with the help of a so-called near-field microscope [9]. Thus, having all the required elements, we started to realize near-field microscopy using resonance energy transfer.

However, the parameters of the resonance energy transfer can substantially change near nanostructures, in particular, near the nanotip. This is a typical problem of nanooptics, which was theoretically studied in Ref. [10] to gain a deeper understanding of the microscope being developed. We modeled the nanotip as a dielectric sphere of radius $a \ll \lambda$ and, indeed, found a noticeable difference between the resonance energy transfer in the case of the surface curvature at the nanometer scale and the cases of free space and plane surface. This difference is mainly caused by a different mismatch between the frequencies of the donor and acceptors located in different regions of a nanomedium (Fig. 2).

Another interesting effect in nanooptics is a change in the probability of forbidden (in particular, quadrupole) transitions near a dielectric nanotip. This problem was theoretically studied in Ref. [11]. Figure 3 shows that a nanosphere of radius $r \ll \lambda$ enhances the ratio of the probability of the quadrupole transition to that of the dipole transition by a factor of $(\lambda/r)^2$.

Of course, many other optical effects are also modified near nanostructures.

Finally, note that a laser photoelectron microscope can be used for parallel and sequential reading of information in systems of ultrahigh-density optical memory (10^{11} bit cm^{-2}) [12].

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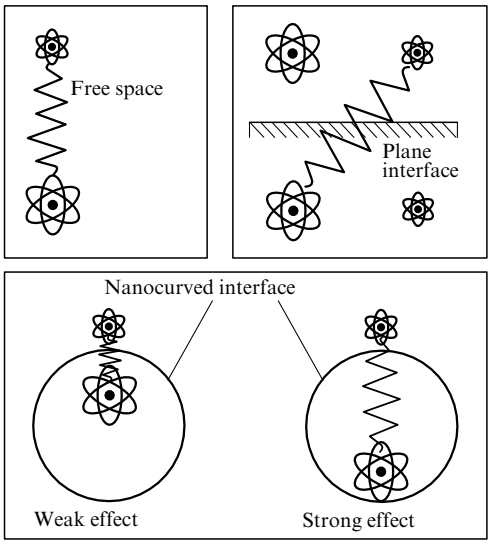

Figure 2. Qualitative explanation of the change in the rate of resonance energy transfer between dipoles separated by a 'nanointerface' compared to the cases of free space and a plane surface. The effect is caused by a change in the matching between frequencies of the donor inside a nanosphere and an acceptor [10].

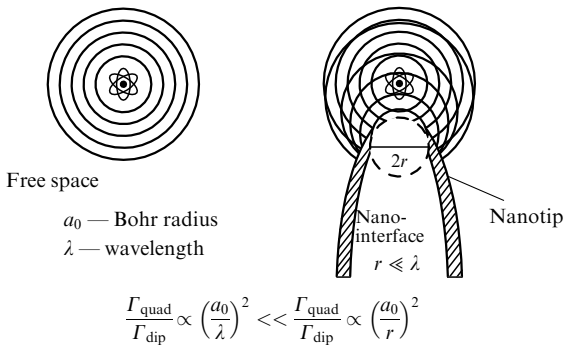

Figure 3. Qualitative explanation of the change in probability of the quadrupole transition compared to that of the dipole transition near a nanosphere [11].

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Spectroscopy of polaritons in a semiconductor microcavity

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1. Introduction

At present, the optical properties of thin dielectric and semiconductor films on metal surfaces, the excitation of polaritons in these structures (see, for example, Refs [1, 2]), and the nonlinear optics of thin-film microcavity structures of Fabry – Perot resonator type [3] are attracting much attention. These systems represent a pithy model of nonlinear optics: planar microcavity structures are a simple physical model, which permits a sequential account of boundaries when a resonance medium of finite thickness is used. In addition, these systems are promising for the study of the nonstationary Kasimir effect and other phenomena of quantum electrodynamics in a cavity (see Ref. [4] and references therein).

This work is devoted to the study of effects of the nonlinear interaction of ultrashort light pulses with microcavity semiconductor/metal structures. We studied the excitation and relaxation of eigenmodes in an A^2B^6 semiconductor microcavity using femtosecond laser spectroscopy [5, 6].

2. Experiment

As planar cavity structures, we used semiconductor films (ZnS, ZnSe) of different thicknesses (0.25 – 1.2 μm) grown on a metal layer (Ni, Cr, Cu) deposited onto a quartz substrate. Excitation was performed by 50-fs optical pulses. The different photon energies $\hbar\omega_{pu1} = 2.34$ eV, $\hbar\omega_{pu2} = 2.75$ eV, and $\hbar\omega_{pu3} = 5.5$ eV used in the experiments allowed us to study the system response upon excitation below the energy gap, when $\hbar\omega_{pu} < E_g$ (E_g is the band gap) and above the energy gap, when $\hbar\omega_{pu} > E_g$ [7 – 11]. The dynamics $\Delta D_R(\hbar\omega) = -\Delta \log_{10}[R(\hbar\omega)]$ of photoinduced reflection was studied over a broad spectral range from 1.6 to 3.2 eV with a 7-fs time step and maximum delay of up to 2.5 ps using 50-fs probe pulses.

Studies of the time evolution of photoinduced reflection spectra for both types of excitation [7 – 11] showed that in the case of excitation below the energy gap ($\hbar\omega_{pu1} = 2.34$ eV for the ZnSe/Cr and ZnSe/Cu structures and $\hbar\omega_{pu2} = 2.75$ eV for ZnS/Ni), the response substantially depends on the metal. The photoinduced response of the ZnSe/Cr structure is of alternating sign for positive time delays $\tau_D = 0.04 – 1$ ps, whereas for the ZnSe/Cu and ZnS/Ni structures it is of fixed sign (except the region near the energy gap edge of ZnSe/Cu) for all time delays used. At the same time, the response of the ZnSe/Cr structure at short negative time delays $\tau_D = -0.04 –$ 0.0 ps is qualitatively similar to that of the ZnS/Ni structure upon excitation below the energy gap.

For the ZnS/Ni structure, two stages can be separated: a fast one, with a characteristic time of the order of the pulse duration, and a slow one, with a characteristic time of the order of 0.4 ps. During the fast stage, the signal achieves a maximum for delays of the order of 0 fs and then decreases. Then, it again increases (on the time scale of 0.4 ps) and after that slowly relaxes to zero for delays $\tau_D > 1$ ps. The response of the ZnSe/Cu structure is similar to that of the ZnS/Ni structure (except the fast stage, which was not observed in ZnSe/Cu).

For excitation above the energy gap ($\hbar\omega_{pu2,3} = 2.75$ and 5.5 eV for ZnSe/Cr and $\hbar\omega_{pu3} = 5.5$ eV for ZnS/Ni), the responses from all the structures under study were qualitatively similar. For short delays $\tau_D < 0.5$ ps, an alternating signal was observed, which reached a maximum for a delay of about 0.1 ps and then relaxed with a characteristic time of about 1 ps to a signal of fixed sign.

3. Discussion

Excitation below the energy gap. Among the physical processes proceeding upon excitation below the energy gap [7, 8], electron excitation in metal and tunneling of nonequilibrium charge carriers into a semiconductor are directly related to the band structure of the metal.

Upon excitation below the energy gap, an optical pulse is mainly absorbed in a thin surface layer of a metal ($\sim l_{ex} = 2/\alpha_{metal}(\hbar\omega) \sim 20 – 30$ nm for Ni, Cr, Cu). During collisions between electrons, their distribution function will change more strongly near the Fermi level. A fraction of the excited nonequilibrium charge carriers in a metal will penetrate into a semiconductor above (or through) the Schottky barrier. The density of the injected electrons is proportional to the density of states in the metal at the Schottky barrier height. The injection time is restricted by the electron-electron relaxation time. The appearance of free charge carriers in the semiconductor will result, in turn, in a change in its dielectric constant.

Our analysis showed [10, 11] that the electron structure of metals determines variations in the photoinduced responses of structures based on these metals. The response of structures containing Ni and Cu is determined for times shorter than 1 ps by the response from a thin metal layer. This response is more pronounced for Cu, and its spectral dependence is determined by the presence of the allowed dipole interband transitions in the vicinity of the Fermi level in the spectral region under study. The response of the Cr-based structures is mainly determined by a change in the dielectric constant of the semiconductor, irrespective of the excitation type — above or below its energy gap. This is explained by the absence of transitions to the vicinity of the Fermi level in Cr in the spectral region under study. At the same time, Cr, in contrast to Ni and Cu, is an efficient injector of excited charge carriers, because the density of states sharply increases with increasing $|E - E_F|$. The injection time of charge carriers into a semiconductor can be estimated as $l_{ex}/v_F \sim 20$ fs. This estimate is in good agreement with experimental data.

Excitation above the energy gap. Upon excitation above the energy gap of a semiconductor, a laser pulse is in fact completely absorbed in a thin near-surface layer of the semiconductor [the thickness of this layer in ZnS excited by the laser pulse $\hbar\omega_{pu3}$ is $l_{ex} = 2/\alpha_{ZnS}(\hbar\omega_{pu3}) \approx 40$ nm] and produces an inhomogeneous distribution of hot carriers in the semiconductor.

Upon such excitation, the main contribution over a time shorter than 1 ps is determined by one-photon absorption [9]. The nonequilibrium electrons excited in a thin near-surface layer of the semiconductor will relax due to the electron – phonon and electron – electron interaction and will penetrate into the unexcited part of the semiconductor. The time evolution of the dielectric constant of the semiconductor is determined by these processes.

4. Conclusions

We have studied excitation and relaxation of eigenmodes in a semiconductor microcavity on a metal substrate. It is shown that femtosecond laser pulses allow us to control selectively boundary conditions in a microcavity and modify the time, spectral, and spatial parameters of the nanostructures under study.

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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