Excitation-power dependence of the near-band-edge photoluminescence of semiconductors

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We present a model calculation for the dependence of the near-band-edge photoluminescence (NBEPL) on the power of the exciting laser light. Our model explains all features of the NBEPL power dependence that were previously observed in experiment: (i) the variation of the excitonic photoluminescence intensity I with L^k, where L is the excitation power and k is an exponent between 1 and 2, (ii) deviations from the $I \sim L^k$ law as L is varied by more than two orders of magnitude, and (iii) the variation of k for exciton emission lines when the wavelength of the exciting laser radiation is varied. Furthermore, our model relates the k values of the free exciton, bound exciton, and the free-to-bound transitions. The results are in excellent agreement with experimental data.

I. INTRODUCTION

The low-temperature photoluminescence (PL) from II-VI and III-V compound semiconductors with reasonably good crystalline quality is dominated by near-band-edge photoluminescence (NBEPL). It has casually been stated that the underlying recombination process can be identified from the behavior of the PL intensity as the excitation power is varied.[1] As a general result it has been found[2–5] that the luminescence intensity I of the NBEPL emission lines is proportional to L^k, where L is the power of the exciting laser radiation and $1<k<2$ for exciton-like transition and $k<1$ for free-to-bound and donor-acceptor pair (D^0A^0) transitions.

In a theoretical study by Taguchi, Shirafuji, and Inuishi,[2] an attempt was made to explain the superlinearity of the bound-exciton PL by a rate equation model assuming free- and bound-exciton annihilation and free-electron to acceptor transitions. The experimentally observed $I \sim L^{1.1}$ power dependence of the free-exciton line was interpreted as a linear dependence of the exciton population of the excitation intensity. The nonlinear behavior of the bound-exciton emission intensity arises from the increase of the density of neutral impurities due to the neutralization by photoexcited carriers. The model yields $k=1$ for the free-exciton and $k=1.5$ for the bound-exciton emission. This is contrasted by recent experiments,[3,4] where several values of k between 1 and 1.7 for the exciton emission and $k<1$ for free-to-bound transitions were measured.

Alternatively, Cooper, Bajaj, and Newmann[3] assumed a constant density of binding sites for bound excitons when the excitation power was varied. They suggested that the observed superlinear power dependence of the exciton emission for above-band-gap excitation is due to the formation of excitons from free electrons and holes. Since the rate of the exciton formation depends on the product of the electron and hole densities, the exciton PL emission is expected to be quadratic in L. If the excitation wavelength is tuned to the exciton energy, the power dependence of the exciton PL emission will become linear, which was observed in the experiment. However, above-band-gap excitation gave $k<2$ for the exciton emission power dependence.

It was stated in Ref. 3 that the discrepancy between experiment and the expected quadratic dependence for above-band-gap excitation is due to the nonuniform distribution of photoexcited carriers or to competitive nonlinear and nonradiative processes.

In this paper we present a calculation of the excitation power dependence of NBEPL lines. The calculation is based on a set of rate equations for the free-exciton, bound-exciton, free-to-bound, and donor-acceptor recombinations. No further assumption on nonradiative processes are made. The steady-state solutions of the rate equations are obtained by a method recently presented by Zulehner.[6] With our model we can explain the features of the NBEPL which were observed in experiment. The model yields the power dependence of all NBEPL lines. Deviations from the $I \sim L^k$ behavior, which are observed in experiments when L is varied by more than two orders of magnitude, are also reproduced by our model. Furthermore, the variation of k for the free-exciton line which was observed[3] when the wavelength of the excitation laser light is varied from above-band-gap to resonant excitation of excitons can also be understood in the framework of our calculation.

We also present analytical relations between the k's of the free exciton, the bound exciton, and the free-to-bound emission. These relations can be used for the unambiguous identification of PL lines from their power dependence.

II. MODEL OF THE NBEPL POWER DEPENDENCE

For the calculation of the power dependence of the NBEPL, we take into account a number of transitions which are shown schematically in Fig. 1. We assume that photoexcited electron-hole pairs can recombine by the following transitions: (A) free-exciton (FE) recombination, (B) and (C) radiative recombination of donor- and acceptor-bound excitons (D^0X, A^0X), (D) donor-acceptor pair recombination (D^0A^0), (E) radiative recombination of a free electron and a neutral acceptor (eA^0), (F) radiative recombination of a free hole and a neutral donor (hD^0), and (G) and (H) nonradiative transitions of free electrons and holes to ionized donors and acceptors, respectively.

Also included in Fig. 1 are the excitations processes (I)–(L): (I) is the excitation of electron-hole pairs by above-band-gap laser light, $h\nu > E_g$, (J) stands for resonant formation of free excitons when the laser energy $h\nu \approx E_g$, and (K) and (L) are the excitation of electrons from neutral donors or ionized acceptors by the laser radiation, respectively.

Assuming that the electron concentration in the conduction band, n, is equal to the concentration of holes in the valence band, p, and neglecting thermal dissociation of free and bound excitons, we can describe processes (A)–(L) by a set of coupled differential equations:

$$\frac{dn}{dt} = iL + an^2 - gn(N_D - N_{D^0}) - enN_{A^0} , \tag{1}$$

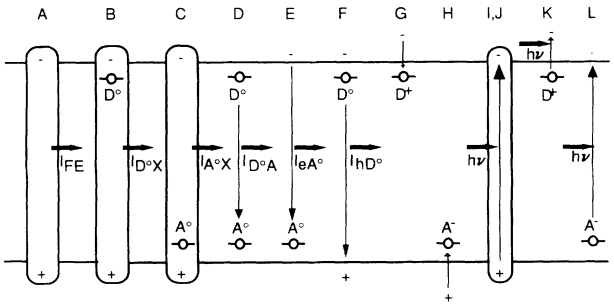

FIG. 1. Radiative and nonradiative transitions which are included in our model calculation of the near-band-edge photoluminescence. (A) Free-exciton recombination (FE), (B) and (C) radiative recombination of donor- and acceptor-bound excitons (D^0X, A^0X), (D) donor-acceptor pair recombination (D^0A^0), (E) radiative recombination of a free electron and a neutral acceptor (eA^0), (F) radiative recombination of a free hole and a neutral donor (hD^0), and (G) and (H) are nonradiative transitions of free electrons and holes to ionized donors and acceptors, respectively. Also included in the figure are the excitation processes (I)–(L): (I) is the excitation of electron-hole pairs by above-band-gap laser light, $h\nu > E_g$, (J) stands for resonant formation of the free excitons with $h\nu \approx E_g$, and (K) and (L) are the excitation of electrons from neutral donors or ionized acceptors by the laser radiation, respectively.

$$\frac{dn_{\rm FE}}{dt} = an^2 + jL - \left[\frac{1}{\tau_{\rm FE}} + \frac{1}{\tau_{\rm FE}^{\rm nr}}\right] n_{\rm FE} - bn_{\rm FE}N_{D^0} - cn_{\rm FE}N_{A^0} , \tag{2}$$

$$\frac{dn_{DX}}{dt} = bn_{\rm FE}N_{D^0} - \left[\frac{1}{\tau_{DX}} + \frac{1}{\tau_{DX}^{\rm nr}}\right] n_{DX} , \tag{3}$$

$$\frac{dn_{AX}}{dt} = cn_{\rm FE}N_{A^0} - \left[\frac{1}{\tau_{AX}} + \frac{1}{\tau_{AX}^{\rm nr}}\right] n_{AX} , \tag{4}$$

$$\frac{dN_{A^0}}{dt} = h(N_A - N_{A^0})n + i(N_A - N_{A^0})L - cn_{\rm FE}N_{A^0} + \left[\frac{1}{\tau_{AX}} + \frac{1}{\tau_{AX}^{\rm nr}}\right] n_{AX} - dN_{D^0}N_{A^0} , \tag{5}$$

$$\frac{dN_{D^0}}{dt} = g(N_D - N_{D^0})n - kN_{D^0}L - bn_{\rm FE}N_{D^0} + \left[\frac{1}{\tau_{DX}} + \frac{1}{\tau_{DX}^{\rm nr}}\right] n_{DX} - dN_{D^0}N_{A^0} - fN_{D^0}n . \tag{6}$$

In Eqs. (1)–(6) N_D and N_A are the concentrations of donors and acceptors, N_{D^0} and N_{A^0} are the concentrations of neutral donors and acceptors, $\tau_{\rm FE}$ and $\tau_{\rm FE}^{\rm nr}$ are the radiative and nonradiative lifetimes of donor and acceptor bound excitons, and $\tau_{DX}, \tau_{DX}^{\rm nr}, \tau_{AX}, \tau_{AX}^{\rm nr}$ are the radiative and nonradiative lifetimes of donor and acceptor bound excitons, respectively. $n_{\rm FE}$, n_{DX}, and n_{AX} are the concentrations of free and bound excitons, respectively, and L is the laser intensity. The coefficients $a, b, \ldots, l$ are transition rates of the processes (A)–(L) indicated in Fig. 1, respectively. The charge neutrality is fulfilled by assuming $n = p$. It is implicitly assumed in Eqs. (4) and (5) that bound excitons recombine, leaving neutral donors and acceptors. This has been observed in experiments on the thermal dissociation of excitons by Taguchi, Shirafuji, and Inuishi.[2] For simplicity, phonon-replica, two-electron and two-hole transitions, and the recombination of excitons bound to ionized donors (D^+X) are neglected in our model since these are weak transitions. The term j in Eq. (2) describes the direct formation of free excitons and can be neglected for laser light energies $h\nu$ exceeding the gap energy E_g. However, when $h\nu$ is close to E_g, this term becomes dominant.

In the steady state we get from Eqs. (2)–(4) the luminescence intensities of free and bound excitons, $I_{\rm FE}$, I_{D^0X}, and I_{A^0X}:

$$I_{\rm FE} = \frac{n_{\rm FE}}{\tau_{\rm FE}} = \frac{\beta}{\tau_{\rm FE}} n^2 , \tag{7}$$

$$I_{D^0X} = \frac{n_{DX}}{\tau_{DX}} = \frac{bN_{D^0}\beta}{1 + \frac{\tau_{DX}}{\tau_{DX}^{\rm nr}}} n^2 , \tag{8}$$

$$I_{A^0X} = \frac{n_{DA}}{\tau_{AX}} = \frac{cN_{A^0}\beta}{1 + \frac{\tau_{AX}}{\tau_{AX}^{\rm nr}}} n^2 , \tag{9}$$

$$\beta=\frac{a}{\left[\frac{1}{\tau_{FE}}+\frac{1}{\tau_{FE}^{nr}}\right]+bN_{D^0}+cN_{A^0}} . \quad (10)$$

Assuming the intensity of free-to-bound transitions to be proportional to the respective transition rates, we can write

$$I_{hD^0}\sim nN_{D^0} , \quad (11)$$

$$I_{eA^0}\sim nN_{A^0} . \quad (12)$$

In order to discuss the dependence of the luminescence intensity I on L, we distinguish between the following assumptions: In Sec. II A we discuss the case of constant N_{D^0} and N_{A^0}, as supposed in Ref. 3. In Sec. II B we discuss the assumption made in Eq. (2) that N_{D^0} and N_{A^0} may depend on the laser intensity L.

A. Constant N_{D^0} and N_{A^0}

It has been supposed in Ref. 3 that N_{D^0} and N_{A^0} are constant. In this case we only need to consider Eqs. (1)–(4).

Equation (1) gives the relation between n and L. We distinguish between three different cases: (i) Only a small portion of the free electron-hole pairs form excitons and most photoexcited carriers recombine via defect states of donors and acceptors. Neglecting the term an^2 in Eq. (1), we then obtain for the steady state

$$n=\frac{i}{eN_{A^0}+g(N_D-N_{D^0})}L . \quad (13)$$

Substituting the expression for n given by Eq. (13) into Eqs. (7)–(9), yields

$$I_{FE}=\frac{\beta}{\tau_{FE}}\frac{i^2}{[eN_{A^0}+g(N_D-N_{D^0})]^2}L^2 , \quad (14)$$

$$I_{D^0X}=\frac{bN_{D^0}\beta}{1+\frac{\tau_{DX}}{\tau_{DX}^{nr}}}\frac{i^2}{[eN_{A^0}+g(N_D-N_{D^0})]^2}L^2 , \quad (15)$$

$$I_{A^0X}=\frac{cN_{A^0}\beta}{1+\frac{\tau_{AX}}{\tau_{AX}^{nr}}}\frac{i^2}{[eN_{A^0}+g(N_D-N_{D^0})]^2}L^2 . \quad (16)$$

Equations (14)–(16) give a quadratic power dependence for the intensities of the free- and bound-exciton recombination lines. For the free-to-bound transitions (eA^0,hD^0), we obtain with Eqs. (11)–(13) a linear power dependence of the PL intensity I.

The radiative and nonradiative lifetimes of free and bound excitons appear in Eqs. (14)–(16). The competitive nonradiative processes decisively influence the intensities of the luminescence lines. Decreasing nonradiative lifetimes yield decreasing PL intensities. However, in contrast to what has been assumed in Ref. 3, nonradiative transitions influence only the magnitude of the factors multiplying L^2, but they have no effect on the exponent of L in the I vs L relations.

(ii) For dominating exciton recombination, the terms which are linear in n can be neglected in Eq. (1). In this case we obtain $n\sim L^{0.5}$ for the steady state, which was supposed in Ref. 2. With Eqs. (7)–(9) we now obtain $I\sim L$ for excitonic PL lines. According to Eqs. (11) and (12), the intensity of free-to-bound transitions then varies as $I\sim L^{0.5}$.

(iii) If neither excitonic nor donor-acceptor recombination dominates, the linear and quadratic terms in n are of the same order of magnitude in Eq. (1), resulting in curved $\log(I)$-$\log(L)$ plots.

At a very low level of excitation power L, a behavior as described in (i) is expected. As the excitation level is increased and a larger number of carriers form excitons, the system approaches the state described in (ii).

The analysis presented in Sec. II A can only be applied to systems where all exciton lines show the same power dependence and where the PL signal of a donor-acceptor pair transition, which is supposed to depend on the product of $N_{D^0}N_{A^0}$, is independent of L.

B. Variation of N_{D^0} and N_{A^0} with L

Assuming that N_{D^0} and N_{A^0} depend on L, Eqs. (1)–(6) now have to be taken into account. No simple analytic solutions like those described in Sec. II A are obtained. However, relations between the k values, e.g., the value of $d(\log I)/d(\log L)$ from different PL lines, may be established. Assuming that $1/\tau_{FE}+1/\tau_{FE}^{nr}\gg bN_{D^0}+cN_{A^0}$, we obtain from Eqs. (7)–(9):

$$n\sim(n_{FE})^{0.5} , \quad (17)$$

$$I_{D^0X}\sim n_{FE}N_{D^0} , \quad (18)$$

$$I_{A^0X}\sim n_{FE}N_{A^0} . \quad (19)$$

With Eqs. (18) and (19) we find

$$k_{D^0X}(L)=k_{FE}(L)+k_{D^0}(L) , \quad (20)$$

$$k_{A^0X}(L)=k_{FE}(L)+k_{A^0}(L) . \quad (21)$$

By k_{D^0X}, k_{A^0X}, and k_{FE} we denote the slope of $\log(I_{D^0X})$, $\log(I_{A^0X})$, and $\log(I_{FE})$ vs $\log(L)$. Equations (20) and (21) show that $k_{D^0X}(L)$ and $k_{A^0X}(L)$ may be written as the sum of $k_{FE}(L)$ and $k_{D^0}(L)$ or $k_{A^0}(L)$, respectively. The latter contain the effect of photoneutralization of the density of neutral donors and acceptors.

With Eqs. (11), (12), (17), (20), and (21) we finally obtain

$$k_{eA^0}=\frac{k_{FE}}{2}+(k_{A^0X}-k_{FE}) , \quad (22)$$

$$k_{hD^0}=\frac{k_{FE}}{2}+(k_{D^0X}-k_{FE}) . \quad (23)$$

Assuming that $I_{D^0A^0}$ is proportional to $N_{D^0}N_{A^0}$ we obtain for the donor-acceptor pair transition

$$k_{D^0A^0}=(k_{D^0X}-k_{FE})+(k_{A^0X}-k_{FE}) . \quad (24)$$

C. Influence of the excitation wavelength on the laser power dependence of PL lines

We have shown in Secs. II A and II B that our model gives $k \geq 1$ for free and bound excitons when $h\nu$ of the exciting laser radiation exceeds the gap energy E_g of the semiconductor. In Ref. 3 it was demonstrated that free- and bound-exciton PL lines exhibit a linear power dependence when $h\nu \approx E_g$, but a pronounced nonlinear behavior for above-band-gap excitation. We explain these experimental data within the framework of our model assuming the process of direct formation of free excitons to be the dominating process when the excitation energy $h\nu$ is close to the gap energy E_g and the term jL in Eq. (2), which describes the direct creation of free excitons, becomes important and the creation of free electrons and holes by dissociation of free excitons is neglected. These assumptions are supported by the experimental observation[3] that for $h\nu \approx h\nu_{\mathrm{FE}}$ no free-to-bound transition hD^0 is observed, indicating that the density of free carriers is small. However, the same sample shows a strong transition hD^0 when $h\nu > E_g$, indicating a large density of free carriers. Using the above assumptions and Eqs. (2)–(4) we obtain for the steady state:

$$I_{\mathrm{FE}} = \frac{n_{\mathrm{FE}}}{\tau_{\mathrm{FE}}} = \frac{j}{1 + \frac{\tau_{\mathrm{FE}}}{\tau_{\mathrm{FE}}^{\mathrm{nr}}} + bN_{D^0}\tau_{\mathrm{FE}} + cN_{A^0}\tau_{\mathrm{FE}}} L \ , \quad (25)$$

$$I_{D^0X} = \frac{n_{DX}}{\tau_{DX}} = \frac{bN_{D^0}}{1 + \frac{\tau_{DX}}{\tau_{DX}^{\mathrm{nr}}}} n_{\mathrm{FE}} \ , \quad (26)$$

$$I_{A^0X} = \frac{n_{AX}}{\tau_{AX}} = \frac{cN_{A^0}}{1 + \frac{\tau_{AX}}{\tau_{AX}^{\mathrm{nr}}}} n_{\mathrm{FE}} \ . \quad (27)$$

For N_{D^0}=const and N_{A^0}=const, which we assume to be fulfilled at low excitation levels, when the effect of photoneutralization is weak, Eqs. (25)–(27) give a linear power dependence of the free- and bound-exciton PL intensity in excellent agreement with the experimental results of Cooper, Bajaj, and Newmann.[3]

IV. EXPERIMENTAL RESULTS AND DISCUSSION

Figure 2 shows a typical 1.6-K near-band-edge PL spectrum from a CdTe epilayer grown on GaAs by hot wall epitaxy.[7,8] The strong emission lines at about 780, 778, and 779 nm are due to the radiative recombination of excitons bound to shallow neutral acceptors (A^0X), shallow neutral donors (D^0X), and ionized donors (D^+X), respectively. Free-to-bound transitions are observed at 778.7 (hD^0) and 800 nm (eA^0). The recombination of free excitons in the ground state (FE, $n=1$) and the first excited state (FE, $n=2$) gives rise to weak emissions at 776.4 and 773.1 nm. (Free and bound excitons also recombine with the emission of a LO phonon, which produces a readily observed phonon replica at energies 21.2 meV less than the principal lines.)

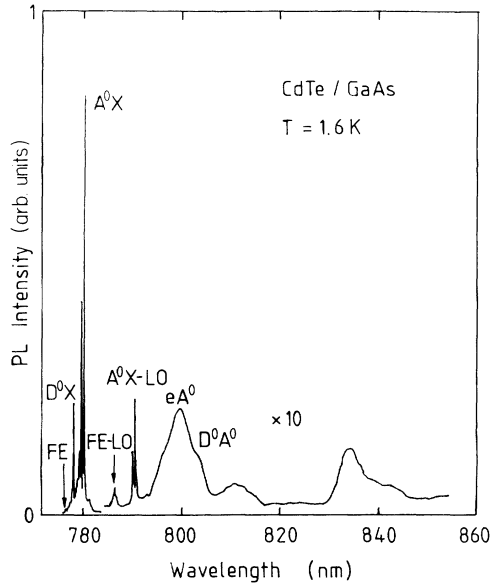

FIG. 2. The 1.6-K near-band-edge photoluminescence from a CdTe epilayer grown on GaAs. The emission lines are due to radiative recombination of free-excitons (FE), recombination of excitons bound to donors and acceptors (D^0X, D^+X, A^0X) and free-to-bound transitions (hD^0, eA^0) and donor-acceptor pair transitions (D^0A^0). Also shown are the LO phonon replica.

Figures 3 and 4 show log(I)-log(L) plots of the intensity I of various CdTe PL lines versus the exciting laser power L. The circles indicate the experimental results. The laser power was varied by almost four orders of magnitude. The data clearly show a deviation from the $I \sim L^k$ law. The curves in Figs. 3 and 4 are calculated us-

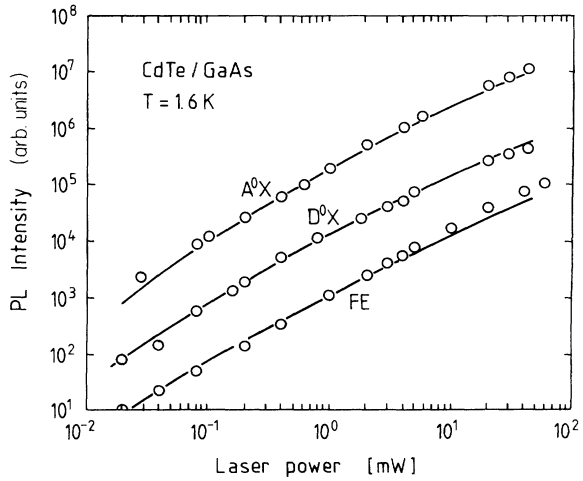

FIG. 3. The intensity of the free (FE) and bound exciton (A^0X, D^0X) photoluminescence from epitaxial CdTe vs the power of the exciting laser radiation. The laser wavelength is 647.1 nm. The laser spot size on the sample surface is 0.7 mm^2. Circles indicate experimental data, and the curves are calculated with our model of the near-band-edge photoluminescence.

TABLE I. The k values of CdTe luminescence lines. The coefficient k is obtained by fitting the measured power dependence of the luminescence lines by an $I \sim L^k$ law (I is the PL intensity, L is the power of the exciting laser light). The numbers in parentheses are calculated using Eqs. (22)–(24).

Reference	FE	D^0X	A^0X	hD^0	eA^0	D^0A^0	Power range (W/cm^2)
This work	1.2	1.4	1.3		0.7		0.003–0.3
This work	1.0	1.1	1.1	0.7	0.6		0.3–6
				(0.6)	(0.6)	(0.2)	
Ref. 3.	1.25		1.25	1.03			10^{-3}–1
		(1.6)			(0.63)		
Ref. 2.	1.1	1,5	1.5,1.7		1–1.1		
				(0.95)	(0.95–1.15)	(0.8–1.0)	
Ref. 4	1.3	1.4	1.4	(0.85)	(0.85)	0.76[a]	0.4–10

[a]This line was attributed to a D^0A^0 transition in Ref. 4. However, from the line energy and the shift of the line with the laser power we tend to interpret it to be due to a free-to-bound transition.

ing our model of the NBEPL power dependence, as described in Sec. II. The parameters in Eqs. (1)–(6) were chosen in order to obtain the best fit between theoretical and experimental data. Our model can satisfactorily reproduce the power dependence of all NBEPL lines.

The experimental results of the PL power dependence obtained by other authors[2–4] are summarized in Table I. In Refs. 2–4 the measured power dependence was fitted by an $I \sim L^k$ law and the values of k are given in Table I. It should be noted, however, that some of the published experimental data show a deviation from this behavior. If we identify the k's with the slope of the log(I)-log(L) curve at a certain level of laser intensity, we can use Eqs. (21)–(23) in order to relate k_{FE}, k_{D^0X} or k_{A^0X}, and k_{hD^0}, K_{eA^0}, $k_{D^0A^0}$. The numbers in parentheses in Table I are calculated using Eqs. (21)–(23), respectively. As can be seen, we find an excellent agreement of all published experimental data with our model calculation. This may be considered as an experimental test of our model.

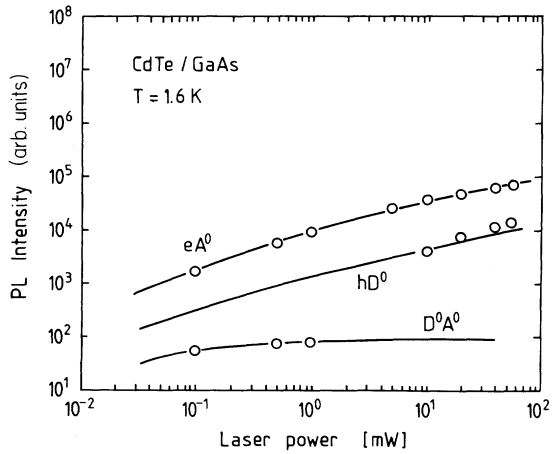

FIG. 4. The photoluminescence intensity of the free-to-bound (hD^0,eA^0) and the donor-acceptor pair recombination (D^0A^0) from epitaxial CdTe vs the power of the exciting laser radiation. The laser wavelength is 647.1 nm. The laser spot size on the sample surface is 0.7 mm^2. Circles indicate experimental data, and the curves are calculated using our model of the NBEPL with the same parameters as in Fig. 3.

V. CONCLUSIONS

The understanding of the photoluminescence power dependence is considered to be important for the identification of the underlying recombination processes and the evaluation of the crystal quality. Earlier models of the photoluminescence power dependence suffered from the fact that they did not cover all features of the measured NBEPL power dependence, which are as follows.

(a) The power dependence of the luminescence lines can be described by an $I \sim L^k$ law (when L is varied over a range of less than two orders of magnitude), where I is the luminescence intensity and L is the excitation laser intensity, k is a coefficient. However, deviations from this behavior are observed, when L is varied over more than three decades.

(b) For excitation laser light with $h\nu$ exceeding the gap energy E_g, the coefficient k is generally $1 < k < 2$ for the free- and bound-exciton emission. k is less than 1 for free-to-bound and donor-acceptor pair recombination.

(c) When $h\nu$ of the laser radiation is tuned to the semiconductor gap energy, the coefficient k for the free-exciton emission becomes equal to 1.[3]

Earlier models of the NBEPL (Refs. 2 and 3) were able to explain only some of the above-mentioned experimental results satisfactorily. In Ref. 2 the superlinearity of the bound-exciton emission was described by a rate equation model taking into account free-exciton, bound-exciton, and free-to-bound transitions. The model, however, is based on the assumption that $k = 1$ for the free-exciton emission, which is in contrast to experimental results, and, furthermore, the model does not contain the important donor-acceptor pair recombination. The nonlinear behavior of the bound exciton was believed to arise from the increase of the population of binding sites due to the neutralization of donors and acceptors by photoexcited carriers. Alternatively in Ref. 3 the density of binding sites for bound excitons is assumed constant with varying laser power. A quadratic power dependence of the free- and bound-exciton emission is found, which is, however, rarely observed in experiment.

In our model of the NBEPL we have solved a set of

rate equations for the free-exciton, bound-exciton, free-to-bound, and donor-acceptor pair recombinations, assuming a set of transition rate coefficients. No further assumptions, e.g., on nonradiative processes, are needed to obtain a good fit of the measured power dependence of all NBEPL lines from CdTe when the excitation power is varied over almost four orders of magnitude. Our model can be used to explain the variation of k of the free-exciton emission when the wavelength of the radiation is varied, as was observed in Ref. 3. As a further important result our model yields analytic relations between the k values of the free exciton, the bound exciton, the free-to-bound, and the donor-acceptor pair emission, respectively. These relations can be used for the identification of near-band-edge photoluminescence lines. As our model is not restricted to one particular semiconductor, it may be applied to any direct semiconductor.

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