center. Results obtained for molecular systems[1,2] with the aid of computers cannot be extended to local states of crystals because of the dispersion of the crystal vibrations that results from the strong dependence of the optical-spectrum parameters on the phonon-density distribution. By calculating the moments of the spectrum and their variations in dichroism effects, we can obtain information on the electron states and the electron-phonon interaction without resorting to a detailed calculation of the band contour.

Let us consider a transition from an orbital singlet initial state A to a multiplet Γ that is split by a perturbation **W** operating in the electronic subsystem. For the n-th central moment of the optical distribution we obtain the matrix expression

$$\sigma_n = (-1)^n \frac{d^n}{dt^n} \langle e^{-iWt} U(t) \rangle_{ph} \Big|_{t=0}, \quad (1)$$

where $\langle \ldots \rangle_{ph}$ represents statistical averaging over the vibrational subsystem,

$$U(t) = T \exp \left\{ \frac{i}{\hbar} \int_0^t [H_{eL}(t_1) + W(t_1)]\, dt_1 \right\} \quad (2)$$

is the evolution operator, and

$$H_{eL} = \sum_{\kappa} v_{\kappa} q_{\kappa} \quad (3)$$

is the operator of the electron-phonon interaction in the approximation linear in the normal coordinates q_K of the crystal vibrations. The matrix operators in formulas (1)—(3) are defined in the basis that diagonalizes the perturbation **W**. The moments (1) are reckoned from the center of gravity of the distribution

$$\bar{\Omega} = \omega_{\Gamma A} \mathbf{1} + \hbar^{-1} W, \quad (4)$$

where $\omega_{\Gamma A}$ is the Franck-Condon frequency in the absence of the perturbation. We obtain for the second and third moments

$$\sigma_2 = \frac{1}{2\hbar^2} \sum_{\kappa} v_{\kappa}^+ v_{\kappa} \operatorname{cth} \frac{\beta_{\kappa}}{2}, \quad (5)$$

$$\sigma_3 = \frac{1}{2\hbar^2} \sum_{\kappa} \left[\omega_{\kappa} v_{\kappa}^+ v_{\kappa} + \sum_{\kappa} v_{\kappa}^+ [W, v_{\kappa}] \operatorname{cth} \frac{\beta_{\kappa}}{2} \right] = \sigma_3' + \sigma_3'' \quad \left(\beta_{\kappa} = \frac{\hbar\omega_{\kappa}}{kT} \right). \quad (6)$$

It follows from (5) and (6) that σ_2 and σ_3' are invariants of the point group of the local center, so that the splitting of the band that is observed in dichroism effects is not accompanied by a change in the second moments of its components. The latter are therefore equal to the second moment of the unsplit band. At the same time, the third moment contains a correction σ_3'' proportional to the matrix **W**. The only contribution to the proportionality coefficient comes from vibrations that are active in the dynamic Jahn-Teller effect (JTE) and have $[\mathbf{W} \times \mathbf{v}] \neq 0$. Under the conditions of the JTE, therefore, an external field not only shifts the band, but also deforms it, so that the hypothesis of the so-called "rigid shift" becomes inapplicable.

The change in the third moment in external fields can be used to extract information on the parmaeters of the Jahn-Teller interaction. Let us demonstrate this for the case of a transition of the A → T type to an orbital triplet in a cubic center when the modes $\overline{\Gamma} = E$, T_2 are active in the JTE. In this case, the matrix $\sigma_3''(\text{trig})$, which corresponds to uniaxial compression along the axis C_3, takes the form

$$\sigma_{3\,(\text{trig})}'' = W_{\text{trig}} \left[-\frac{1}{2} \sigma_2(T_2) - \frac{3}{2} \sigma_2(E) \right], \quad (7)$$

where $\sigma_2(\overline{\Gamma})$ are the contributions of the respective modes to σ_2. For compression along the C_4 axis we obtain

$$\sigma_{3\,(\text{tetr})}'' = W_{\text{tetr}} \left[-\frac{3}{2} \sigma_2(T_2) \right]. \quad (8)$$

The matrices $\mathbf{W}_{\text{trig(tetr)}}$ can be determined from (4) for an observed change $\bar{\Omega}$ on transition from σ- to π-polarized light. Then Eqs. (7) and (8) are used to determine $\sigma_2(E)$ and $\sigma_2(T_2)$ from the measured values of σ_3'', i.e., to find the contributions of the Jahn-Teller vibrations to the second moment:

$$\sigma_2 = \sigma_2(A_1) + \sigma_2(E) + \sigma_2(T_2). \quad (9)$$

Measuring the total second moment σ_2, we also determine $\sigma_2(A_1)$. These results apply for arbitrary Jahn-Teller coupling and are equally valid for both local and crystal vibrations. Reference[3] gives a specific calculation of $\sigma_2(\overline{\Gamma})$ for the U band of ruby ($^4A_{2g} \rightarrow {}^4T_{2g}$ transition in the Cr^{3+} ion).

If the dynamic JTE is relatively small, the band retains its near-Gaussian bell shape on strong electron-phonon interaction. Using the well-known Edgeworth expansion for the band maximum, we obtain the formula

$$\Omega_{\max} = \bar{\Omega} - \frac{\sigma_3}{2\sigma_2}. \quad (10)$$

Accordingly, we have for trigonal dichroism

$$\Omega_{\max}(\pi) - \Omega_{\max}(\sigma) = W_{\text{trig}} \left[1 + \frac{3\sigma_2(E) + \sigma_2(T_2)}{4\sigma_2} \right]. \quad (11)$$

The second term of (11) is a small correction to the frequency of the band maximum, but in calculation of the shift of the maximum it yields a quantity of the same order as the change in the first moment. Relation (11) must be taken into account in determining the parameters of crystal fields of low symmetry from the dichroism of the band maxima. The multiplier in the square brackets is different for different A → T bands (for example, the U and Y bands in ruby), together with the multiplier $\mathbf{W}_{\text{trig}}$ (which is also, generally speaking, different for different bands). It is therefore more reliable to determine $\mathbf{W}_{\text{trig}}$ from the shift of the first moment instead of from the shift of the maximum. Estimates for ruby indicate that the multiplier in the square brackets of (11) equals 1.27 and 1.5 for the U and Y bands, respectively[3].

A detailed exposition will be submitted in the author's monograph "Effekty élektronno-kolebatel'nogo vzaimodeĭstviya v opticheskikh spektrakh primesnykh paramagnitnykh ionov" (Electron-Vibrational Interaction Effects in the Optical Spectra of Impurity Paramagnetic Ions), which will be publsihed in 1974 by "Shtiintsa" (Kishinev).

*$[W, v_{\mathcal{H}}] = W \times v_{\mathcal{H}}$.

[1] R. Englman, M. Caner, S. Toaff, J. Phys. Soc. Japan **29**, 306 (1970).
[2] M. C. M. O'Brien, J. Phys. **C5**, 2045 (1972).
[3] Yu. E. Perlin, L. S. Kharchenko, and B. S. Tsukerblat, Fiz. Tverd. Tela **11**, 1065 (1969) [Sov. Phys.-Solid State **11**, 873 (1969)].

V. A. Kovarskiĭ. Features of the Absorption, Emission, and Scattering of Light by Atoms and Impurity Centers of Crystals with Degenerate Energy Spectra in a Strong Electromagnetic Field. The development of

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 localiced 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 depe n on all higher-order correlation functions.

If, as was first noted by Keldysh[1], the energy spectrum of the localized electron contaisn 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[2]. 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)[3], a result of the quasienergy structure of the localized-electron spectrum[4]. The intensity of the m-th harmonic is determined by the formula

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

here $J_m(x)$ is a Bessel function of real argument, $\rho_i = 3e_0 F_0 a_0/\hbar\omega$, ω is the frequency of the laser radiation, Ω_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 δ-function[5] and Gaussian (G)[5] 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 γ and shift $\delta\epsilon_F$ of the atomic line. When

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

it was found[3] that

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

where $I_m(x)$ is a modified Bessel function and $\chi_m = m!$ when $\rho_i \ll 1$. With increasing intensity F_0 of the EM field ($\rho_i \gtrsim 1$), χ_m becomes less dependent on F_0 ($1 < \chi_m$ $\delta\epsilon_F$), the absorption-line shapes are found to differ substantially for δ and G radiation: the Lorentz shape of the photon replicas for δ 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[6] was also calculated with consieeration 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[7]. 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 nonardiative tunnelling transitions at LC of crystals was studied[8]. The photon replicas of adiabatic potentials affect the conditions under which they intersect and cause a sharp increase in the transition probability.

[1]L. V. Keldysh, Zh. Eksp. Teor. Fiz. 47, 1945 (1964) [Sov. Phys.-JETP 20, 1307 (1965)].
[2]V. A. Kovarskiĭ, N. F. Perel'man, and A. V. Belousov, Paper at Conference on the Interaction of Electrons with Strong Electromagnetic Fiels (Balatonfured, Spptember 11—16, 1972). Invited Papers, Budapest, 1973, p. 125.
[3]V. A. Kovarskiĭ, Zh. Eksp. Teor. Fiz. 57, 1217 (1969) [Sov. Phys.-JETP 30, 663 (1970)].
[4]Ya. B. Zel'dovich, ibid. 51, 1492 (1966) [24, 1006 (1967)]; V. I. Ritus, ibid., p. 1544 [(1041)].
[5]R. Glauber, Transl. in: "Kvantovaya optika i kvantovaya radiofizika" (Quantum Optics and Quantum Radiophysics), Mir, Moscow, 1967.
[6]V. A. Kovarskiĭ and N. F. Perel'man, Zh. Eksp. Teor. Fiz. 60, 509 (1971) [Sov. Phys.-JETP 33, 274 (1971)]; Fiz. Tverd. Tela 13, 1888 (1971) [Sov. Phys.-Solid State 13, 1582 (1972)].
[7]V. A. Kovarskiĭ and N. A. Ferdman, ZhETF Pis. Red. 15, 483 (1972) [JETP Lett. 15, 342 (1972)].
[8]V. A. Kovarskiĭ, N. F. Perel'man, and É. P. Sinyavskiĭ, Fiz. Tverd. Tela 15, 1809 (1973) [Sov. Phys.-Solid State 15, 1207 (1973)].

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.