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Several Steps toward Fundamentals of Physics of Light-Matter Coupling

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Making no pretense at exhaustiveness we essay below a survey of several fundamentals of physics of light-matter coupling.

The microscopic theory of optical phenomena in crystals is closely related to the theory of excitons, which provides an appropriate tool for elucidation of specifics of interaction between electromagnetic fields and crystals and for construction of the physical picture of energy transfer in crystalline media. It is well established that in a crystal, where the translation symmetry allows for excitation of any of its elementary cells (or any of the comprising structural units), the energy transfer of electronic excitations occurs due to the motion of quasi-particles called excitons. The concept of "exciton" was originally introduced into physics by the pioneering works of Frenkel [1]. Among the first to consider excitonic states was also Peierls [2]. These works gave a theoretical explanation for the experimentally observed photoelectrically negative light absorption. Utilizing Heitler-London-Heisenberg method in his study of electronic states Frenkel [1] demonstrated that the absence of photoconductivity under the light absorption by an electronic subsystem in an ideal crystal stems from excitations of crystalline structural units moving in waves (due to translation invariance) along a crystal. Such excited states were given the name of "Frenkel excitons".

Another excited no-current state of crystal electrons was considered by Wannier [3] and Mott [4]. It consists of an electron (in conductivity band) and a hole (in valence band) bound by coulombic forces. The center of inertia of such a hydrogen-like formation moves freely along a crystal. Wannier-Mott exciton motion can be described by the effective mass method. If to consider an excited state of a molecule as an electron-hole pair, the Frenkel exciton can be termed a small-radius exciton, while that of Wannier-Mott should be recognized as a big-radius one (with radius much bigger than the lattice constant). Excitations of the first type are mostly observed in molecular and alkali-halide crystals, whereas the second type is characteristic to semiconductors with narrow band gaps and a large dielectric permittivity.

A fairly exhaustive coverage of exciton theory and a detailed bibliography on the subject can be found in [5-9]. Here we shall mention some of the works of principal importance.

An experimental evidence for the existence of Wannier-Mott excitons was obtained by Gross et al. [10] by observing hydrogen-like spectra near the band-gap absorption edge. According to a well-known Rydberg formula the pattern of optical absorption lines in CuO_2 stands in correspondence with the excitonic levels. Important investigations have also been carried out by Davydov [11] and his co-authors in the papers devoted to molecular excitons [11], where they for the first time considered the splitting of exciton states resulting from the presence of more than one molecule in an elementary cell. It was named Davydov splitting in contrast to Bethe splitting of degenerate atomic energy levels caused by intracrystalline field. Worthy of mentioning is also the paper of Heller and Marcus [12], which for the first time reports on the splitting of energy levels of transverse and longitudinal excitons. The discontinuous character of exciton energy dependence on the wave vector k for k=0 was established by Pekar [13], where he took a proper account of long-range electric interactions between structural units in a crystal. Pekar [13] generalized the concept of exciton by calling "excitonic" an excited state in a non-conducting crystal characterized by a single continuous quantum number (quasi-impulse) with the rest of quantum numbers being discrete. Apart from the above mentioned Frenkel and Wannier-Mott electronic excitons this general definition applies to optical phonons, molecular vibrons (electron-oscillation excitations), spin waves etc.

The problem of electromagnetic field oscillations "coupled" with crystal elementary excitations in the infrared spectrum region (where of significance are the optical branches of lattice oscillations) was for the first time solved on the basis of a semi-classical approach by Tolpygo [14] and independently by his sidekicks Huang and Born [15, 16]. Various solutions of this problem based on polariton approach were given by Fano [17], Hopfield [18] and Agranovich [19]. It will be recalled that the term "polariton" was introduced by Hopfield [18] to denote the quantum of polarization oscillations of crystal structural units, which does not interact with a light wave. He proposed a crystal model, where atoms are replaced by uncoupled charged harmonic oscillators. Hopfield [18] considered a combination of "polariton" and vacuum photon, the so called crystal photon. It was for the latter that the term "polariton" was subsequently reserved.

There is an additional concept of exciton polariton introduced by Pekar [13] to describe the mixed exciton-electromagnetic waves in crystals. The theory of exciton polaritons developed by Pekar [13] and his co-authors proved extremely fruitful for the solution of problems of modern crystal optics with due account for interaction delay between crystal structural units and also for spatial dispersion, long-range coulombic interactions and a strong back action of crystal particles on electromagnetic waves. Numerous works devoted to this subject have been published in 1970-1980 [20-24].

According to modern understanding a crystal is constituted by a multitude of charged particles – electrons and atomic nuclei (for big kinetic energies one should also account for production and annihilation of electron-positron pairs). Due to electromagnetic interactions (which for the most part define the physical and chemical properties of crystals) particles execute a complex motion, which results in a certain ordered structure of a crystalline medium. The generally formulated problem of the dynamics of "crystal-electromagnetic field" system is reduced to a self-consistent solution of equations, defining the evolution of crystal structural units under the action of the field combined with field equations. Performing the necessary calculations

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usually requires an employment of certain approximation chosen individually in for each particular case. The choice depends on a crystal model as well as on the nature of excitations involved.

Results in crystal optics obtained during the past fifty years provide a solid foundation for the progress of modern photonics. Concepts developed in the physics of crystalline solids can potentially be applied to the physics of photonic supercrystals. While the theory of impurity bands and excitons in semiconductor crystals has been constructed in 1970-1980, an analogous theory for photonic crystals is yet to be completed. Recent experiments and theoretical investigations reveal an intense interest for polaritonic structures and systems of coupled microresonators [25], whose applications include fabrication of clockworks of unprecedented accuracy [26,27] as well as the sources of coherent irradiation. There has been a significant advance in the photonics of imperfect structures. A number of our recent works have been devoted to optical activity of imperfect photonic crystals [28] and to dispersion of exciton-like electromagnetic excitations in non-ideal lattices of coupled microresonators [29,30]. Such investigations provide the necessary theoretical background for manufacturing of composite structures with controllable propagation of electromagnetic waves.

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