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Excitons and optical nonlinearities in hybrid organic-inorganic nanostructures

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Properties of electronic excitations in nanostructures based on organic materials and inorganic semiconductors, having respectively Frenkel excitons and Wannier – Mott excitons with close energies, are discussed. It is known that Frenkel excitons can have large oscillator strengths. At the same time, Wannier – Mott excitons are characterized by large resonance optical nonlinearities because of comparatively low saturation concentrations. In nanostructures containing organic and inorganic semiconductor quantum wells, the resonance interaction between exciton states in quantum wells results in the hybridization of Frenkel and Wannier – Mott excitons [1]. New exciton states may have, as Frenkel excitons, large oscillator strengths of the transition and, at the same time, as Wannier – Mott excitons, they may exhibit large resonance optical nonlinearities. As a result, these nonlinearities increase several hundreds times compared to nonlinearities inherent in the semiconductor quantum well [2]. A similar effect was also considered in a microcavity, where exciton resonances are close to the photon resonance in a microcavity [3]. For the case of small resonance splittings compared to the width of the exciton resonance in an organic layer, the irreversible energy transfer from an exciton in a semiconductor quantum well to the organic material is considered. This transfer is analogous to the Förster transfer. For sizes of a semiconductor quantum well and a barrier of the order of 100 A, energy transfer occurs over a time which is much shorter than the exciton lifetime in the semiconductor quantum well [4]. This effect can be of special interest for applications: electric pumping of excitons in a semiconductor quantum well can be used to produce bright luminescence of organic molecules (see review [5]).

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The outlook for nanolocal femtosecond spectroscopy and nanolithography

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Achievements in nanophysics and technology in the field of nano- and optoelectronics are associated with the advancement to progressively smaller spatial and time scales. This requires the development of fundamentally new methods for fabricating nanostructures and nondestructive control. One of the important problems of nanophysics is the development of optical methods that combine high spatial, time, and spectral resolution and allow one to study ultrafast processes in single nanostructures, clusters, and molecules.

In this connection, we will discuss several problems related to the action of laser pulses on a system consisting of the tip of a scanning probe microscope (SPM) and a substrate, and the use of the possibilities appearing in connection with these problems in nanooptics and nanotechnology.

Consider first an auxiliary problem about natural local plasma vibrations in a system consisting of the SPM metal tip and a substrate and their excitation by laser radiation (see Refs [1, 2] and references therein). If the distance d between the tip and a substrate is considerably smaller than the radius of curvature R of the tip, then the localization radius L of a plasmon is of the order of $\sqrt{dR}$ and increases with increasing d. The eigenfrequencies of the system also depend on R and d, and, of course, on the material of the tip and substrate, the minimum frequency related to in-phase vibrations of electrons in the tip and substrate decreasing as d decreases (detailed calculations are reported in Ref. [1]).

An external electromagnetic field focused in the region under the tip sharply increases near it due to the 'lightning rod effect' and also if the above modes are resonantly excited. In this case, the external field frequency, the distance between the tip and substrate, etc. are controlling parameters of the problem. The localization region of a strong near field is determined by the polarization of the tip-substrate system and decreases at $d < R \ll \lambda$ to a size considerably less than the wavelength. Thus, for $R \sim 20$ nm and $\lambda = 620$ nm, the localization radius of a strong field under a tungsten tip is approximately 20, 7, and 4 nm for d equal to 5, 0.5, and 0.3 nm, respectively. The field strength under the tip can be, depending on its shape, a nonmonotonic function of d and, in addition, it strongly depends on the value ε of the one-particle decay at a given frequency. In this connection, it would be profitable to use silver as a substrate and (or) coating for the SPM tip. As for the Landau damping of local plasmons (LPs), its ratio to the frequency is of the order of r_{TF}/L (where r_{TF} is the Thomas – Fermi screening radius) [2], so that it can be considerable only for LPs that can appear on irregularities of the SPM tip.

Thus, the 'lightning rod effect' and (or) resonance excitation by an external laser field of plasma vibrations induce a strong near field in the SPM tip-substrate system. This field can be used for both optical studies and with a subwavelength resolution (nanooptics) and nanotechnology.

As for nanotechnology, a strong field in the subwavelength region can be used to modify a surface at the nanometer scale (see below for a discussion of the relevant experiment), to induce nanolocal chemical reactions, to perform nanolocal ultrahigh-density magneto-optical (or