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LATTICES OF ISLANDS OF ELECTRON-HOLE LIQUID IN DICHALCOGENIDES UNDER OPTICAL PUMPING

The formation of islands of the electron-hole liquid in the case of uniform light irradiation of transition metal dichalcogenides such as MoS₂ and MoTe₂ has been simulated numerically. The kinetics of exciton capture by the islands has been considered, and the distribution of the exciton density around the islands has been calculated accounting for the correlation of islands' positions in the exciton gas and the boundary conditions under various uniform pumping shapes. The size of the electron-hole liquid islands has been estimated. The optimal spatial arrangement of the electron-hole liquid islands and the dependence of the formed structure on the system's parameters have been found.

Keywords: dichalcogenides, exciton gas, electron-hole liquid, two-dimensional lattices.

1. Introduction

The formation of structures in a nonuniform density distribution of the electron-hole liquid (EHL) in dichalcogenides is a challenging topic in solid-state physics and materials science. To date, substantial experimental data on the EHL properties in various crystals and under different conditions have been accumulated (see reviews [1–4]). The theory of the EHL droplet formation in bulk Ge and Si materials has been studied in works [5–7]. The development of nanophysics brought about the discovery of the EHL in several semiconductor structures with quantum wells, in particular, in the Si/Si_{1-x}Ge_x/Si system [8, 9], the SiO₂/Si/SiO₂ system of various thicknesses [10], and double quantum wells in SiGe/Si [11] and GaAs/AlGaAs [12–14] heterostructures.

In recent years, the EHL was discovered in new van der Waals heterostructures, namely, transition metal dichalcogenides (TMDs) [15, 16]. In these materials, electron-hole liquids, which are a result of the electron or hole excess, can be formed. Single-

layer dichalcogenides have a layered structure, where atoms in a layer are bound via strong covalent bonds, whereas weaker van der Waals forces act between the layers. This circumstance allows the easy separation of the layers to be performed, thus creating new possibilities for manipulating the electronic structure. A TMD is a thin monolayer semiconductor consisting of a transition metal and a chalcogen. One layer of metal atoms is sandwiched between two layers of chalcogen atoms, which is expressed by the formula MX₂, where M stands for the metal, and X for the chalcogen. The family of semiconductor TMDs is a particularly promising platform for a fundamental research of two-dimensional (2D) systems with their potential applications in optoelectronics, because they have a direct band gap within a monolayer and a highly efficient “light-matter” coupling. Their crystal lattice with the broken inversion symmetry together with strong spin-orbit interaction leads to a unique combination of spin and valley degrees of freedom. Furthermore, the 2D character of the monolayers and weak dielectric screening from the environment lead to a considerable enhancement of the Coulomb interaction. The resulting formation of bound electron-hole (e-h) pairs, or excitons, dominates over the optical and spin properties. Thus, atomically thin TMDs – such as MoS₂, WS₂, and others – possess unique electronic prop-

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erties, which can be important for a wide range of applications [17].

In bulk semiconductors, if the excitation intensity is not very high, the EHL consists of individual EHL droplets. In 2D systems, the EHL is formed as separate islands located in the quantum well plane. As a result of the carrier recombination processes, every island has a finite radius, whose dimensions are determined by the recombination rate and the irradiation intensity. Important are the issues concerning the size of EHL islands and their mutual arrangement driven by the interaction between individual islands. The lattice in the islands can arise as a result of the interaction between electrons and holes, which leads to their organization into spatial structures similar to crystal lattices. Such structures can have topological properties, which opens new possibilities for information storage and processing. In particular, the emergence of topological phases in systems with the EHL, which can affect the electronic properties of dichalcogenides, was discussed in work [13]. A possibility to control the EHL by changing the synthesis conditions such as the temperature or the pressure, which can substantially affect their properties, was pointed out in work [18].

Currently, there is almost no information in the literature concerning EHL droplets in TMDs, and the interaction between the islands has not been considered so far. The task of studying the EHL in low-dimensional structures remains challenging, because spatial structures created and controlled making use of light in 2D systems are promising for optoelectronics and spintronics as fast-switching models or energy-efficient memory elements. The EHL can demonstrate new collective phenomena, which opens new horizons for applications in quantum electronics [9]. Important are interdisciplinary studies in this area including physics, chemistry, and materials science [16]. The EHL capability in spintronics and quantum computers was discussed in work [17], where the importance of such research for future technologies was emphasized. The authors of the research article [19] showed how the EHL can be used in new sensors and elements based on single-layer dichalcogenides. Also interesting is the study of the nuclear irradiation effect on the TMD surface structure, which is of interest for radiation physics [20, 21].

In general, the problem of EHL formation in single-layer dichalcogenides is challenging, but promising for

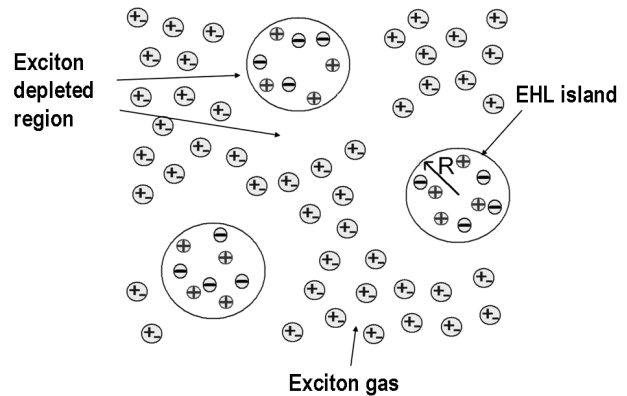


Fig. 1. Schematic diagram for the formation of EHL islands from the exciton gas

research and potential applications, which opens up new opportunities in materials science and promises both new discoveries in materials physics and applications in various technologies [22].

2. Problem Formulation and Applied Consideration Method

Light irradiation excites excitons in a dichalcogenide monolayer. We consider the case where the light flux differs from zero only on some part of the dichalcogenide 2D surface, being uniformly distributed over it. The illuminated section can have the shape of a square, disk, stripe, and so forth. It is a region, where excitons are excited. Since the energy of the “electron-hole” pair in the EHL is higher than the exciton binding energy, the EHL is formed in the system. In the considered 2D system, the EHL has the form of islands (Fig. 1).

Excitons are captured by the islands, whereas the e-h pairs can leave the latter. So, there is an exchange between the islands and the environment. The task is to find the island sizes, the mutual island arrangement, and the distribution of exciton density outside the islands. It was solved for the islands of exciton condensate [23], applied to the EHL [24], and the results are used in this work to calculate the parameters of the EHL islands in dichalcogenides. In this approach, we will estimate the probabilities for the structure itself and its specific configuration to appear making use of a distribution function.

It is assumed that the islands have a circular shape, the distribution of e-h pairs in the islands is uniform, and the sizes of the islands are much smaller than the

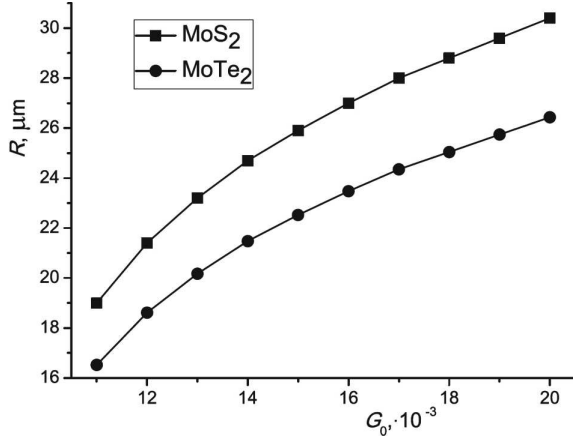


Fig. 2. Critical radius of EHL island

distance between them. In works [23, 24], a common solution was found and used:

- 1) for the kinetic equation describing the distribution function of the condensed exciton phase islands over their size, relative position, and so on;
- 2) for the equation describing the distribution of exciton density $c(\mathbf{r})$ outside the islands,

$$D_X \Delta c - \frac{c}{\tau_X} = -G(\mathbf{r}), \quad (1)$$

where D_X and τ_X are the diffusion coefficient and lifetime, respectively, of excitons, and G is the pumping intensity.

Boundary conditions to Eq. (1) are imposed at the surface of every island. The value of the exciton density at the surface of a particular island depends on the positions of other islands, which determines the connection between the islands. The interaction between the islands, which can lead to a redistribution in the gas density and the formation of structures with an increased density, occurs through the diffusion fields of free excitons, which are created by an external source and form the islands. A neighboring island reduces the concentration of excitons near a considered island and slows down its growth. Thus, it is beneficial for two islands to move away from each other. But, at a certain distance between them, due to the constant pumping, there appears a region where the exciton concentration exceeds a critical value, and conditions for the appearance of a new island are formed. Therefore, there is an optimal (most probable) spatial arrangement of the islands.

The calculations were performed in Mathematica and using parameter values taken from experimen-

tal data [15, 25–31]. For some parameters, the literature data are either different or not established, so they were estimated. For a monolayer MoS_2 , the critical density, and the binding energy per one e-h pair of the EHL are $c_i = 3.8 \times 10^{11} \text{ cm}^{-2}$ and $E_{\text{EHL}} = 380 \text{ meV}$, respectively; the diffusion coefficient and the binding energy of excitons are $D_X = 2.1 \text{ cm}^2/\text{s}$ and $E_X = 240 \text{ meV}$, respectively, the EHL (exciton) lifetime is $\tau = 4 \times 10^{-7} \text{ s}$ ($\tau_X = 9.5 \times 10^{-7} \text{ s}$), and the exciton diffusion length is $l_D = \sqrt{D_X \tau_X} = 0.14 \text{ mm}$. For a MoTe_2 monolayer, $c_i = 5 \times 10^{12} \text{ cm}^{-2}$, $E_{\text{EHL}} = 150 \text{ meV}$, $D_X = 5 \div 8 \text{ cm}^2/\text{s}$, and $E_X = (250 \div 400) \text{ meV}$. The temperature $T = 20 \text{ K}$ was assumed in all calculations.

3. Study of the EHL Structure at Various Pumping Forms

Before studying the interaction and arrangement of EHL islands, we will firstly solve the problem of how a single island is formed from the exciton gas. Then we will consider the 2D problem concerning the arrangement of EHL islands in TMDs in the case where the irradiation is performed by pumping in the form of an infinitely long uniform stripe of finite thickness.

The plots in the Figures below correspond to the most probable values of relevant physical quantities described by the distribution functions: the radii of the islands, the distances between them, and so forth.

3.1. Formation of a single island

The distribution function of a single EHL island determines the most probable value of its radius R for a given pumping intensity G . It is used to determine the threshold pumping intensity value G_{cr} , required for the formation of a single island. In dimensionless units, the distribution function of a single island has the form

$$f(R) = f_0 e^{S(R)}, \quad (2)$$

where the quantity $S(R)$ accounts for the probability that an exciton will be captured by the island, as well as the probability of e-h pair emission by the island. The corresponding procedure was described in detail in work [24].

The maximum of the distribution function (2) corresponds to the most probable value of the island radius at a given pumping intensity (Fig. 2). Hereafter,

the pumping value is given in $G_{cr}\tau/c_i$ units. Numerical calculations showed that the critical radius R of the EHL islands equals $28 \mu\text{m}$ in MoS_2 dichalcogenide and increases with the growth of pumping intensity. At the same time, for MoTe_2 , the critical radius equals $R = 26 \mu\text{m}$. The pumping intensity threshold becomes lower as the exciton lifetime or the temperature decreases.

3.2. Narrow-stripe pumping mode

After determining the critical island radius, we can find the optimal distance between the islands; it corresponds to the maximum in the common distribution function of the nearest islands. Let us consider the formation of EHL islands in a TMD under uniform irradiation of the dichalcogenide layer by its pumping in the form of an infinite horizontal stripe, the vertical size of which is such that the formation of islands is possible only along the stripe line, i.e., horizontally (Fig. 3). The vertical size of the stripe will be called the stripe thickness. Let the stripe thickness equal $2h$.

Let us find the exciton density distribution in the absence of other islands. In the system under consideration (irradiation in the form of an infinite narrow stripe), we can obtain the exact solution of Eq. (1),

$$c_0(\mathbf{r}) = G_0\tau_X \begin{cases} 1 - \exp\left(-\frac{h}{l_D}\right) \text{ch}\frac{y}{l_D}, & |y| \leq h \\ \exp\left(-\frac{|y|}{l_D}\right) \text{sh}\frac{h}{l_D}, & |y| > h. \end{cases} \quad (3)$$

Now, we should determine the maximum of the distribution function (2) for a chain of islands, where c_0 enters as a parameter, and account for the boundary conditions. In this case, we can consider the interaction of only closely located islands.

The periodic arrangement of islands is assumed to be the most probable. To assess the probability of system deviations from the periodic arrangement, the following analysis was performed. Let only one island shift from its equilibrium position, without changing the positions of the other islands. The relevant calculations show that a sharp peak in the probability density takes place exactly in the section corresponding to the periodic arrangement of islands along the stripe, whereas the positions after the island displacement are less probable. Furthermore, the relative shift of the island position in the stripe (this is the root-mean-square deviation of the shift of one island

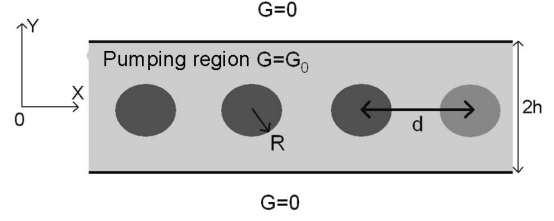


Fig. 3. Formation of EHL droplets from the exciton gas in the case of uniform irradiation of TMD in the narrow-stripe pumping mode

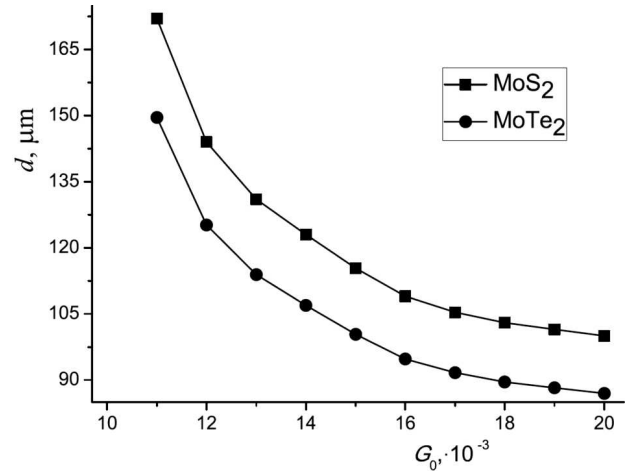


Fig. 4. Distance between EHL islands in a dichalcogenide monolayer in the case of uniform irradiation of TMD in the narrow-stripe pumping mode

divided by the distance between the islands) decreases substantially as the pumping intensity grows. This fact means that the system has attributes of periodicity, so that the system can be considered as ordered at high pumping intensity values.

According to calculations, in a system of equidistant EHL islands, the most probable distance d between the islands in MoS_2 dichalcogenide is about $200 \mu\text{m}$ and decreases with the irradiation intensity growth (Fig. 4). Accordingly, for a MoTe_2 single crystal, $d \approx 150 \mu\text{m}$.

3.3. Wide-stripe pumping mode

Let us consider the problem of EHL formation under a uniform pumping of the dichalcogenide layer, as its thickness $2h$ increases. Owing to diffusion processes, it becomes possible to arrange several islands in the vertical direction (the OY -axis). In this case, the interaction between the nearest islands should be

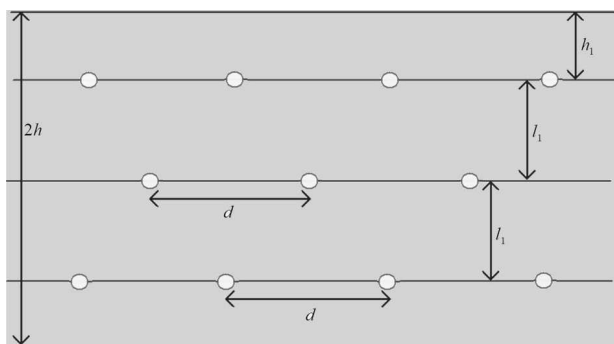


Fig. 5. Optimal arrangement of EHL islands in the case of uniform irradiation of TMD in the wide-stripe pumping mode

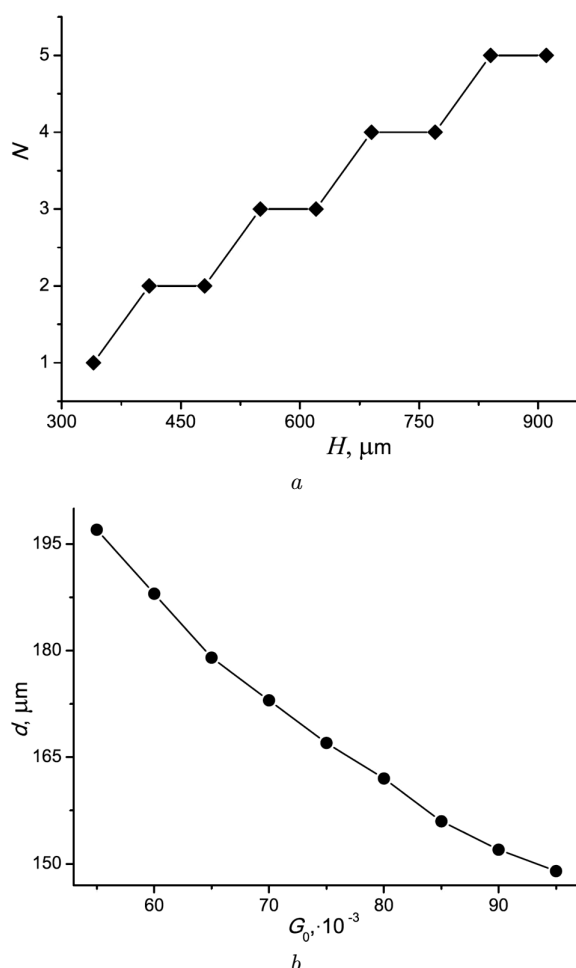


Fig. 6. Dependence of the number of EHL island rows in the stripe on the stripe thickness at fixed uniform stripe pumping (a); dependence of the distance between islands in a row on the pumping intensity (b)

taken into account not only in the horizontal direction, but also in the vertical one. Assuming that the most probable distance d between the islands in a row has already been established and using the distribution function for the nearest islands in neighboring rows, it was found that the most optimal arrangement of rows with equidistant EHL islands is reached, if the islands in a new row are located in the middle between the islands in the neighboring row, i.e., in the checkerboard pattern (Fig. 5). The distances between the islands in a row decrease, as the irradiation intensity increases.

Using numerical calculations, it was found that the most optimal for a MoS₂ monolayer is the following arrangement of three rows of EHL islands with the radius $R = 36 \mu\text{m}$ at the pumping intensity $G_0 = 81 \times 10^{-3}$ and the stripe thickness $2h = 620 \mu\text{m}$: $d = 162 \mu\text{m}$, $l_1 = 183 \mu\text{m}$, and $h_1 = 127 \mu\text{m}$. For a MoTe₂ monolayer with the islands of radius $R = 30 \mu\text{m}$ and at the pumping intensity $G_0 = 52 \times 10^{-3}$, the following parameter values are optimal: $2h = 500 \mu\text{m}$, $d = 132 \mu\text{m}$, $l_1 = 156 \mu\text{m}$, $h_1 = 94 \mu\text{m}$.

The number of rows of the EHL islands as a function of the irradiated stripe thickness $H = 2h$ was analyzed separately. This dependence is “stepwise”, i.e., the number of rows increases abruptly as the thickness H grows, with the distances l_1 between separate rows being somewhat smaller than the distance d between islands in a row. The corresponding result for MoTe₂ is presented in Fig. 6.

4. Conclusions

The formation of the EHL spatial structure in monolayers of dichalcogenides MoS₂ and MoTe₂ under light irradiation has been considered in the framework of stochastic approach. The sizes of EHL islands in those dichalcogenides equal tens of microns, and the distance between them equals hundreds of microns, which is substantially larger than the corresponding EHL sizes in semiconductor structures with quantum wells (silicon and germanium). Structures with such scales, which are close to macroscopic, have not been observed yet in the experiment, because specimens of TMD monolayers with rather large sizes should be used for this purpose.

The formation of EHL islands in TMD was studied in the case of a uniform pumping of a dichalcogenide monolayer in the shape of a stripe. The stripe thick-

ness was varied from a value at which the formation of islands was possible only along the stripe line to those at which the formation of several rows of EHL islands was possible. In the case of narrow stripe irradiation, the most probable was the equidistant arrangement of round islands. In the case of wide stripe irradiation, the most optimal was the equidistant arrangement of the islands in a checkerboard pattern, with the distances between the islands in a row decreasing, as the irradiation intensity grew. The number of island rows increased with the growth of the irradiated stripe width.

The calculations were performed for the MoS₂ and MoTe₂ parameters and can be repeated for other TMDs and other pumping geometries.

The formation of EHL islands in TMDs is a phase separation in a heterosystem under non-equilibrium conditions. The ordered arrangement of EHL islands in the dichalcogenide layer is an example of a new type of macroscopic lattice in 2D structures.

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ҐРАТКИ ОСТРІВЦІВ
ЕЛЕКТРОННО-ДІРКОВОЇ РІДИНИ
В ДИХАЛЬКОГЕНІДАХ
ПРИ ОПТИЧНОМУ НАКАЧУВАННІ

Виконано чисельне моделювання формування електронно-діркової рідини у випадку однорідного світлового опромінення дихалькогенідів перехідних металів типу MoS₂ та MoTe₂. Досліджено кінетику захоплення екситонів острівцями та обчислено розподіл густини екситонів навколо острівців, враховуючи кореляцію в положеннях острівців у газі екситонів та межові умови при різних формі однорідного накачування. Оцінено розміри острівців електронно-діркової рідини. Встановлено оптимальне просторове розміщення острівців електронно-діркової рідини та залежність утвореної структури від параметрів системи.

Ключові слова: дихалькогеніди, екситонний газ, електронно-діркова рідина, двовимірні ґратки.