Optical Properties of Fast-Diffusing Solid-State Plasmas

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Transmission and emission spectra of fast-diffusing nonequilibrium electron-hole plasmas in semiconductors are calculated with use of displaced Fermi distributions. The carrier drift significantly alters the plasma spectra and removes previously reported incomprehensible discrepancies between experimental and theoretical plasma parameters, indicating the necessity to reinterpret entirely the spectroscopic data from nonequilibrium plasmas.

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Equilibrium and nonequilibrium electron-hole plasmas (EHP) have been studied extensively in optically excited semiconductors.[1,2] In the case of the equilibrium electron-hole liquid (EHL) in indirect-gap materials quantitative agreement between theory and experiment has been reported for all important parameters. Above the EHL critical temperature T_c a nonequilibrium electron-hole plasma is observed for which this agreement abruptly ceases: In Fig. 1 we show experimental and theoretical values for the many-body band-gap renormalization $E_g' - E_g$ (in units of the respective exciton Rydberg) versus the interparticle distance (measured in exciton Bohr radii) for EHL and EHP in various semiconductors.[3] Very recently it has been demonstrated theoretically[4] that in these units the renormalization is highly insensitive to details of the semiconductor band structure. In Fig. 1 this is *verified experimentally* by EHL data from unstressed Ge and Si as well as from Si stressed uniaxially along the ⟨111⟩ axis. In contrast, for the EHP in the *indirect-gap* materials the experimental renormalization is *smaller* by about 1 Ry than calculated. On the other hand, as shown here for GaAs, the experimental values of the band renormalization in *direct-gap* semiconductors are typically *larger* by 1 Ry than predicted.[2,4]

Recently the EHP expansion due to gradients in the plasma density and temperature in GaAs has been studied by spatially resolved luminescence spectroscopy.[5] From the spatial distribution of the EHP luminescence intensity a drift velocity v_D of about 1×10^7 cm/s was estimated. In Ge velocities close to 3×10^6 cm/s have been assigned to the plasma expansion previously.[6] Note that the Fermi velocities v_F in the plasma range typically from 10^6 to 10^7 cm/s. Since the carrier distributions represent the momentum of the plasma expansion, $v_D \cong v_F$ implies large deviations of the distributions from the equilibrium Fermi-function forms.

In this paper we demonstrate for the first time that the fast ambipolar expansion of optically excited nonequilibrium EHP in semiconductors is essential for the understanding of its optical response. Including the plasma drift in the evaluation of the experimental spectra removes the striking discrepancies between experimental and theoretical results with respect to line shape, band-gap renormalization, and binding energy and shows that the discrepancies are due to the

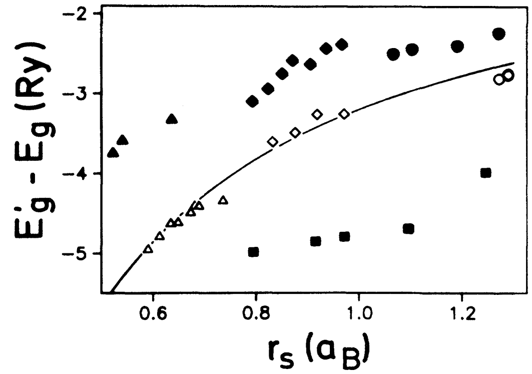

FIG. 1. Experimental band renormalization in EHL (open symbols) and EHP (full symbols) in unstressed Ge (triangles), unstressed Si (lozenges), Si stressed along ⟨111⟩ (circles), and GaAs (squares) (Ref. 3). The Ge data were obtained under cw excitation (10 kW/cm²) by a neodymium-doped yttrium aluminum garnet laser focused to a 100-μm-diam spot, the other data by pulsed excitation (intensities of ≥ 100 kW/cm²). Solid line: theory (Refs. 1 and 4); see text.

unjustified use of equilibrium Fermi functions in the calculation of the spectra. As predicted by our model, luminescence from a stationary plasma produced by stress confinement is in good agreement with the equilibrium theory.

We approximately characterize the nonequilibrium EHP by distribution functions $f_{e,h}(\vec{k}) = f_{e,h}^{0}(\vec{k} - \vec{k}_{De,h})$,[7] where $f_{e,h}^{0}$ are equilibrium Fermi functions defined by a characteristic plasma density and temperature[8] and k_D is an effective drift vector. Ambipolar diffusion requires equal drift velocities of electrons and holes. For a one-component electron system this is a standard procedure and is used to study, e.g., electrical transport.[7] The principally new feature here is that these distributions determine the optical response of the ambipolar plasma and are thus directly accessible to experiment.

Including the drifted Fermi functions[7] in the well known formulas for the optical spectra[1,2] straightforwardly yields transmission and emission spectra of drifting plasmas. In the upper part of Fig. 2 calculated gain spectra of an EHP in CdS are shown for increasing drift. The plasma drift leads to two striking consequences:

(i) The crossover from gain to absorption, which occurs at the chemical potential for $v_D = 0$, shifts with increasing v_D to lower energies. As the position of the renormalized gap ($E = 0$ in Fig. 2) is unaffected by the drift, the gain width is significantly reduced at high v_D.

(ii) The low-energy part of the gain line shape, which varies proportional to $(h\nu)^{1/2}$ for $v_D = 0$, increases proportional to $(h\nu)^2$ at high v_D. Experimentally the gain varies as $(h\nu)^2$ near the renormalized energy gap, which previously motivated a "no-k selection" model of the gain,[3,9] although in direct transitions the momentum is conserved principally. Similar but weaker drift-induced changes are found for the spontaneous emission.

In the recombination in indirect-gap semiconductors momentum is conserved by phonon participation. Hence the spectra depict the distribution of the carriers in energy space. The luminescence broadens for increasing drift since the carriers are distributed over a wider range of energies (bottom of Fig. 2).

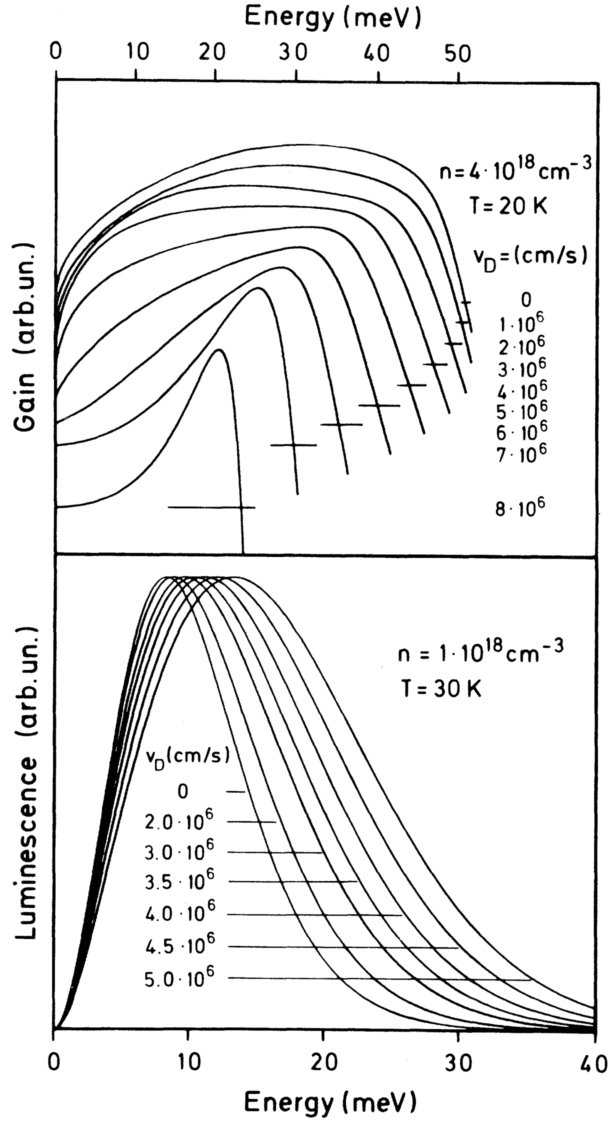

FIG. 2. Top: Theoretical line shapes of the EHP transmission in CdS at different drift velocities. The horizontal lines indicate the crossover from gain to absorption. Bottom: EHP emission in Si for different velocities.

We now compare our calculated line shapes to experimental luminescence and gain spectra. In Fig. 3 the open circles depict an equilibrium EHL spectrum of Ge. The full circles show a plasma emission at $T = 7$ K, slightly above T_c.[1] Assuming equilibrium Fermi distributions ($v_D = 0$), we obtain $n = 1.9 \times 10^{17}$ cm^{-3} for the EHL ($T = 4$ K) and $n = 2 \times 10^{17}$ cm^{-3} for the EHP (dashed lines). The band renormalization in the EHL, however, is larger by about 4 MeV (corresponding to 70% of the emission half-width) than the renormalization in the plasma. As shown in the inset in Fig. 3 and in Fig. 1 this difference is typical for EHL and EHP of apparently similar densities. The discrepancy is entirely resolved if we include the carrier drift in the line-shape calculation (solid line, Fig. 3). For a density of 6×10^{16} cm^{-3}, the respective theoretical renormalization $E_g(n)$, and a drift velocity of $\cong 3 \times 10^6$ cm/s, a good fit of

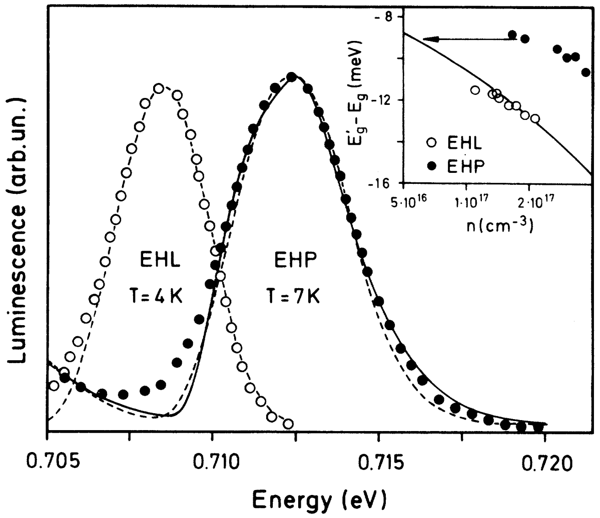

FIG. 3. Luminescence of EHL and EHP in Ge. The fitted temperatures are indicated. Dashed line: fit using equilibrium Fermi functions and a freely adjustable band gap. Solid line: fit including drift and the calculated position. Inset: renormalization in EHL and EHP obtained with use of equilibrium distributions. Solid line: theory (Ref. 4). The arrow indicates the change in renormalization and density due to the drift.

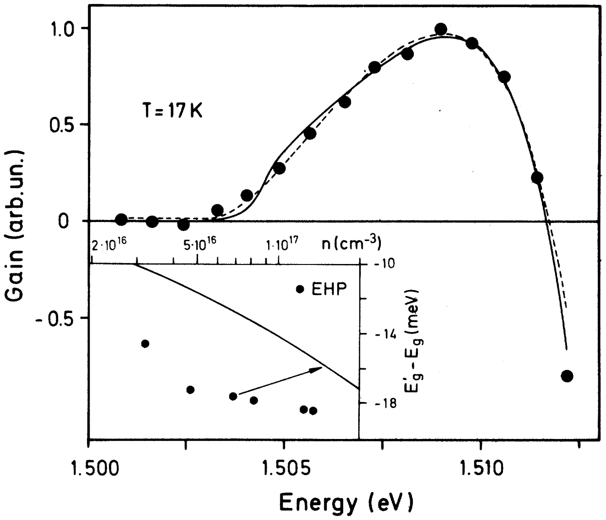

FIG. 4. Experimental transmission spectrum of EHP in GaAs (Ref. 3). The temperature was derived from the spontaneous emission. Dashed line: fit neglecting k conservation and using an adjustable position. Solid line: fit including k conservation, theoretical renormalization, and drift. Inset: renormalization of EHP ($v_D=0$) (Ref. 3). Solid line: theory (Refs. 4 and 12).

the shape *and* position of the spectrum is obtained.[4,10,11]

Figure 4 displays a transmission spectrum of GaAs ($T = 17$ K).[3] The dashed line indicates the usual fit neglecting k conservation and gives a density of 7×10^{16} cm^{-3}. As depicted in the inset the experimental position of the renormalized gap E_g' at this density is lower by more than 1 Ry (3.7 MeV) than calculated.[3,12] This difference is completely unexpected as the calculations of E_g' even include the small-polaron effects. Under the assumption of equilibrium Fermi distributions, the crossover from gain to absorption occurs at the chemical potential of the EHP. This seems to indicate a strongly bound EHL state in this direct-gap material, but contradicts the experimental data for the density temperature dependence[3] and the results of various calculations.[1,2,12] Including the drift and maintaining k conservation we calculate the spectrum shown by the solid line in Fig. 4. We obtain a drift velocity of 7×10^{6} cm/s, a density of 1.5×10^{17} cm^{-3}, and vanishing binding energy for the plasma, in agreement with the theoretical band renormalization. The spectroscopically determined velocity agrees well with a value obtained independently by Romanek *et al.*[5,13]

Our paper implies that the optical spectra of plasmas above the Mott density[1] are described rather accurately by the single-particle properties and therefore many-body effects (apart from a rigid band renormalization) are comparatively small. This contrasts with a recent theoretical approach[14] which explains the shapes of the transmission spectra of EHP by strong many-body effects. This approach, however, does not resolve the inconsistent plasma band renormalization

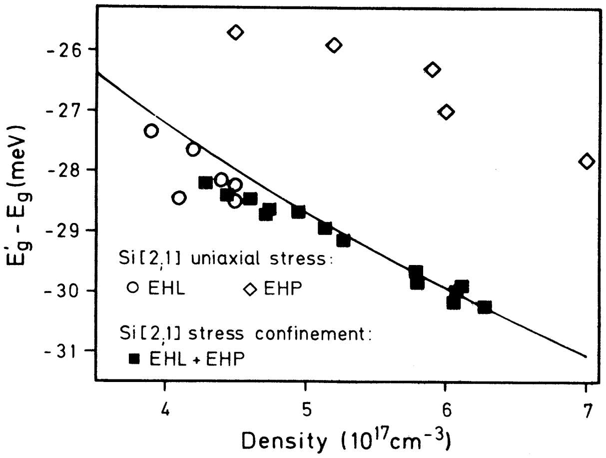

FIG. 5. Renormalization in EHL and EHP in highly stressed Si. Open symbols, uniaxial stress; full squares, stress confinement (Ref. 15). See text.

shown in Fig. 1.

Finally we directly demonstrate the significant changes in the optical response if the drift is excluded. Figure 5 shows the band renormalization of EHL (circles) and EHP (open squares) in uniaxially $\langle 100 \rangle$-stressed Si in comparison to data from EHL and EHP (full squares) under $\langle 100 \rangle$-stress confinement.[15] With use of equilibrium Fermi functions the band renormalization in the plasma in uniformly stressed Si[2; 1] is always smaller by $\cong 3$ MeV than in the EHL at the same density. In contrast, a sufficiently high gradient in the stress well may counterbalance the plasma expansion under stress-well conditions. In this case we expect for EHL and EHP a band renormalization in agreement with the equilibrium theory. As shown in Fig. 5 this is indeed observed: The experimental data for the band-gap renormalization under stress confinement are consistent with the theory for all densities, although the data correspond to temperatures ranging from 7 to 30 K, which is significantly higher than the critical temperature for EHL in Si.

In conclusion, our model for the optical spectra of diffusing plasmas removes long-standing discrepancies between experimental and theoretical plasma parameters which are traced to the usual equilibrium description of the EHP. We find that equilibrium evaluations of the spontaneous luminescence in indirect-gap semiconductors typically overestimate the plasma density by a factor of about 3, whereas from transmission in direct-gap materials n is underestimated by a factor of about 2. Our results underline the importance of drift effects for the physical properties of non-equilibrium plasmas in general. This includes, e.g., the Auger recombination and Raman scattering in EHP. Drift effects are expected to be particularly important at the extreme excitation intensities used for pulsed laser annealing where they may strongly modify the calculation of the plasma reflectivity.[16]

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