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Abstract

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PHYSICS

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A UNIVERSAL RELATION BETWEEN THE ABSORPTION AND LUMINESCENCE SPECTRA IN SEMICONDUCTORS AT HIGH PUMPING INTENSITIES

In work ([1]) a universal relation was derived between the absorption and luminescence spectra of complex molecules. Later, a number of considerations were put forward that made it possible to generalize it to the case of high pumpings, which cause depletion of the lower electronic state and the appearance of an inverted population of levels ([2–4]). In the present work a derivation of an analogous relation for semiconductors is given. Some aspects of this problem were discussed in ([5],[6]).

The universal relation between the absorption and luminescence spectra, together with the energy-balance equations, significantly expands the possibilities for experimental investigation of the properties of semiconductors. It is valid for an equilibrium distribution of electrons in the conduction band and on the levels adjacent to it, and also for an equilibrium distribution of holes in the valence band and on the adjacent levels. In this case the absorption coefficient and the power of spontaneous emission at frequency ν can be represented in the form ([6])

$$k(\nu) = \frac{h\nu}{v_g}\left[B'_{12}(\nu) - B'_{21}(\nu)\right], \tag{1}$$

$$W(\nu) = h\nu \int g(E^*)f(E^*)A(\nu, E, E^*)g(E)[1 - f(E)]\,dE, \tag{2}$$

where

$$B'_{12}(\nu) = \int g(E)f(E)B(\nu, E, E^*)g(E^*)[1 - f(E^*)]\,dE,$$

$$B'_{21}(\nu) = \int g(E^*)f(E^*)B(\nu, E, E^*)g(E)[1 - f(E)]\,dE; \tag{3}$$

$f(E)$ and $f(E^*)$ are the Fermi-Dirac functions determining the distribution of electrons over the energy levels of the valence band E and the conduction band E^*; $g(E)$ and $g(E^*)$ are the densities of states; $E^* - E = h\nu$; v_g is the group velocity of light. The quantities $A(\nu, E, E^*)$ and $B(\nu, E, E^*)$ are analogous to the Einstein coefficients for spontaneous and induced transitions, with $A(\nu, E, E^*)/B(\nu, E, E^*) = 8\pi h\nu^3/v^2 v_g$, where v is the phase velocity of light. The integration in (^{2})-(^{4}) is carried out over all levels E at a given frequency ν.

Since

$$f(E)[1 - f(E^*)]/f(E^*)[1 - f(E)] = e^{(h\nu - \Delta F)/kT},$$

where $\Delta F = F^* - F$ is the difference of the Fermi quasi-levels, it follows from (1)-(3) and (2) that

$$B'_{21}(\nu) = B'_{12}(\nu) e^{-(h\nu - \Delta F)/kT}, \tag{4}$$

$$\frac{W(\nu)}{k(\nu)} = \frac{8\pi h\nu^3}{v^2} \frac{1}{e^{(h\nu - \Delta F)/kT} - 1}. \tag{5}$$

Relation (7) is universal. Since not all spontaneous emission is luminescence, but only the excess over the background of thermal emission, then, taking (5) into account,

$$W_{\text{lum}}(\nu) = W(\nu) - v_g k(\nu) u^0(\nu) = \frac{1 - e^{-\Delta F/kT}}{1 - e^{-h\nu/kT}} W(\nu), \tag{6}$$

where $u^0(\nu)$ is the Planck function. Therefore,

$$\frac{W_{\text{lum}}(\nu)}{k(\nu)} = \frac{8\pi h\nu^3}{v^2} \frac{1 - e^{-\Delta F/kT}}{1 - e^{-h\nu/kT}} \frac{1}{e^{(h\nu - \Delta F)/kT} - 1}. \tag{7}$$

Usually $h\nu \gg kT$. If, in addition, $\Delta F \gg kT$, then formula (7) becomes (5). Conversely, under thermodynamic equilibrium, when $\Delta F = 0$, relation (5) coincides with Kirchhoff' s law, and $W_{\text{lum}}(\nu)/k(\nu) = 0$. In what follows, the background of thermal radiation is not taken into account. The parameters of the semiconductor and the properties of the irradiating radiation enter into (7) only through the difference ΔF.

Formula (7) describes not only the absorption regime, but also the gain regime that arises at sufficiently high pumpings. If $h\nu > \Delta F$, then the value of $k(\nu)$ is positive; for $h\nu < \Delta F$ it is negative.

If the form of the function $k(\nu)$ is known from experiment, then relation (7) makes it possible to calculate the absolute values of the luminescence power and its spectral distribution. The value of ΔF is found directly from the frequency ν_{inv}, i.e., the frequency corresponding to the transition from gain to absorption:

$$\Delta F = h\nu_{\text{inv}}. \tag{8}$$

The study of the function $k(\nu)$ is best carried out with the aid of quantum generators based on organic dyes, which make it possible to obtain powerful light sources with a smoothly varying frequency ($^{2-4}$).

If the form of the function $k(\nu)$ is not known, it can be calculated from the photoluminescence spectrum. At the same time the difference of the Fermi levels ΔF is determined. To do this it is necessary to use relation (7) and the energy-balance equation

$$\int v_g k(\nu) u(\nu)\, d\nu + Q_{12} = \int W(\nu)\, d\nu + Q_{21}. \tag{9}$$

The first term on the left-hand side of (9) determines the power expended on excitation of electrons by the radiation flux with spectral density $u(\nu)$; the first term on the right-hand side is the power emitted in the process of luminescence. The quantities Q_{12} and Q_{21} denote the powers of the corresponding nonoptical transitions.

Substituting (9) into (7), we obtain the relation

$$\int \frac{v_g v^2}{8\pi h\nu^3} W(\nu) u(\nu) \left[e^{(h\nu-\Delta F)/kT} - 1\right] d\nu = \int W(\nu) d\nu + Q_{21} - Q_{12}, \tag{10}$$

which relates the value of $W(\nu)$ to ΔF. Under thermodynamic equilibrium $u_{\text{equil}}(\nu)$ is determined by Planck' s formula, $Q_{21} = Q_{12}$, and therefore $\Delta F = 0$. As $u(\nu)$ increases, the value of ΔF grows. If the excitation is carried out by intense monochromatic radiation of frequency ν_{H} with integral density u ($u \gg \int u_{\text{equil}}(\nu) d\nu$), and $Q_{12} = 0$, then from (10) it follows that

$$\frac{\Delta F}{kT} = \frac{h\nu_{\text{H}}}{kT} - \ln \frac{1 + \alpha(\nu_{\text{H}})u}{\alpha(\nu_{\text{H}})u}, \tag{11}$$

where

$$\alpha(\nu_{\text{H}}) = \frac{v^2 v_g}{8\pi h\nu^3} \frac{\eta W(\nu_{\text{H}})}{\int W(\nu) d\nu}, \tag{12}$$

and $\eta = \int W(\nu) d\nu / [Q_{21} + \int W(\nu) d\nu]$ is the energy yield of luminescence.

In the general case the contour of the band and the energy yield of luminescence depend on the pumping intensity, and therefore $\alpha(\nu_{\mathrm{H}})$ also depends on u. To determine $\alpha(\nu_{\mathrm{H}}, u)$, it is simplest to measure experimentally the quantity $\Delta\nu = \int W(\nu)d\nu/W(\nu_{\mathrm{M}})$, where ν_{M} is the frequency of the maximum of the luminescence band—

and $\sigma = W(\nu_{\mathrm{H}})/W(\nu_{\mathrm{M}})$. Then $a(\nu_{\mathrm{H}}, u) = u^2\nu_g\eta\sigma/8\pi h\nu^3\Delta\nu$. For parameter values corresponding to gallium arsenide, $a \sim 0.015\eta\sigma$ erg^{-1}cm^3. For complete saturation ($a(\nu_{\mathrm{H}})u \gg 1$), radiation densities of $\sim 10^3$ erg/cm^{-3} are required. Such values are attainable with solid-state quantum generators operating in the pulsed regime. For $a(\nu_{\mathrm{H}})u \gg 1$, the value of ΔF is equal to $h\nu_{\mathrm{H}}$ and, consequently, $\nu_{\mathrm{inv}} = \nu_{\mathrm{H}}$. Knowing ΔF and the shape of the luminescence band, with the aid of (7) one can readily find the shape of the gain and absorption band (the values of $k(\nu)$ in relative units). From (1) and (10) it follows that

$$k(\nu) = \frac{h\nu}{\nu_g}B'_{12}(\nu)\left[1 - e^{-(h\nu-\Delta F)/kT}\right] =$$

$$= \frac{h\nu}{\nu_g}B_{12}(\nu)\,\frac{1 + a(\nu_{\mathrm{H}}, u)u\left[1 - e^{-h(\nu-\nu_{\mathrm{H}})/kT}\right]}{1 + a(\nu_{\mathrm{H}}, u)u}. \tag{13}$$

If

$$h\nu > h\nu_{\mathrm{H}} - kT\ln\left[\frac{(1 + a(\nu_{\mathrm{H}}, u)u)}{a(\nu_{\mathrm{H}}, u)u}\right], \tag{14}$$

then the absorption coefficient $k(\nu)$ is positive; otherwise it is negative. A broad gain band can be created only at sufficiently large ν_{H} and u.

According to (11), for $a(\nu_{\mathrm{H}}, u)u \gg 1$ the distance between the quasi-Fermi levels tends to its limiting value, equal to $h\nu_{\mathrm{H}}$. This means that, at sufficiently high pump levels, the quantities $B'_{12}(\nu)$ and $a(\nu_{\mathrm{H}}, u)$ cease to depend on u. Consequently, formula (13) gives explicitly the dependence of the absorption-gain coefficients on the energy density of the incident radiation at high excitation levels. In this case the absorption coefficient at the frequency of the exciting light is equal to

$$k(\nu_{\mathrm{H}}) = \frac{h\nu_{\mathrm{H}}}{\nu_g}\,\frac{B'_{12}(\nu_{\mathrm{H}})}{1 + a(\nu_{\mathrm{H}})u} = \frac{\mathrm{const}}{1 + a(\nu_{\mathrm{H}})u} \to \frac{\mathrm{const}}{a(\nu_{\mathrm{H}})u}. \tag{15}$$

When comparing the absorption-gain and luminescence spectra in semiconductors and in complex molecules, far-reaching analogies can be traced. Thus, for example, the universal relation between the absorption and luminescence spectra, applicable under strong excitation of complex molecules, has the form

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$$\frac{W(\nu)}{k(\nu)} = \frac{8\pi h\nu^3}{\nu^2}\,\frac{1}{\dfrac{C_1 n_1}{C_2 n_2} e^{h(\nu-\nu_{\mathrm{el}})/kT} - 1}, \tag{16}$$

where C_2 and C_1 are the statistical weights of the upper and lower electronic states; n_2 and n_1 are their populations; and ν_{el} is the frequency of the purely electronic transition. The difference ΔF in semiconductors corresponds in this case to the quantity $h\nu_{\mathrm{el}} - kT\ln(C_1 n_1/C_2 n_2)$. Formula (11) in semiconductors can be matched, in complex molecules, with the formula

$$\frac{C_2 n_2}{C_1 n_1} e^{h\nu_{\mathrm{el}}/kT} = e^{h\nu_{\mathrm{H}}/kT}\,\frac{a(\nu_{\mathrm{H}})u}{1 + a(\nu_{\mathrm{H}})u}, \tag{17}$$

which makes it possible to calculate n_2 and n_1 from the value of $a(\nu_{\mathrm{H}})$, known from the luminescence spectrum. For $a(\nu_{\mathrm{H}})u \gg 1$, $C_1 = C_2$, and $h\nu_{\mathrm{H}} = h\nu_{\mathrm{el}}$, half of the particles are in the upper electronic state. If, at larger u, the value of ν_{H} is much greater than ν_{el}, then the overwhelming majority of the particles are transferred to level 2.

The absorption coefficient in complex molecules is determined by the formula

$$k(\nu) = \frac{h\nu}{v_g} B_{12}(\nu) n_1 \frac{1 + \alpha(\nu_H)u\{1 - e^{-h(\nu-\nu_H)/kT}\}}{1 + \alpha(\nu_H)u}, \tag{18}$$

which is very close to (13). Formula (14) is the same for both classes of substances. The principal difference between (13) and (18) is the impossibility of representing the coefficient $B'_{12}(\nu)$ in the form of two factors, one of which depends only on ν, and the other only on u. In addition, in complex molecules the parameter $\alpha(\nu_H)$ does not depend on the density of the exciting radiation.

The analogy between the optical properties of complex molecules and semiconductors makes it possible to apply to semiconductors certain conclusions obtained in the study of the generation of complex molecules (see ($^{2-4}$)).

In conclusion, we note that the universal relation is valid not only for optical excitation, but also for any other method of excitation, in particular, for excitation by an electric current or by a beam of fast electrons.

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