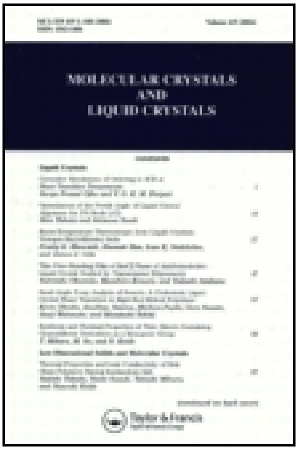

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Correlation Effects in a System of Exciton Drops

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Correlation Effects in a System of Exciton Drops

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A theoretical investigation of condensed exciton phase development was fulfilled. It was shown that in the region of phase diagram where existence of two phases (exciton liquid and exciton gas) is possible the finite value of exciton lifetime causes the following peculiarities: 1) the sizes of exciton condensed phase drops are restricted; 2) strong correlation between space positions of exciton drops exists; 3) with changing the exciton concentration in vicinity of transition point of the system from the two-phase range to the one-phase range the sharp narrowing of the drop radius distribution function occurs.

Keywords: exciton; condensed phase; lifetime; periodical structure

INTRODUCTION

The appearance of new phases under light or nuclear irradiation of crystal is a intriguing problem of phase transitions in non-equilibrium systems[1,2]. The crystal with high concentration of excitons is the example of such systems. Due to attractive interaction between excitons the condensed phase of exciton (exciton liquid) may be created. Stirring problem is problem of Bose condensation of excitons[3,4] with its interesting physical consequences: superfluidity and others. The modern sources of light irradiation allow to create the exciton density enough to phase transformation in crystal system. But usually in semiconductors the electron-hole and electron- electron correlation energies in condensed phase are larger than energy of interaction between excitons, and bond energy of electron and hole is week and consequently excitons lose their individuality and the well known and well good studied state of electron-hole liquid is created [5,6]. Since narrow value of exciton bands in organic crystals the space distance between excitons in condensed phase should have order of period of crystal lattice. In this case the processes of exciton-exciton annihilation become very intensive that hamper the creation of condensed phase. For this reason in organic crystal the condensed exciton phase was not observed so far. Several years ago for two types of semiconductors Cu_2O [7] and ZnP_2 [8], which have intermediate exciton radius value between exciton radiuses in organic and semiconductor crystals, the convincing arguments about existence of exciton condensed phases were obtained. But such phase is not equilibrium system because excitons are created by light and disappear by emission of photon or phonons. In the works [9,10] it was shown that system with high concentration of excitons is unstable with respect to their periodical space distribution. In presented article the influence of lifetime on parameters of exciton phases and spatial correlations are investigated.

QUALITATIVE ANALISIS OF PHASE TRANSITION IN SYSTEM OF UNSTABLE PARTICLES

First let us make qualitative analysis of peculiarities of phase stratification caused by finite lifetime of excitons. Typical temperature-concentration phase diagram for stable particles is presented in Fig.1. In the region 1 there is only one phase, in the region 2 two phases exist. Usually for stable particles the dynamic of phase transitions is studied at uniform distribution of particles in initial time. Such state can be realized when the system is transferred from one

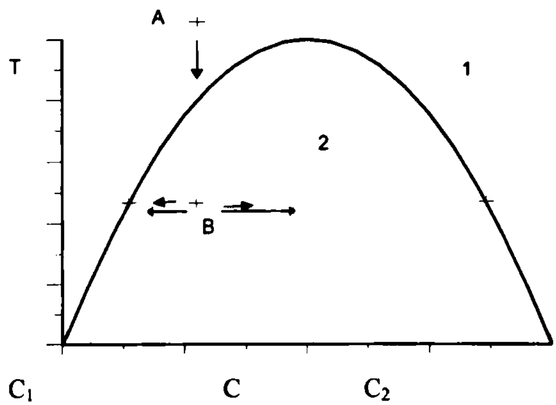

FIGURE 1. Typical phase diagram.

phase region (from point A) to point B in two phases region by some way (for example, by prompt cooling). Afterwards the creation new phases in system takes place. According Lifshits-Slyezov theory the radius of new phase increases at large time as $t^{(1/3)}$. Thus, at $t \to \infty$ the system consists of macroscopic large regions of two phases with concentrations c_1 and c_2 (see Fig.1). Also in considered problem of exciton phase development the steady state generation creates first uniform distribution of excitons. Afterwards similar to spinodal decomposition processes the phase stratifications occur. The processes try to create the states with concentrations c_1 and c_2. But due to finite value of exciton lifetime we expect appearance following peculiarities:

1) exciton condensed phase state is non-equilibrium state. Its parameters depend on lifetime of excitons and may significantly differ from parameters of equilibrium state at the same concentration of particles;

2) the size of new phase domain should be limited; created by light concentration c of excitons is less, than concentration of excitons in condensed phase c_2 (see Fig.1). Therefore, to support necessary concentration the exciton drop should derive excitons from environment due to diffusion processes. The number of incident excitons should be equal to number of excitons, which disappears inside of drop as result of finite lifetime . First value is proportional to surface of the drop, second value is proportional to volume of drop. As the result radius of

drops should be restricted and has maximal value of order of exciton diffusion length $l = (D\tau)^{1/2}$, where D is the diffusion coefficient, τ is the lifetime of exciton.

3) The spatial correlation in position of exciton drops should exist. Since the drops derive excitons from environment they can not be situated close one to another because exciton resources of environment are restricted. They can not be situated far one from another since in this case there is the possibility of appearance of new drop in space between them.

DISTRIBUTION FUNCTION OF EXCITON DENSITY

In the steady state case the system is described by the distribution function $\rho(c(\mathbf{r}))$ where exciton density $c(\mathbf{r})$ is the order parameter. To find $\rho(c(\mathbf{r}))$ it is necessary to solve kinetic equation which depends on the kind of interaction between particles. To consider the problem in general we will present the dependence of the free energy on the fluctuating fields $c(\mathbf{r})$ in Landau- Ginzburg form. Fokker - Planck functional equation is given by

$$\frac{\partial \rho}{\partial t} = -\int d\mathbf{r} \frac{\delta (j_d + j_r)}{\delta c(\mathbf{r})}, \tag{1}$$

where j_d is the probability current due to diffusion processes

$$j_d = M\Delta\left(\frac{\delta F}{\delta c(\mathbf{r})} + \kappa T \frac{\delta \rho}{\delta c(\mathbf{r})}\right), \tag{2}$$

M is the mobility, which is proportional to diffusion coefficient D, F is Landau - Ginzburg free energy , j_r is the probability current caused by processes of creation and decay of particles

$$j_r = -\frac{c(\mathbf{r}) - c_0}{\tau} - \frac{1}{2}\frac{\delta ((G + c(\mathbf{r})/\tau)\rho)}{\delta c(\mathbf{r})}, \tag{3}$$

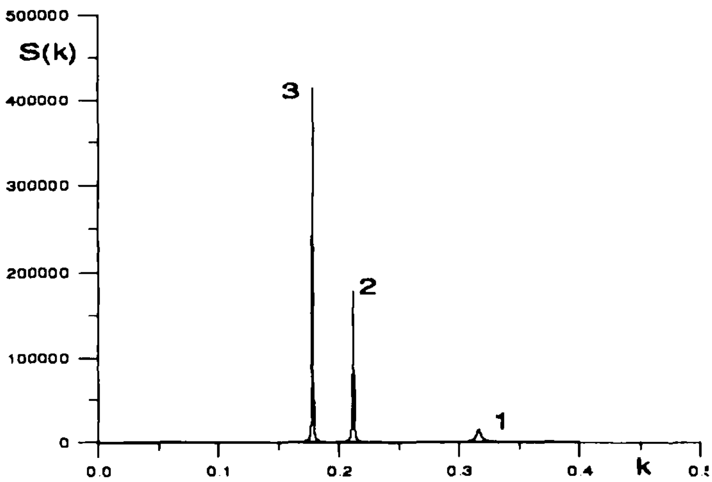

FIGURE 2. Fourier transform of correlation function for different values of dimensionless lifetime τ:1)100, 2)500, 3)1000.

$c_0 = G\tau$ is the mean value of particle concentration, G is the generation rate. The term j_d is well known in literature, the term j_τ takes into account the finite value of exciton lifetime [10].

The correlation function of exciton density and Fourier transform equal

$$S(\mathbf{r}-\mathbf{r}') = < u(\mathbf{r})u(\mathbf{r}') >, \quad (4)$$

$$S(\mathbf{k}) = \int d\mathbf{r} S(\mathbf{r}) \exp(-i\mathbf{k}\mathbf{r}). \quad (5)$$

where $u(\mathbf{r}) = c(\mathbf{r}) - c_0$.

In order to solve Eq.(1) we used the approximation of two-point distribution function [11] in following form

$$\rho_2(u(\mathbf{r}), u(\mathbf{r}')) = \rho_1(u(\mathbf{r}))\rho_1(u(\mathbf{r})[1 + \frac{S(\mathbf{r}-\mathbf{r}')}{< u^2 >} u(\mathbf{r})u(\mathbf{r}')] \quad (6)$$

Further we present the results only for correlation function.

In stratification range the Fourier transform of correlation function $S(\mathbf{k})$ has sharp maximum at $k \approx \tau^{-1/4}$ (Fig.2) that provides the oscillations of correlation function $S(\mathbf{r})$ as function of coordinate. At large lifetime the period of oscillations tends to infinity. The Fourier transform $S(\mathbf{k})$ describes the intensity of light scattering by system. In scattering picture side by side with center spot that origins from incident bunch of light the ring should appear which is connected with formation of regular structure with period $\lambda = 2\pi / k$. At spinodal decomposition of stable particles the periodical structures arise also in initial stage and the radius of ring decreases with time and tends to zero. In contrast to spinodal decomposition phenomenon in considered system of unstable particles the period of structure depends on diffusion coefficient and radius of the light scattering ring is fixed.

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