

nonlinear optics has led to the observation of various multiphoton processes. But a number of difficulties were reported, along with the obvious advances, even in the very first theoretical studies of higher-order nonlinear processes by the standard methods of perturbation theory. Foremost among these is failure of the expansion of the transition amplitudes in powers of the intensity  $F = F_0 \cos \omega t$  of the electromagnetic (EM) field at characteristic interaction energies comparable in magnitude with the energy of the localized electron. Various difficulties arise in study of all of the possible resonance effects, whose description requires summation of a class of diagrams in perturbation theory that correspond to reradiation of photons. Nor is the probability of the  $n$ -photon process always expressed in this case simply in terms of the  $n$ -th-order correlation function of the EM field amplitude; it begins to depend on all higher-order correlation functions.

If, as was first noted by Keldysh<sup>[1]</sup>, the energy spectrum of the localized electron contains a group of crowded or degenerate states, these states may be strongly perturbed by the EM field, and the contribution of this perturbation may be decisive in some cases.

Here we shall set forth results obtained in the Physical Kinetics Laboratory of the Moldavian Academy of Sciences Institute of Applied Physics in studies of absorption, emission, and scattering of light by quantum systems with degenerate multiplets in a strong EM field. The formalism of Green's functions in nonstationary perturbation theory was used for consistent description of these processes. Dipole interaction terms belonging to degenerate levels were included in the zeroth approximation<sup>[2]</sup>. Equations of the Dyson type were derived for the Green's functions and served as a base for the analysis. The following results were obtained. The emission spectrum for transitions, e.g., from a doubly degenerate multiplet to a nondegenerate ground level is a superposition of harmonics (photon replicas)<sup>[3]</sup>, a result of the quasienergy structure of the localized-electron spectrum<sup>[4]</sup>. The intensity of the  $m$ -th harmonic is determined by the formula

$$W_{ig}^{(m)} = \frac{4}{3} \frac{e_0^2 (\Omega_0 + m\omega)^2}{\hbar c^3} r_{ig}^2 J_m^2(\rho_1); \quad (1)$$

here  $J_m(x)$  is a Bessel function of real argument,  $\rho_1 = 3e_0 F_0 a_0 / \hbar \omega$ ,  $\omega$  is the frequency of the laser radiation,  $\Omega_0$  is the frequency of the electron transition, and  $a_0$  is the Bohr radius. It follows from (1) that the luminescence spectrum of the system is significantly realigned and that suppression of the fundamental luminescence line may occur as a result of rising of harmonics. The strength and shape of the absorption and luminescence lines were also investigated as functions of the coherent properties of the radiation. In the case of  $\delta$ -function<sup>[5]</sup> and Gaussian (G)<sup>[5]</sup> laser-radiation sources, a variety of information can be obtained, depending on the relation between the spectral width  $\Delta\Omega$  of the absorbed light and the width  $\gamma$  and shift  $\delta\epsilon_F$  of the atomic line. When

$$\max(\gamma, \delta\epsilon_F) \ll \Delta\Omega \ll \omega \quad (2)$$

it was found<sup>[3]</sup> that

$$\chi_m = \frac{W_{ig}^{(m)}(G)}{W_{ig}^{(m)}(\delta)} = \frac{e^{-\frac{\rho_1^2}{2}} I_m\left(\frac{\rho_1^2}{2}\right)}{J_m^2(\rho_1)}, \quad (3)$$

where  $I_m(x)$  is a modified Bessel function and  $\chi_m = m!$  when  $\rho_1 \ll 1$ . With increasing intensity  $F_0$  of the EM

field ( $\rho_1 \geq 1$ ),  $\chi_m$  becomes less dependent on  $F_0$  ( $1 < \chi_m \ll \delta\epsilon_F$ ), the absorption-line shapes are found to differ substantially for  $\delta$  and G radiation: the Lorentz shape of the photon replicas for  $\delta$  radiation is replaced by a one-sided, sharply asymmetrical broadened contour in the case of G radiation. The cross section for resonant scattering of light by atoms and local centers (LC) of crystals<sup>[6]</sup> was also calculated with consideration of the quasienergy structure of the spectrum. New resonances were observed in the cross section for scattering on photon replicas. The essential dependence of the cross section on the coherent properties of the EM field was established. A property of scattering on LC was the appearance of a vibrational structure that can be transferred by photon replicas from one region of the optical spectrum to another. Experiments to observe photon replicas in luminescence spectra were set up at the Moldavian Academy of Sciences Institute of Applied Physics<sup>[7]</sup>. Both spontaneous and induced luminescence on an antistokes photon replica was observed in an impurity crystal of n-InP.

Another trend has been theoretical investigation of resonant shifts (splittings) of atomic multiplets that occur when the frequency of a strong EM field is resonant with the natural frequencies of the atom. The cross sections for light scattering and multiphoton ionization were calculated for this case. These results made it possible to obtain additional information both on the spectrum of the localized electron in a strong EM field and on the laser radiation itself. Finally, the influence of a strong EM field on multiphoton nonradiative tunneling transitions at LC of crystals was studied<sup>[8]</sup>. The photon replicas of adiabatic potentials affect the conditions under which they intersect and cause a sharp increase in the transition probability.

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**S. A. Moskalenko.** Collective Properties of Excitons and Biexcitons. The discovery of powerful light sources opened the way to a new phase in the study of excitons. Using lasers, it has been possible to produce high exciton concentrations in crystals. The interexciton interaction becomes substantial at low temperatures, and qualitatively new, collective exciton properties emerge.

Depending on the nature of the interaction and on the temperature, we may expect: 1) scattering of excitons by one another<sup>[1]</sup>; 2) formation of biexcitons<sup>[1-3]</sup>; 3) Bose-Einstein condensation of excitons or biexcitons<sup>[1,4-8]</sup>; 4) condensation of excitons into metallic drops consisting of nonequilibrium electrons and holes<sup>[9]</sup>. The first three effects are discussed below.

The mechanism of radiative Auger recombination of two free excitons<sup>[5]</sup> is used to explain the new luminescence band observed experimentally in<sup>[10]</sup>. The shape of the band was calculated in<sup>[11]</sup> on the assumption that one large-radius exciton is annihilated, yielding part of its energy for dissociation of the other exciton. The theory agrees well with experiment only in the region of the longwave tail of the band. The shortwave tail is probably due to interserial excitation of the second exciton.

A computer was used in<sup>[12]</sup> to obtain the energy of the ground state of the biexciton from the Hamiltonian of the electrons and holes. An analytical relation for the biexciton energy levels as functions of the quantum numbers can be obtained only for a definite model. It is assumed in<sup>[13]</sup> that the binding energy in the exciton is much larger than the binding energy of the biexciton. It then becomes possible to introduce an interaction potential of two excitons that depends on the distance between their centers of gravity. It was taken in the form of a Morse potential whose parameters depend on the mass ratio  $\sigma$  of the electron and hole and are so selected that the values of the biexciton's ground-state energy would agree with the calculation of<sup>[12]</sup>. It was used to find the excited levels of the biexciton as functions of  $\sigma$ , vibrational quantum numbers  $n = 1, 2$ , and 3, and rotational quantum numbers  $l = 1$  and 2. The first vibrational level exists for  $\sigma \leq 0.1$ , the second for  $\sigma \leq 0.03$ , etc.

The resulting energy spectrum makes it possible to propose a new mechanism for the radiative recombination of two biexcitons, one in which the second biexciton remains in the excited state. The luminescence band  $N_1$  of the CuCl crystal<sup>[14]</sup> may be due to this transition if its intensity is proportional to the square of the intensity of the M band.

Saturation of the attractive forces between excitons on formation of biexcitons may result in predominance of repulsion between biexcitons and hence a possibility of their Bose condensation<sup>[4]</sup>.

Kuroda, Shionoya, Saito, and Hanamura observed a narrow emission line in the luminescence spectrum of a CdSe crystal against the background of the M band<sup>[15]</sup>. Its width is less than 0.17 meV, and it is observed when the pump power lies in a certain range. These facts suggested that it might be related to the appearance of Bose-condensed biexcitons. If we follow<sup>[16]</sup>, the expected width of the narrow luminescence line is of the order of 0.01 meV. Bose condensation of excitons in CdSe was reported on earlier in<sup>[17]</sup>. Further experimental studies in this area are absolutely necessary.

In a system of dipole-active excitons and photons, the energy spectrum depends strongly on the relation between the frequency  $\nu_{\text{ex-ex}}$  of collisions between excitons and the frequency  $\nu_{\text{ex-ph}}$  of exciton-to-photon transformations. If  $\nu_{\text{ex-ex}} > \nu_{\text{ex-ph}}$  and repulsion between excitons (e.g., singlet excitons) predominates at low temperatures, when scattering by phonons is in-

significant, a Bose condensate of excitons appears. It was shown in<sup>[1,6-8,16]</sup> that a Bose condensate of excitons with a wave vector  $k \neq 0$  may generate a coherent electromagnetic wave with macroscopic amplitude and the same wave vector  $k$ . If the total number of excitons and photons is preserved, a single oscillation is established in the system with participation of macroscopic numbers of photons and excitons. There are two possible values of the vibration frequency  $\omega(k)$  of a macroscopically filled mode for each value of the wave vector  $k$ , but they occur only one at a time. The two branches of values of  $\omega(k)$  resemble the polariton spectrum<sup>[1,6]</sup>. If the Bose condensation of the excitons was to a state with  $k = 0$ , the excitons can be transformed into light, simultaneously emitting either acoustic quanta in the system of the excitons themselves or lattice phonons. This indicates the existence of instability in the system and the possibility of designing a combined degenerate-exciton laser and phaser.

The coherent properties of excitons and photons may be inhomogeneous in space. Known examples of such states include quantum vortices in liquid helium and the self-focusing of laser radiation. Being approximately equivalent to bosons, excitons may also form quantum vortices<sup>[1,4]</sup>, and, in virtue of their ability to undergo transformation to light, may produce an inhomogeneous distribution of the radiation density in the plane perpendicular to the vortex axis. In particular, if a vortex is formed by longitudinal excitons, we shall see a light filament having the frequency of the longitudinal exciton and concentrated for the most part in the region of the core of the vortex. Its intensity and lifetime will be determined by the exciton concentration and lifetime.

There are also other aspects to the interaction of excitons with laser radiation:

1) The interaction of excitons with resonant laser radiation during time intervals smaller than the relaxation time in the system. Experimental conditions for observation of such phenomena have now been developed. Thus, for example, the frequency of an argon laser is resonant with the energy of formation of the exciton level  $A_1$  in the  $\text{CdSe}_x\text{S}_{1-x}$  crystal at  $x = 0.005$ <sup>[18]</sup>.

2) Self-induction of transparency<sup>[19]</sup> on excitons.

3) Creation of a superradiant state in a high-density exciton system as a result of intraserial excitation of excitons, and observation of radiation echo<sup>[1,20]</sup>.

4) Intense electromagnetic radiation is capable not only of mixing exciton states and producing effects of the double optical resonance type<sup>[21]</sup>, but also of affecting the structure of the individual exciton in much the same way as a strong constant magnetic field that predominates over the Coulomb electron-hole interaction causes the formation of diamagnetic excitons<sup>[22]</sup>. Under these conditions, excitons of high density exhibit qualitatively different properties.

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**D. V. Gitsu.** Features of Transfer Phenomena in Crystals of the Bismuth Type. The results of a coordinated investigation of transfer phenomena in bismuth and bismuth-antimony alloys doped with donor (tellurium, selenium) and acceptor (tin, lead) impurities<sup>[1-8]</sup> are discussed. The limitations imposed by the point group of the crystal on the dynamics of development of the anisotropy of galvanothermomagnetic effects in arbitrary nonquantizing magnetic fields are analyzed on the basis of a polynomial representation of the kinetic coefficients<sup>[9,10]</sup>.

Analysis of the expressions for the kinetic coefficients that were obtained in the relaxation-time approximation for various localizations of the vital extrema<sup>[11]</sup> indicates that the structure of the polynomial coefficients of the generalized resistivity tensor is determined by the number of vital extremes and by the orientation of the isoenergetic surfaces corresponding to them, while the structure of the coefficients of the generalized Seebeck tensor also depends on the dispersion law and relaxation mechanism. If only diffusion processes in the principal crystallographic directions are considered, the most important existence condition for odd terms of the magnetothermal emf reduces to

the following: there exist several groups of nonequivalent vital energy extremes, and the principal axes of the effective-mass tensor of at least one of the groups do not coincide with the principal crystallographic directions.

In the experimental part of the study, attention is concentrated on the dynamics of development of the anisotropy of the galvanothermomagnetic effects in magnetic fields up to 4 T.

It is shown that in pure bismuth, the experimental angle diagrams are accurately reproduced theoretically within the framework of a two-band model (the L and T extremes are vital)<sup>[12]</sup>. The carrier concentration varies with temperature as  $T^{3/2}$ , and electron mobility nearly in accordance with a  $T^{-5/2}$  law. The commutation effect of the magnetothermal emf at  $T > 77^\circ\text{K}$  is an effect of third order in the magnetic field in all crystallographic directions, a direct demonstration of the absence of phonon dragging of carriers. The results of a study of bismuth doped with donor impurities indicate that the dispersion in the conduction band is approximated more accurately by a nonellipsoidal nonparabolic model, and that the energy gap at point L increases with rising temperature. It was shown that the increase of the Hall effect with increasing field that is observed in certain crystallographic directions is due to strong anisotropy of electron mobility. In all alloys of this type, magnetoresistance is not saturated in strong fields, but increases almost linearly with the field; this is explained by a kind of inelastic scattering of electrons by quasilocal impurity states.

It follows from the aggregate of the data for alloys doped with acceptor impurities that the parabolicity of the principal valence band of bismuth persists over a broad energy range, and that no topological features are observed between the bottom of the conduction band and the L maxima<sup>[13]</sup>. Participation of L holes in transfer phenomena is registered reliably in strongly doped alloys from the increase in the anisotropy of the thermal emf and magnetoresistance and through the secondary appearance of a commutation effect of the magnetothermal emf. The observed uncommonly strong variation of the thermal emf with the magnetic field in weakly doped alloys of this type is explained by the fact that the asymptotic value of the emf is inversely proportional in strong fields to the electron-hole concentration difference. The basic kinetic parameters of L holes were determined. It was clearly shown that the effects of tin and lead impurities in bismuth and bismuth-antimony alloys differ not only quantitatively, but in certain cases even qualitatively.

In  $\text{Bi}_{1-x} - \text{Sb}_x$  alloys, all measured quantities vary nonmonotonically with composition. These results clearly indicate that the energy spectra of these alloys vary substantially<sup>[14]</sup> over the entire concentration range. Here there are multiple changes in the vital character of the energy extrema, and this results in changes of varying nature in the anisotropy of the galvanothermomagnetic effects in the various concentration ranges. Four such ranges can be distinguished near  $77^\circ\text{K}$ :  $0 \leq x < 0.07$ ,  $0.1 < x < 0.25$ ,  $0.3 < x < 0.65$ , and  $0.75 < x \leq 1$ , together with the transitional ranges corresponding to them. It was shown that considerable realignment of the spectrum occurs on variation of the temperature. As a result, for example, inversion of the L bands<sup>[14]</sup> occurs at  $77^\circ\text{K}$  at compositions with  $x \approx 0.2$ .