angular distribution of fluorescence by crystal impurities, analogous to the well-established studies of radiations from single atoms and from nuclei.

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Coulomb Effects on the Gain Spectrum of Semiconductors

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We have investigated the effect of Coulomb interactions on the gain spectra of highly excited semiconductors, specifically GaAs. It is shown that at low temperatures there is an enhancement of the direct excitation of approximately a factor of 2 and a sideband due to particle-hole pair excitations that extends into the gap. As the temperature increases, it is argued by analogy to the electron-phonon problem that the sideband changes into an Urbach tail with gain of the order of 10–100 cm^{-1}.

Stimulated recombination in direct-gap semiconductors has been under intensive study in the last decade.[1,2] There exist extensive calculations of gain versus temperature and degree of inversion based on independent-particle models taking into account band-structure effects[3] and impurity-band tailing.[4,5] Recently, direct measurements of the gain spectrum have become available for optically pumped systems at low temperature and for a variety of pump powers.[2] One of the most striking features is that gain begins only at one or two exciton energies below the gap. This effect has been explained[6,7] as being a result of exchange and correlation. We discuss in this paper two additional effects due to this Coulomb interaction: (i) an enhancement of the band-to-band gain, and (ii) emission below the band-to-band processes that takes the form of a plasmon sideband at low temperatures and a self-induced band tail at high temperatures. We argue that this band tail may be quite important in understanding the properties of undoped or lightly doped ($<10^{18}$ cm^{-3}) GaAs lasers operating at room temperature. This sideband effect has previously been discussed.[8] However, there are large cancelations omitted by previous authors, and consequently our results are very different.

We shall limit ourselves to isotropic electron and hole bands described by effective density-of-states masses m_e and m_h. We shall also specialize to GaAs as an illustration and take $m_h/m_e = 9.5$. The absorption and gain of an inverted semiconductor is given by the imaginary part of the dielectric function. This is calculated to lowest order in the dynamically screened Coulomb interaction, $V(q) = (4\pi e^2/q^2)[\epsilon(q, \omega)]^{-1}$, by summing the three diagrams shown in Fig. 1. The dielectric function $\epsilon(q, \omega)$ of the electron-hole plasma is further approximated by the single-plasmon-pole approximation that has been used extensively by Hedin and Lundquist.[9] However, in order to attempt to represent the true spectrum of a zinc-blende material,[10] we broaden the plasmon into a Lorentzian whose width is $\frac{1}{10}$ of the plasma frequency. When the gain spectrum is calculated, we find an enhancement of the band-to-band recombination. This enhancement varies somewhat with frequency and is approximately a factor of 2 at low temperature. The slightly distorted gain spectrum is compared with the single-particle result in the lower right-hand corner of Fig. 1 for a typical electron-hole density at low temperature. It is seen that the enhancement is considerably reduced compared to that of the exciton problem[11] because of the screening of the Coulomb interaction. We have crudely estimated the effects of multiple scattering by using the work of Mahan[12] and found them to be small. In the same calculation, we can look at Auger processes involving the emission

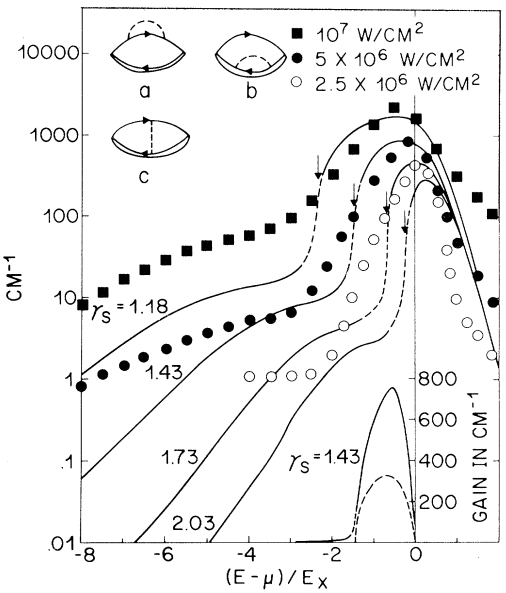

FIG. 1. Spontaneous-emission spectrum for GaAs at $T=0.25E_x$ ($E_x \approx 40$°K is the exciton binding energy) for four densities [$n=3/4\pi(r_s a_B)^3$]. $r_s=1.43$ corresponds to $n=2\times10^{16}$ cm^{-3}. Energy is measured in exciton units and the curves are lined up so that different densities have the same chemical potential μ. Arrows indicate the bottom of the band in a noninteracting model. The data points, measured at 2°K...are taken from Ref. 2. The calculation is based on the three diagrams labeled *a*, *b*, and *c*, where single and double lines denote electron and hole propagators, and dashed lines denote elementary excitations. The lower right-hand corner shows the gain spectrum calculated in the noninteracting-particle model (dashed line) and including Coulomb effects (solid line).

of a plasmon or a single-particle excitation. These processes are responsible for the tailing into the band gap of the gain curve shown in the inset of Fig. 1. This effect is shown more clearly in Fig. 1 in the logarithmic plot of the spontaneous-emission spectrum at $T=0.25E_x$ for four different densities. The curves have been lined up so that energy is measured from the same chemical potential μ, below which laser action begins. Arrows mark the bottom of the band in a noninteracting model. The curves clearly show a sideband which extends deeper into the gap for increasing density. The sideband is quite small when compared to the direct recombination because of the large cancelation between the three diagrams in Fig. 1. Physically, this cancelation is due to the fact that an electron-hole pair is a neutral object, and interacts less strongly with a plasmon than an electron or hole separately. Previous authors[8] have neglected vertex correction [Fig. 1(c)], and their result is about a factor of 10 too large. It should be noted that the calculation of the sideband from these diagrams actually diverges at the band edge because of the low-frequency excitations. We have simply interpolated through this region with the dashed lines shown in Fig. 1. The spontaneous emission can be compared with luminescence data. In Fig. 1, we have also plotted data[2] taken at 2°K and at three pump powers. The peak positions of each curve and the absolute intensity are adjusted to give a best fit. The measured gain at each pump power is comparable to the calculated value. The high-energy side of the data indicates that there is considerable heating as pump power is increased. Since we are comparing the data with curves calculated at the same temperature, ~10°K, the agreement is quite reasonable.

For room-temperature lasers, there is little doubt that for highly compensated materials ($n > 10^{18}$ cm^{-3}) the impurity-band tail is responsible for the laser action. If the impurity band is involved, the calculated threshold current should increase when the impurity content is decreased.[4,5] Experimentally, however, it is found that between an impurity content of 10^{16} and 10^{18} there is essentially no change in the laser threshold.[13] While the plasmon sideband is responsible for a gain of several tens of reciprocal centimeters at low temperature and high degree of inversion, at room temperature our calculations indicate that it contributes only a few reciprocal centimeters. However, our perturbation calculation is expected to break down at high temperature when the thermal occupation of excitations is large, i.e., when kT is larger than the heavy-mass plasma frequency. This is completely analogous to the situation in ionic crystals where phonon sidebands at low temperature broaden into an Urbach tail at high temperature.[14] While there has been some success in treating the multiple-phonon problem by summing selected classes of diagrams the most successful approach has been to treat the phonon excitation as a static distribution of fluctuating potentials,[15] and we attempt a similar discussion here. We first note that because of the heavy mass, the hole distribution is Boltzmann even under lasing conditions. Therefore, we try treating them as classical particles. It has been pointed out, in connection with the problem of trapping of electrons in He,[16] that interaction of electrons with classical particles is equivalent to the impurity-band-tail problem.

A pure material when inverted to an electron and hole density n ($n \approx 10^{18}$ at room temperature) will then have an Urbach tail similar to an n-type material with n ionized impurities. In particular, this predicts that laser threshold at room temperature should not be sensitive to doping up to $\sim 10^{18}$. Further, the luminescence spectrum should tail increasingly into the band gap as the pump power is raised at a given temperature. This latter effect has been observed in GaAs and CdS and was given the above interpretation.[17] A natural question to raise is to what extent the motion of the holes will destroy the analogy with a static distribution of impurities. The same question has to be faced in existing treatments of phonon-induced Urbach tail. Halperin[18] has examined this question by going to an exactly solvable model involving a two-level system interacting with a phonon bath. To study the classical approximation in a more general situation, we would like to start with a system of n interacting electrons and holes described by $H = H_0 + H_1$, where H_0 is the hole kinetic energy and H_1 is the sum of the electron kinetic energy and the Coulomb energy of the electron-hole system.

We would like to write down a formal expression for the dielectric function in an approximation in which only holes are treated classically. Formally this is an expansion in $\hbar$, coming from the commutator involving hole coordinates, $[P_i, R_j] = \hbar\delta_{ij}$, but keeping intact the commutators involving electron coordinates. We have developed such an expression for

$$e^{\lambda H} = \exp(\lambda H_0)\exp(\lambda H_1)u(\lambda), \tag{1}$$

and calculated $u(\lambda)$ to second order in $\hbar$. Our result reproduces the well-known correction $\hbar^2(\nabla\nabla)^2/24m(kT)^3$ to the energy of a completely classical system,[19] and contains further terms for the present semiclassical situation. (Here we denote by ∇V the average of the gradient of the potential seen by each of the positive charges.) These results will be discussed in a separate publication. The dielectric function can be calculated from the correlation function

$$D(\tau) = \sum_{k,k'} \langle b_{k'}(\tau) c_{k'}(\tau) c_k^{\dagger}(0) b_k^{\dagger}(0)\rangle,$$

where $c_k^{\dagger}$, $b_k^{\dagger}$ create an electron and a hole, respectively. The zeroth-order approximation is given by

$$D(\tau) = Z_{c1} \sum_{k,k'} \exp[-\tau(\hbar k')^2/2m_h]\,\mathrm{Tr}[\exp(-\beta H_1)\exp(-\tau H_1) b_{k'} c_{k'} \exp(\tau H_1) c_k^{\dagger} b_k^{\dagger}], \tag{2}$$

where $Z_{c1} = \mathrm{Tr}[\exp(-\beta H_1)]$. Going to a spatial representation we can take the trace over the hole coordinates and obtain

$$D(\tau) = Z_{c1}^{-1} \sum_{\{R_i\}} \sum_{k',r,r'} \exp[ik'(r-r')]\exp[-\tau(\hbar k')^2/2m_h] \times \mathrm{Tr}\left\{\exp[-\beta+\tau)H_1(\{R_i\})]\,\Psi(r)\exp[\tau H_1(\{R_i, r'\})]\,\Psi^{\dagger}(r')\right\}, \tag{3}$$

where $\Psi^{\dagger}(r)$ creates an electron at r. On Fourier transforming and then continuing to real frequency E, we note that if $(\hbar k')^2/2m_h \ll E$ then the k' integral gives $\delta(r-r')$, and formally Eq. (3) describes the annihilation of an electron at a static impurity site subject to a distribution of potentials determined by the Boltzmann factor due to all other electrons and holes. The recoil energy $(\hbar k')^2/2m_h$ is also present in the impurity-band problem since one hole is created in the valence band in optical absorption. Thus the above result is exactly the same as the impurity-band problem except that the distribution of the impurities is characterized by kT.[20]

We want to study the conditions under which the corrections to $D(E)$ of order $\hbar^2$ are small. We anticipate that $D(E)$ varies exponentially with E. Thus a reasonable criterion is that corrections to $\ln D(E)$ should be small. We have shown that this is satisfied provided that

$$\frac{1}{4(kT)^3}\frac{\hbar^2}{m}\left\langle\left(\frac{\partial V}{\partial r}\right)^2\right\rangle\left[\frac{\partial \ln D(E)}{\partial E}\right]^4 \ll \ln D(E), \tag{4}$$

where $V(r)$ is the potential seen by a single hole and the angular brackets mean the thermodynamic average. The above conclusion has been reached under the assumption that $\partial[\ln D(E)]/\partial E \gtrsim 1$. Otherwise the condition is simply

$$(kT)^{-3}(\hbar^2/m)\langle(\partial V/\partial r)^2\rangle \ll 1. \tag{5}$$

For a Coulomb gas, $\langle(\partial V/\partial r)^2\rangle = \langle\partial^2 V/\partial r^2\rangle k_B T = 4\pi n e^2 k_B T$, where n is the charge density at the site of the hole, which is approximately the density. Assuming $D(E) = \exp(\alpha E/k_B T)$, Eq. (4) then becomes

$$\tfrac{1}{4}(\hbar\omega_1/kT)^2\alpha^3 \ll (E/kT), \tag{6}$$

where $\omega_1 = (4\pi e^2 n/m_h)^{1/2}$ is the hole plasma frequency. This condition indicates that fluctuations must be sufficiently strong that the exponential dependence of the Urbach tail is relatively slowly varying over an energy of order kT.

From the data of Kressel *et al.*[17] we infer values of α of 1.6, 1.0, and 1.0 for $T = 300$, 145, and 77°K, respectively. Using a value of $\hbar\omega_1/kT = \frac{1}{2}$ in Eq. (6) we find that our condition is fairly well satisfied. Furthermore, these values of α should be compared with that of the impurity-band tail of n-type semiconductors. Using the recent absorption data of Hwang[21] we find comparable values of α at a doping level of 3×10^{18} at room temperature and 1×10^{18} at 77° K. These are the typical densities near laser threshold and the agreement is quite satisfactory.

Finally, as pointed out by Kressel *et al.*,[17] the fluctuation model discussed here also gives a more consistent explanation of the fact that the laser action occurs 20–30 meV from the peak in the luminescence spectrum. It does not appear likely that the average band gap can be renormalized by this amount as the renormalization effects tend to decrease with temperature and probably are not more than 20 meV at low temperatures.

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