Many-electron effects in acceptor-related radiative recombination of quasi-two-dimensional electrons

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The radiative recombination of electrons from a quasi-two-dimensional electron gas with holes localized on acceptors is investigated. The emission spectra for a single heterojunction are calculated including screening and the dynamical response of the Fermi sea. The line-shape analysis provides unambiguous identification of the Fermi-edge singularity in the emission spectra of the two-dimensional electron gas.

The radiative recombination of electrons in the conduction band with photoexcited holes in the valence band provides information about all occupied electron states in modulation-doped semiconductor microstructures, such as quantum wells, wires, and dots. This is to be contrasted with transport measurements which involve only states at the Fermi surface. The insight into electronic states from emission spectrum is, however, usually hindered by the nonequilibrium distribution of photoexcited holes. This problem can be circumvented by a selective doping of semiconductor microstructures with acceptors.[1] Acceptor-related recombination has been used in the study of ground-state properties of the metallic state,[1,2] incompressible liquid,[3] Wigner crystal,[3] and in hot carrier relaxation.[4] We shall concentrate on a metallic state here.

Typically when acceptors are present, a well-defined emission line below the band-to-band recombination spectrum appears.[1,2] This line corresponds to a recombination of an electron with a hole strongly bound to a negatively charged acceptor, and one can indeed focus on the electronic system. Unfortunately the many-electron response to a localized perturbation, such as recombination on an acceptor, is divergent and the line shape of the emission spectrum does not reflect the single-particle density of states in a simple way. This phenomenon is known as the Fermi-edge singularity[5] (FES). The Fermi-edge singularity in emission spectra of modulation-doped quantum wells has been observed by Sholnick *et al.*[6] It manifests itself in the enhancement of the emission spectrum at high energies, i.e., in the vicinity of the Fermi surface. This is to be contrasted with experimental emission lines associated with recombination on acceptors[2] which peak at low energies corresponding to transitions from the bottom of the conduction band. The explanation of the experiments of Skolnick *et al.* required holes to be localized by, e.g., potential fluctuations. Hence the experimental spectra of Kukushkin *et al.*[1] and Skolnick *et al.*[6] appear to be contradictory. We shall demonstrate that in fact both of these experiments measured the Fermi-edge singularity.

Since the standard perturbation theory based on the summation of an arbitrary class of diagrams (e.g., ladder diagrams) fails completely in the understanding of FES, nonperturbative methods are a necessity. We present here a nonperturbative calculation of the emission spectrum due to the recombination of electrons with a hole localized on an acceptor. The calculation includes screening, excitonic effects, shakeup of the Fermi sea, and the effect of the whole wave function.

We consider a single heterojunction with electrons occupying a single subband at the average position z_0 and acceptor at position z_h, away from quasi-two-dimensional electrons. The physical picture is very simple: prior to illumination our system consists of N electrons and a single negatively charged acceptor which acts as a repulsive scattering center for electrons. The single-particle states and energies for N conduction electrons in a heterojunction and one electron trapped on the acceptor are denoted by $|\lambda\rangle$ and e_λ, and $|h\rangle$ and $-\omega_a$, respectively. This is our final basis and the normal state one would like to probe optically. Upon illumination one electron is added to the conduction band and a hole from the valence band is localized by the negatively charged acceptor, making it a neutral, weak-scattering center. The single-particle states and energies of $N+1$ conduction electrons in a heterojunction in the presence of neutral acceptor are denoted by $|k\rangle$ and e_k.

The emission spectrum $E(\omega)$ involves the emission of a photon with frequency ω with one of the $N+1$ conduction electrons making a transition to the empty level (hole) localized on the acceptor. The annihilation of the hole changes the potential seen by all electrons in the conduction subband from charge neutral to a negatively charged acceptor. This makes the transition a many-electron effect. The emission spectrum $E(\omega)$ can be derived directly from Fermi's golden rule (we set $\hbar=1$):

$$E(\omega)=2\pi\sum_f \left|\left\langle \Psi_f(N+1)\left|\sum_{j=1}^{N+1} p_j\right|\Psi_i(N+1)\right\rangle\right|^2 \times\delta(E_i(N+1)-E_f(N+1)-\omega)\ . \quad (1)$$

The initial state $|\Psi_i(N+1)\rangle$ is the Slater determinant of $N+1$ particles occupying the $N+1$ lowest single-particle conduction states $|k\rangle$ in the presence of a neutral acceptor. The final states $|\Psi_f(N+1)\rangle$ are Slater determinants

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of $N+1$ particles with one particle occupying the localized acceptor level $|h\rangle$ and N particles occupying all possible single-particle states $|\lambda\rangle$, i.e., conduction states in the presence of a repulsive acceptor potential. The main difficulty in using the golden rule directly is the enormous number of final many-particle states, for which both matrix elements and energies have to be evaluated. This problem was circumvented by Combescot and Nozières,[7] Mahan,[8] and Ohtaka and Tanabe.[9] We give here a brief derivation.

The total interband momentum operator couples conduction states only to the localized acceptor state. Expanding final states with respect to the localized state and using the Fourier transform of a δ function allows us to rewrite Eq. (1) as

$$E(\omega)=2\,\mathrm{Re}\int_0^\infty dt\, e^{-i(\omega-\omega_{\max})t}E(t)\ . \tag{2}$$

The maximum photon frequency is given by the difference between the ground-state energies of $N+1$ particles before and after emission: $\omega_{\max}=E_i(N+1)-[E_f^0(N)+(-\omega_a)]$. Here $E_f^0(N)$ is the ground-state energy of the normal (final) state, i.e., without the photoexcited electron. The time-dependent emission spectrum $E(t)$ of course ensures that no frequencies larger than the maximum allowed frequency $\omega_{\max}$ contribute to the frequency spectrum. From Eq. (1) $E(t)$ is given by

$$E(t)=\sum_f\left|\sum_{n=1}^{N+1}m_{k_n}(-1)^n\langle\Psi_f(N)|\Psi_i^{k_n}(N)\rangle\right|^2 \times e^{-iE_f(N)t}e^{itE_f^0}\ . \tag{3}$$

In Eq. (3) transition matrix elements are expressed in terms of single-particle transition matrix elements $m_k=p_{vc}\langle h|k\rangle$ corresponding to transition from a conduction state $|k\rangle$ to a localized state $|h\rangle$. p_{vc} are conduction to valence momentum matrix elements and $\langle h|k\rangle$ is the overlap of the conduction and localized electron envelope wave function. Each single-particle matrix element is weighted by the overlap of initial and final N electron states. The initial state $\Psi_i^{k_n}(N)\rangle$ is formed from the $N+1$ lowest $|k\rangle$ states except for the state $|k_n\rangle$. It therefore represents a hole in the Fermi surface. Equation (3) can now be manipulated further to explicitly involve the summation over final states:

$$E(t)=\sum_{n=1}^{N+1}\sum_{n'=1}^{N+1}m_{k_n}(-1)^n\left\langle\Psi_i^{k_n}(N)\left|\sum_f\right|\Psi_f(N)\right\rangle \times e^{-iE_f(N)t}\langle\Psi_f(N)|\Psi_i^{k_{n'}}(N)\rangle \times m_{k_{n'}}(-1)^{n'}e^{itE_f^0}\ . \tag{4}$$

The summation over a complete set of final states can now be eliminated by introducing a final-state Hamiltonian H_f which produces a correct final single-particle spectrum. H_f is the sum of single-particle Hamiltonians h_f. Using the final-state Hamiltonian we can write formally

$$E(t)=\sum_{n=1}^{N+1}\sum_{n'=1}^{N+1}m_{k_n}(-1)^n\langle\Psi_i^{k_n}(N)|e^{-itH_f}|\Psi_i^{k_{n'}}(N)\rangle \times m_{k_{n'}}(-1)^{n'}e^{itE_f^0}\ . \tag{5}$$

The time-dependent overlap of N-electron wave functions of the initial basis propagated by the final-state Hamiltonian $\langle\Psi_i^{k_n}(N)|e^{-itH_f}|\Psi_i^{k_{n'}}(N)\rangle$ is equal to the determinant $D_{k,k'}$ of matrix Φ of order $N+1$ build out of matrix elements $\phi_{p,p'}$ ($\phi_{p,p'}=\langle p|e^{-ith_f}|p'\rangle$, with p,p' occupied) whose nth row and n' column have been deleted. Using the relationship between the inverse of matrix Φ and determinants $D_{k,k'}$ we obtain the Combescot-Nozières result[7] for emission:

$$E(t)=e^{iE_f^0t}\det[\Phi(t)]\sum_{k,k'\le k_F}m_k\Phi^{-1}_{k,k'}(t)m_{k'}\ . \tag{6}$$

The first term in Eq. (4) [$\det(\Phi)$] describes the shakeup of the Fermi sea due to the disappearance of the valence hole, while the last term describes vertex corrections, i.e., the scattering of the hole inside the Fermi surface by a repulsive potential in the final-state Hamiltonian. This scattering process is mediated by exchange of the photocreated hole with holes (empty states) above the Fermi surface. Equation (6) involves the states of the photoexcited system while one wants to measure the states of the final basis. This is done by transforming Eq. (6) into the final basis. We define matrix $G_{\lambda,\lambda'}$, which describes the propagation of the hole in the Fermi sea in the final-state basis as

$$G_{\lambda,\lambda'}(t)=\sum_{k,k'\le k_F}\langle\lambda|k\rangle\Phi^{-1}_{k,k'}(t)\langle k'|\lambda'\rangle\ ,$$

where $\langle k|\lambda\rangle$ are the overlap matrix elements between the initial and final single-particle states. Using the identity

$$\varphi_{k,k'}=\sum_\lambda\langle k|\lambda\rangle e^{-ie_\lambda t}\langle\lambda|k'\rangle\ ,$$

the relationship between the initial matrix elements m_k and final basis matrix elements m_λ given by $m_k=\sum_\lambda m_\lambda\langle\lambda|k\rangle$, and the identity $\det(\Phi)=\exp\{\mathrm{Tr}[\ln(\Phi)]\}=\exp[-iC(t)]$, a set of *nonlinear* differential equations for the time evolution of the vertex (G) and self-energy (C) functions can be derived:

$$\begin{aligned}\frac{\partial}{\partial t}G_{\lambda,\lambda'}(t)&=-ie_\lambda G_{\lambda,\lambda'}(t)\\&\quad+i\sum_{\lambda''}G_{\lambda,\lambda''}(t)e_{\lambda''}G_{\lambda'',\lambda'}(t)\ ,\\\frac{\partial}{\partial t}C(t)&=2\sum_\lambda e_\lambda G_{\lambda,\lambda}(t)\ .\end{aligned} \tag{7}$$

The final expression for the emission[10] function $E(t)$ is now given simply in terms of the vertex G and self-energy corrections C:

$$E(t)=e^{iE_f^0t}e^{-iC(t)}\sum_{\lambda,\lambda'}m_\lambda e^{+ie_\lambda t}G_{\lambda,\lambda'}(t)m_{\lambda'}\ . \tag{8}$$

An important consequence of working in the final-state

basis is that *all* single-particle states of the final basis contribute to the frequency spectrum of the emission $E(t)$, irrespective of whether they are occupied or empty in the final ground state of the system, i.e., in the absence of the hole. The filling of phase space of initial states enters via the initial condition for the matrix $G(0)$:

$$G_{\lambda,\lambda'}(0)=\sum_{k\le k_F}\langle\lambda|k\rangle\langle k|\lambda'\rangle \ .$$

The overlap matrix elements $\langle k|\lambda\rangle$ between the initial and final states are solutions of the Wannier equation:

$$e_k\langle k|\lambda\rangle+\sum_{k'}V_{k,k'}\langle k'|\lambda\rangle=e_\lambda\langle k|\lambda\rangle \ . \tag{9}$$

The interaction $V_{k,k'}$ is the change in the one-electron potential between initial and final bases. This change corresponds to a repulsive scattering potential due to the screened charge of the hole localized on an acceptor. The matrix element is written in the initial basis, i.e., basis of the neutral acceptor.

We now apply our theory to the single heterojunction with only lowest subband occupied. The electron states in the absence of an acceptor can be written as a product of plane waves and the lowest subband wave function, which we take to be in the Stern-Howard form: $\xi_0(z)=\sqrt{b^3/2}ze^{-bz/2}$. The average position of the electron layer is given by $z_0=3/b$. The parameter b is obtained from variational calculations as a function of electron density. The acceptor and the hole are located away from the electron layer at position z_h. The effect of electrons and heterojunction structure on the hole is very small and we treat the acceptor as a bulk acceptor. We expand the hole wave function in terms of Gaussians and retain only a single term in a first approximation. The hole wave function is therefore written as $\Psi(x,y,z)=(2/\pi a^2)^{3/4}\exp[-(x^2+y^2+z^2)/a^2]$ with parameter a playing the role of the effective hole radius. Since the hole screens the charge of the acceptor very effectively, it is a very good approximation to assume a perfect screening condition, i.e., the initial basis is the basis in the absence of an impurity. In this approximation the matrix elements of the hole potential can be written as $V_{k,k'}=V(q)f(q)F(q)$, with $q=|k-k'|$ and $V(q)$ the statistically screened interaction. $F(q)$ is a standard electron-hole form factor, which can be well approximated by $F(q)=\exp(-q|z_0-z_h|)$, and $f(q)=\exp(-q^2a^2/8)$ is the Fourier transform of the in-plane hole charge density. $V(q)$ is the screened Coulomb interaction.[10] In a similar way matrix elements m_k can be effectively approximated by

$$m_k=m_0(2\pi a^2b^6/4)^{3/4}z_he^{-bz_h/2}\exp(-k^2a^2/4) \ ,$$

where m_0 is a constant. Hence the coupling of conduction electron states with the hole is largest at the bottom of the band and decreases rapidly as the energy of the carriers increases.

In Fig. 1 we show calculated emission spectra for Fermi energy $E_F=2$ Ry. Energies are measured in bulk rydbergs and the Fermi energy is related to the carrier density n by $n=E_F/2\pi a_0^2$ where a_0 is the effective Bohr radius. For GaAs, $a_0=100$ Å and $E_F=2$ Ry corresponds

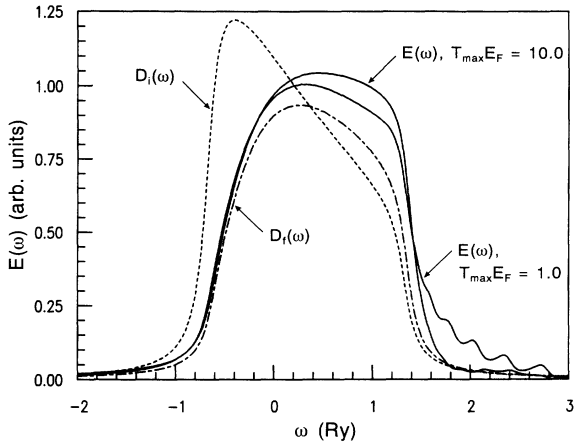

FIG. 1. The emission spectrum for $E_F=2.0$ Ry (solid lines) for two different times of evolution. The joint density of states of the initial D_i and final D_f bases are also shown. The acceptor is located at $z_h=1.5a_0$.

to a carrier density $n=3\times10^{11}$ cm^{-2}. The emission spectra were calculated[10] by propagating vertex and self-energy corrections up to a maximum time $T_{\max}$. The emission spectra (solid lines) for two different time cutoffs $T_{\max}E_F=1.0$ and 10.0 are shown. They are compared with the joint single-particle density states of conduction electrons and a hole in the absence of impurity D_i and in the presence of impurity D_f, $D_i(\omega)=\sum_{k\le k_F}m_k^2\delta(e_k+\omega_a-\omega)$ and $D_f(\omega)=\sum_{\lambda\le\lambda_F}m_\lambda^2\delta(e_\lambda+\omega_a-\omega)$, respectively. As illustrated in Fig. 1, the effect of self-energy and vertex correction is to modify the final joint density of states D_f in the vicinity of the Fermi energy. The two different times illustrate nicely the fact that as the system relaxes after emission of a photon, the removal of forbidden frequencies $\omega>\omega_{\max}$ corresponds to the buildup of the oscillator strength below $\omega_{\max}$, i.e., the formation of the Fermi-edge singularity. We have broadened the singularity by adding an imaginary part of 0.1–0.2 Ry to the frequency ω. The final joint density of states D_f approximates reasonable well the line shape of the spectrum apart from FES. The joint density of initial states D_i is a poor approximation to the line shape. However, the total integrated intensity of the emission line $E(t=0)$ is given exactly by the total integrated intensity of the initial joint density of states D_i. In our model this is given by $E(0)=m_0^2\xi_0^2(z_h)\sqrt{2\pi a^2}[1-\exp(-E_F)]$, where E_F is given in bulk rydbergs. This allows a very easy interpretation of the integrated intensity as a function of carrier density, in contrast to a complicated line shape. This should be compared with carrier densities extracted from the width of the emission line.

The dependence of the emission spectrum on the density of free carriers is shown in Fig. 2. The width of the line increases and the intensity decreases with increasing carrier density. The excitonic effects are reduced due to screening and the fact that as the electron density increases, the electron layer moves away from the hole. The emission line increasingly resembles the final joint density of states D_f, with small remnants of the FES at

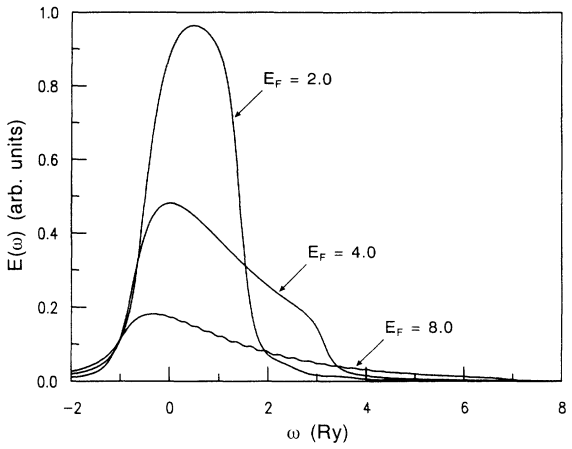

FIG. 2. The emission spectrum for 3 different carrier densities corresponding to Fermi energies $E_F=2,4,8$ Ry and fixed hole position at $z_h=1.5a_0$.

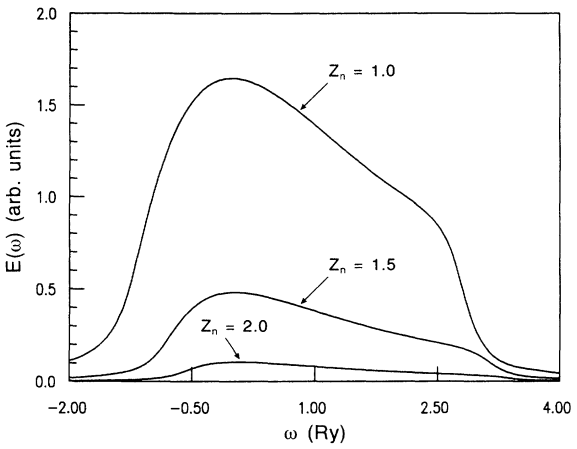

FIG. 3. The emission spectrum for a fixed carrier density corresponding to Fermi energy $E_F=4.0$ Ry for different positions of the hole.

higher energies. A similar effect is obtained by moving the acceptor away from the electron layer. The emission spectra for three different hole positions $z_h=1.0,1.5,2.0$ and carrier density corresponding to $E_F=4.0$ Ry are shown in Fig. 3. The average position of the electron layer is $z_0=0.7a_0$.

Let us now comment on the experiments of Skolnick *et al.*,[6] Kukushkin *et al.*,[1] and Petrou *et al.*[2] In Ref. 6 it was assumed that in the initial state the hole was localized by, e.g., potential fluctuation. Hence the initial state was that of a positively charged localized hole in the electron gas, while in the final state electrons relaxed to their unperturbed states. This relaxation is equivalent to the switching of a repulsive hole potential in the basis of initial states. In acceptor related transitions the initial state is unperturbed and the final state contains electrons relaxed around the negatively charged repulsive center. Hence the only difference is in the single-particle density of states and both experiments measure the Fermi-edge singularity.

In summary, the emission spectra associated with recombination of electrons in a quasi-two-dimensional gas with holes localized on acceptors have been calculated. The calculation shows that when the single-particle joint density of states is properly taken into account, the spectra show an enhancement in the vicinity of the Fermi energy due to correlations of many electrons with the localized hole.

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