MULTIPHOTON LUMINESCENCE SATELLITES OF AN IMPURITY CRYSTAL IN A STRONG ELECTROMAGNETIC FIELD

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One of us considered in [1] the singularities of optical transitions to degenerate levels of the hydrogen atom in the presence of powerful nonresonant laser emission. Under these conditions it was predicted that the spontaneous luminescence at the frequency of the transition of the electron from the degenerate level to the ground level would be weakened ("suppressed") as a result of a redistribution of the intensity in the frequency spectrum of the luminescence. Since the hydrogen-like model of local centers is well realized in a number of semiconductors, it is of interest to generalize the theory to include this case. If the ionization energy of the local center is $\varepsilon_{1c} << \hbar\omega$ (ω is the emission frequency of the external laser), then the energy levels of the local center can be regarded approximately as almost degenerate [2, 3], and the results of [1] can be directly used for the subsequent analysis.

It is assumed that the average electron kinetic energy $\varepsilon_F^c = e_0^2F_0^2/4\mu_c\omega^2$ in the field of an electromagnetic wave of intensity $F = F_0 \cos \omega t$ in the conduction band of a semiconductor and the hole energy $\varepsilon_F^v = e_0^2F_0^2/4\mu_v\omega^2$ in the valence band satisfy the inequality $\varepsilon_F^c << \varepsilon_D$ and $\varepsilon_F^v << \varepsilon_A$, where ε_D and ε_A are the donor and acceptor ionization energies, respectively. For example, for $F_0 = 10^6$ V/cm, $\hbar\omega = 1.17$ eV, $\mu_c = 0.1$ m we have $\varepsilon_F \sim 10^{-3}$ eV, and $\varepsilon_D \sim 10^{-2}$ eV. For acceptors in III-V semiconductors, these conditions are even better satisfied. Since $\varepsilon_{1c} << \hbar\omega$ (the antiadiabatic case), the levels of the local center do not decay in practice by tunneling [4].

Let us write out the final formulas for the probability of spontaneous luminescence $W(\Omega)$ when an electron goes from a donor level into the valence band (DV transition) and separately for the transition of an electron from the conduction to the acceptor level (CA transition):

$$W_{DV}(\Omega)d\Omega = \sum_k B_k^{(DV)} \sum_n J_n^2(\rho_D)\delta\{\epsilon_v(k) + \Delta_{cv} - \epsilon_D - \hbar\Omega - n\hbar\omega\}\rho_\Omega d\Omega, \tag{1}$$

$$W_{CA}(\Omega)d\Omega = \sum_k B_k^{(CA)} \sum_n J_n^2(\rho_A)\delta\{\epsilon_c(k) + \Delta_{cv} - \epsilon_A - \hbar\Omega - n\hbar\omega\}\rho_\Omega d\Omega. \tag{2}$$

Here $J_n(x)$ is a Bessel function of real argument. $B_k^{(DV)}$ and $B_k^{(CA)}$ are constants that depend on the matrix elements of the dipole transition from the band to the impurity center:

$$\epsilon_{cv}(k) = \frac{\hbar^2k^2}{2\mu_{cv}}; \quad \hbar\omega\rho_{D,A} = e_0F_0 < \psi_{1s}^{D,A} | Z | \psi_{2p}^{D,A} > = 1{,}3e_0F_0a_B^{D,A}$$

ρ_Ω is the spectral density of the radiation; $a_B^{D,A}$ is the radius of the Bohr orbit of the donor or acceptor. $F_0 = [(n_1^2 + 2)/3]F_{laser}$; n_1 is the refractive index.

As follows from (1) and (2), in a strong electromagnetic field the spontaneous-luminescence spectrum of the impurity crystal is a superposition of bands whose maxima are separated by an amount equal to the laser quantum $\hbar\omega$, and the intensity of the n-th band is determined by the weight function

$J_n^2(\rho_{D,A})$. At $\rho_D \ll 1$ (or $\rho_A \ll 1$), the main contribution is made by the term with $n = 0$, and formulas (1) and (2) go over into the usual formulas for the probability of the spontaneous luminescence. In cases when ρ_A (or ρ_D) is larger than unity, the main contribution is made by the photon satellite with $n \sim \rho_D$ (or $n \sim \rho_A$). It should be noted that the "anti-Stokes" photon satellites with energies larger than $|\Delta_{cv} - \varepsilon_D|$ (or $|\Delta_{cv} - \varepsilon_A|$) are not limited in number, whereas the "Stokes" satellites are limited, as follows directly from formulas (1) and (2).

Observation of the photon replicas is of particular interest in connection with the possibility of experimentally observing the harmonics of quasi-energies of the local center in a strong electromagnetic field [5]. Since $\rho_{lc} \sim 1$ is reached at fields $F_0 \sim 10^6$ V/cm, $\hbar\omega = 1.17$ eV, and $a_B^{lc} \sim 10^{-6}$ cm, the organization of a suitable experiment for the observation of the first "anti-Stokes" photon satellite in the spectrum of the spontaneous luminescence with frequency

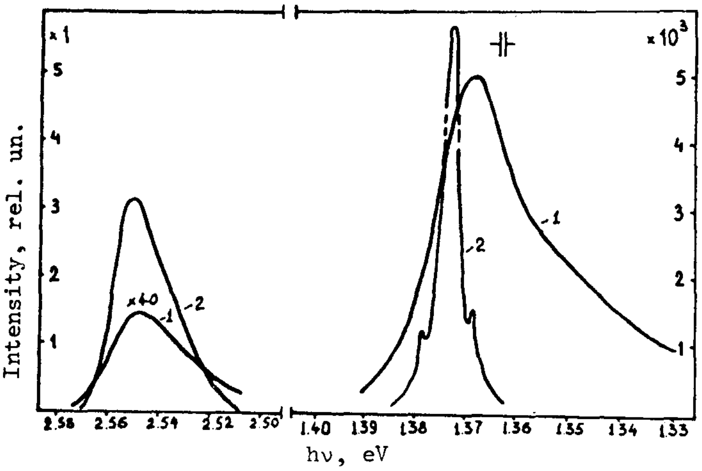
$$\hbar\Omega_1^+ = \Delta_{cv} - \epsilon_{lc} + \hbar\omega. \qquad (3)$$

is assumed to be perfectly realistic.

We chose the semiconducting crystals ZnTe ($\Delta_{cv}^{77°K} \simeq 2.2$ eV), CdSe ($\Delta_{cv}^{77°K} \simeq 1.8$ eV), and InP ($\Delta_{cv}^{77°K} \simeq 1.4$ eV), which contain hydrogen-like local centers with $\varepsilon_{lc} \approx 0.05$ eV. The luminescence was excited by volume two-photon pumping with a W-switched neodymium laser (pulse energy ∿1 J, duration at half-width ∿40 nsec). The luminescence spectra were registered with an MDR-2 high-transmission monochromator, with photomultipliers, and with a high-speed long-persistence oscilloscope.

Inasmuch as in all the chosen samples the excited carrier had time to go over to shallow local levels during the duration of the laser pulse, the luminescence spectra were recorded both as DV transitions and as CA transitions, i.e., transitions with frequencies lower than the width of the forbidden band by an amount $\sim\varepsilon_{lc}$ were realized.

Let us stop to discuss the indium phosphide crystal in greater detail. It is known [6] that a CA-transition line at a frequency 1.37 eV ($\varepsilon_A \sim 0.045$ eV)



Luminescence spectra of indium phosphide under two-photon excitation for two values of the neodymium-laser intensity: curve 1 - intensity 4×10^{25} kV/cm^2-sec; curve 2 - intensity 6.5×10^{25} kV/cm^{2}sec, T = 77°K.

is observed in the single-photon excitation spectrum of this crystal at 77°K. Under two-photon excitation, this line was distinctly recorded in the luminescence spectra with short kinetics (on the order of the laser-pulse duration). The expected "anti-Stokes" photon satellite was recorded at a frequency $\hbar\Omega_1^+ = 1.37 + 1.17$ eV (see the figure). Obviously, luminescence at the frequency Ω_1^+ is absorbed to a considerable degree in the volume of the crystal, so that it was registered in essence only from the surface layers. (The last fact is precisely evidence of the high intensity of the Ω_1^+ luminescence in the volume.) The kinetics of the relaxation of the photon satellite followed strictly the laser pulse. It is precisely this circumstance which made it possible to use a neodymium laser both as the source of carrier pumping (with sufficiently rapid relaxation of the carriers to the local centers, $\sim 10^{-11}$ sec), and simultaneously as a source controlling the elementary luminescence act. With further increase of the neodymium-laser power, the photon satellite increased sharply in intensity. When the intensity of the exciting laser radiation was changed by a factor ~ 1.6, the intensity of the satellite increased 80 times! Such a rapid growth of the photon satellite intensity is possibly connected with singularities in the indium-phosphide material, in which at these intensities we observed a narrowing of the fundamental luminescence line, thus evidencing that stimulated transitions make a contribution. With increasing temperature $T > 77$°K, the intensity of the Ω_{CA} luminescence, and accordingly the intensity of the photon satellite Ω_1^+, has decreased strongly, this being connected with the thermal ionization of the local centers A.

The photon satellites Ω_1^+ were also observed in zinc telluride and cadmium selenide crystals at frequencies ~ 360 and ~ 428 nm respectively. In these crystals, however, the main luminescence band is sufficiently broad and the photon satellite has a similar structure.

We note in conclusion that the appearance of the frequency Ω_1^+ in the luminescence spectrum might be connected also with the superposition addition of the laser frequency ω and the frequency Ω_{CA} of the CA luminescence at the local centers of the crystal. However, the probability of such a process, which is a process of higher order than that considered above (formulas (1) and (2)), is proportional to the intensity of the Ω_{CA} liminescence and consequently is much lower.

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