0, 0.5)	(-1.0, 0.0)
$(G_1, G_2, G_3)/\omega$	(0.30, -0.03, 0.15)	(0.30, 0.0, 0.0)

3.**Method and Approximations**

At first, we introduce a set of basis states with n_l (= 1 or 0) exciton and m_l (= 0, 1, 2,…) Einstein phonons at each lattice site l, as

$$\prod_l [\ (B_l^+ U_l)^{n_l} \frac{(b_l^+)^{m_l}}{\sqrt{m_l!}}\ ] |0>, \quad U_l \equiv e^{-\sqrt{S/\omega}(b_l - b_l^+)}, \quad \rangle\rangle\rangle \quad |0> \equiv \text{ exciton-phonon vacuum,} \qquad (3)$$

where U_l denotes the operator of phonon displacement, which appears or disappears, according to the presence or absence of an exciton at site l. Even if we have used this basis set, however, we still have serious difficulty, since the total number of excitons changes from 0 to about 100, while m_l also changes from 0 to about S/ω, almost independently at each site. Thus the direct calculation of this time evolution leads to too large dimensional ones. In order to overcome this numerical difficulty, we derive the following iterative method for the exciton proliferation.

We focus only on the most forwardly expanding part (the most front) of the exciton domain boundary, wherein an exciton with the excess energy coming from the photo-excitation is always included. This most front is determined by try and error method, so that it will be the most efficiently growing part of the domain boundary. The contributions from other excitons not in this front are approximated by the mean field. As proliferation proceeds by using the excess energy, the position of this front also moves. For the practical reason mentioned before, the size of this front can not be so large. As shown in the left part of Fig.2(B), we take the shaded 4 lattice sites as this front. This front (the 4 sites) is our relevant system, within which we calculate excitons, Einstein-phonons and their interactions, full-quantum-mechanically, as well as various dampings mentioned before.

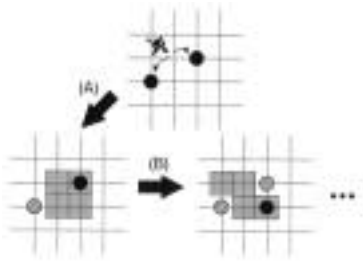

FIGURE 2. Iterative procedure in the 2-D lattice. (A) Photo-generated two excitons (for example) are replaced by a mother exciton (black circle) and a frozen one (shaded circle). (B) The shaded 4 lattice sites in the left part of (B) denotes the front. The shaded 4 lattice sites in the right part of (B) is the new front of the next generation.

Here, we define aliases of our exciton states; (1) "Mother exciton", denoted by the black circle in Fig.2(B). It is always in the front, and has an excess phonon energy inherited from the light. (2) "Frozen exciton", denoted by the shaded circle in Fig.2(B). It is in the outside of the front, and is always in the zero phonon state $m_l = 0$ defined in eq.(3)

As the proliferation proceeds, the total number of exciton in the front increases from 1 to 2, as shown in Fig.2(B). At this stage, we reconstruct a new front as schematically shown in the right hand side of Fig.2(B). The mother exciton is now frozen, and the new exciton becomes a new mother exciton. This new mother inherits the excess energy, which has now somewhat decreased from the initial excess energy, because of the dampings mentioned before. In this reconstruction, the site with the largest exciton density within the front, is taken as the site wherein the new mother resides. We call this reconstruction "generation crossover". While, the new 4 sites (new front) of the new generation is chosen by try and error method, so that they will be the most efficiently growing part around the new mother. The total energy in the system is conserved before and after this generation

crossover. We iterate this procedure, until we can get a large domain. Thus, using this method, we can numerically calculate the temporal evolution of a large system involving many excitons and phonons. It should be noted that this approximation is valid only when the excitons are rather localized, $S >> |T_1|$.

4.Results and Discussion

Figure.3 shows the exciton domain pattern calculated by using the anisotropic interactions. The shaded circle in Fig.3(A) denotes the exciton generated just after the early stage, that is, just after the successive excitation by six light pulses with a time interval 100 period. One period is $2\pi/\omega$. We can see that this domain has a fractal pattern, and it has an “island”, many “capes” and many “peninsulas” stretched outside of the domain, with a “strait”, “bays” and “gulfs” in between. These characteristics just reflect the anisotropy of the inter-exciton interactions.

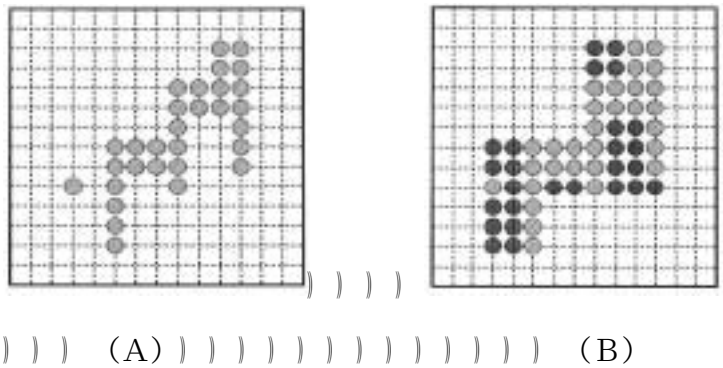

FIGURE 3. Domain pattern formed by anisotropic interactions.（A）The early stage. The shaded circle denotes the exciton.（B）The retarded stage. The black circle represents the newly grown exciton. The shaded one is same as (A).

While, the black circle in Fig.3(B) denotes the exciton generated in the retarded stage, that is, about 100000 periods after the pulse excitation, and this domain pattern is due to the tunneling type slow proliferation. We can see the aforementioned strait, bays and gulfs are now filled up by newly grown excitons, which are denoted by the black circles. Thus, the characteristic pattern is lost by the tunneling process. The pattern peculiar to the anisotropy appears only in the early stage. However, this tunneling process itself is the result of the anisotropy, and the resultant domain has become large enough to be called the PSPT.

Figure.4 shows a domain pattern calculated by using the isotropic interactions. The calculations are performed under the entirely same conditions as that of previous anisotropic case except for the inter-exciton interaction. In this isotropic case, however, only some small block type patterns are formed, and the PSPT is not successful.

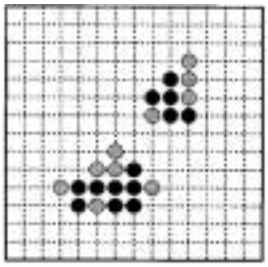

FIGURE 4. Domain pattern formed by isotropic interactions. Others are same as that of Fig.3.

5.Conclusion

We can conclude that the spatial pattern of the exciton domain formed in the early stage, sensitively inherits the spatial anisotropy of inter-exciton interactions. It is also shown that this anisotropy makes the aforementioned tunneling quite efficient, and the proliferation quite successful in the retarded stage, as compared with the case wherein the inter-exciton interaction is isotropic.

References

1.K.Nasu, P.Huai and H. Mizouchi, J.P.CM 13 (2001)R693.
2.S.Koshihara,Y.Takahashi,Y.Tokura and T.Luty, J. Phys. Chem. B103 (1999) 2592.
3.Y.Ogawa,S.Koshihara,K.Koshino,T.Ogawa,C.Urano and H.Takagi, Phys. Rev. Lett. 84 (2000) 3181.