Infrared and Polarization Anomalies in the Optical Spectra of Modulation-Doped Semiconductor Quantum-Well Structures

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(Received 19 June 1985)

We investigate the physical processes which determine the optical spectra of modulation-doped semiconductor quantum-well structures as a function of doping. We argue that even at low doping concentrations, excitons are heavily dressed by charge- and spin-density excitations of the Fermi sea, implying, already in that limit, a Stokes shift between emission and absorption. For all doping concentrations, the charge and spin polarization of the Fermi sea due to many-body effects leads to an enhancement of the symmetry breaking in quantum-well structures, which may explain polarization anomalies observed in luminescence.

PACS numbers: 78.65.Jd, 71.35.+z, 78.55.Ds

In the last few years, there has been growing interest, from both a device as well as a physics point of view, in the properties of n- and p-type modulation-doped semiconductor quantum-well structures. These systems provide idealized realizations of (quasi) two-dimensional Fermi systems. Most importantly, the carriers are spatially separated from the dopants, so that the mobilities are extremely high. In spite of the wealth of experimental information obtained predominantly by luminescence and excitation spectroscopy,[1,2] a clear understanding of the optical properties of these systems is still lacking. Regrettably, little effort has been made in identifying and separating the one-particle (band-structure) effects from the many-particle aspects of the problem.[3] In this paper we will concentrate on the latter. In particular, we will demonstrate their relevance to the dramatic circular-polarization reversal of the photoluminescence at low temperatures from n-type modulation-doped GaAs quantum-well structures when resonantly excited with circularly polarized light at the $n=2$ heavy-hole exciton transition.[1]

A number of n-type modulation-doped GaAs/$Al_xGa_{1-x}As$ quantum-well structures (QWS) grown by molecular-beam epitaxy demonstrate this effect. The layers were grown on (100) Cr-doped substrates under conditions adjusted to yield abrupt interfaces and high mobilities. Excitation is obtained from a tunable dye laser at normal incidence to the layers and the photoluminescence (PL) is detected in the reflection direction 24° off normal incidence. Circular-polarization excitation and detection techniques are employed.

Spectra at 5 K are shown in Fig. 1(a) for an undoped multiple QWS with wells ≈ 120 Å wide clad with $x \simeq 0.3$ alloy barriers. Of main interest are the excitation spectra of the PL and its polarization. The excitation spectra are obtained by scanning of the incident photon energy with the detection set at the peak of the PL curve, so that emission is from what is usually thought to be the $n=1$ heavy-hole exciton, E_{1h}. The transitions of interest are labeled E_{nj} where n denotes the subband quantum number and j the hole character, either light (l) or heavy (h). The polarization spectrum exhibits positive peaks for the heavy-hole (hh) transitions and a decreased, or even negative, polarization for the light-hole (lh) transition. A positive polarization indicates the same sign as the incident polarization.

The polarization spectrum in Fig. 1(a) is consistent with that expected from the valence-band structure of GaAs QWS, but markedly different from that shown in Fig. 1(b) for n-type modulation-doped GaAs QWS with an electron concentration $n \simeq 2\times 10^{11}$ cm^{-2}. In this case, the polarization is negative (−5%) for resonant excitation at $E=E_{2h}$. This behavior is characteristic of GaAs QWS with $n \gtrsim 10^{11}$ cm^{-2}. When the temperature is increased to 20 K, the anomalous decrease in polarization for $E \simeq E_{2h}$ is greatly reduced or even absent in some cases. Furthermore, the electron polarization accompanying the anomaly is rather insensitive to a weak transverse magnetic field (≈ 4.5 kG), whereas in the low-doping limit it usually exhibits the characteristic depolarization behavior (Hanle effect).[1] Also note the increased Stokes shift between emission and absorption in Fig. 1(b).

In order to explain these phenomena, we depart from the rigid-Fermi-sea picture traditionally used to describe the optical spectra of doped semiconductors. We illustrate the simplest consequences of dynamical effects by first discussing the absorption process. We then explain how these effects may resolve the polarization anomaly in luminescence excitation. (We limit ourselves to n-type samples; similar considerations can be carried over to p-type materials and we will only comment on possible differences.) For clarity, we begin by considering some of the physics already contained in the rigid-Fermi-sea picture in which the electrons are not allowed to respond dynamically to the

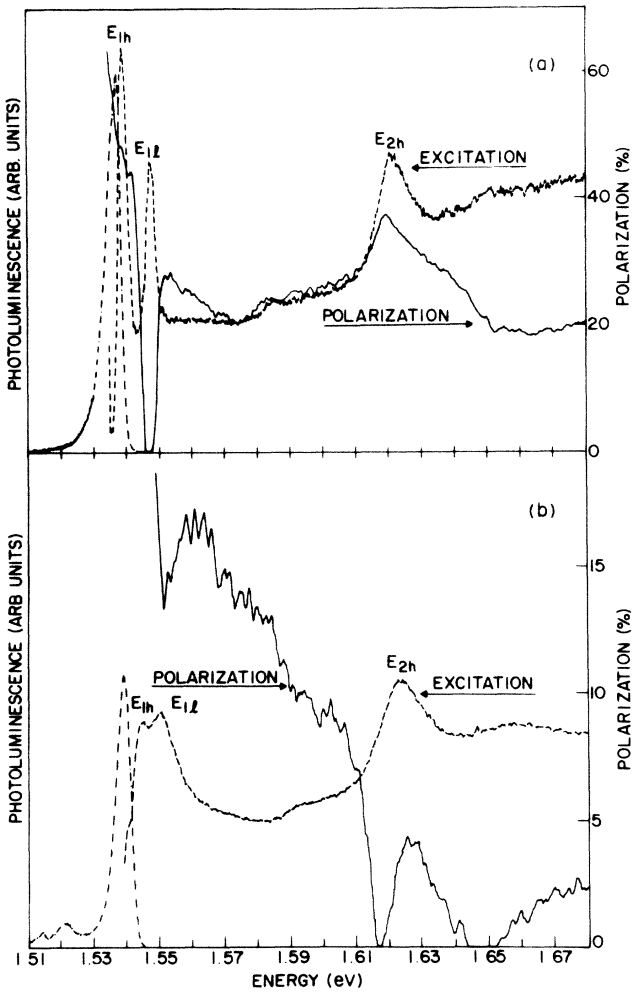

FIG. 1. Photoluminescence (dot-dashed curve), excitation (dashed curve), and circular-polarization (solid curve) spectra of GaAs/$Al_xGa_{1-x}As$ quantum wells ($L \simeq 120$ Å, $x \simeq 0.3$) excited with σ^+ circularly polarized light at 5 K. Sample (a) is undoped and exhibits the usual polarization behavior, the polarization increases at the $n=1$ and 2 heavy-hole exciton transitions (E_{1h} and E_{2h}) and decreases markedly or even goes negative (σ^-) at the $n=1$ light-hole exciton transition E_{1l}. Sample (b) is n-type modulation-doped with $n \simeq 2\times 10^{11}$ cm^{-2}. The emission peak is red shifted from E_{1h} by ≈ 6 meV compared with ≈ 1 meV in (a). The phase of the detected polarization signal was changed by 180° between 1.617 and 1.647 eV, so that in this case the polarization actually becomes negative (σ^-) at E_{2h}.

sudden appearance (or disappearance) of the hole in the course of an optical transition.

At low temperatures and doping concentrations, and for frequencies close to the effective band gap, the absorption spectrum for QWS displays the usual atomic excitons, as demonstrated in Fig. 1(a). With increasing doping, exchange and correlation effects lead to a renormalization of the one-particle Hartree energies (a decrease of the band gap) and a weakening of the exciton binding energy. Usually, there is a large degree of cancellation of these two effects (i.e., of self-energy and vertex corrections), so that the absolute exciton energy changes less rapidly than the band gap.[4] In quasi two-dimensional systems one expects that atomic bound states survive up to higher free carrier concentrations than in the bulk. This is mainly because in two dimensions one only needs an arbitrarily weak potential to support a bound state and, moreover, because the screening saturates as a result of the constant density of states. At low temperatures, when $k_F a_0$ becomes of order unity (where k_F is the Fermi wave number and a_0 is the exciton Bohr radius) the atomic exciton unbinds[5-7] and one must think in terms of one hole correlated with the entire Fermi sea. In that limit the analog of the atomic exciton is the so-called Mahan exciton,[8] which can be thought of as a bound state with respect to the Fermi level.

Once one allows for the dynamical response of the Fermi sea to the appearance of the hole, the picture given above changes drastically. Qualitatively, this can be most easily understood by following the elegant discussion of the soft x-ray spectra of metals due to Schotte and Schotte.[9] In three dimensions this poor-man's method yields the rigorous results of Combescot and Nozières.[10]

In analogy with Refs. 9 and 10, we consider a localized (i.e., infinitely massive) hole with spin coupled through contact direct (U) and exchange (J) interactions to the Fermi sea. Its sudden appearance in the course of the absorption process leads to a readjustment of all the electrons into a new (scattering) state, in local equilibrium around the hole. In the (asymptotically) weak-U and -J limit and within the Schotte-Schotte picture, this readjustment shows up as an infinite number of bosonlike low-energy charge and spin excitations of the Fermi sea. In order to allow for the possibility of excitons, we add a bound-state contribution orthogonal to the electron scattering states. The optical absorption probability $I(\omega)$ can then be easily calculated along the lines described in Ref. 9. The details are straightforward and will be presented elsewhere.[11] Here, we only summarize the results.

For an infinite hole mass, the absorption spectrum has the form

$$I(\omega) \sim A(\omega-\omega_1)^{-\alpha_1} + B(\omega-\omega_2)^{-\alpha_2}.$$

The two threshold energies ω_1 and ω_2 can be obtained from Fumi's theorem[12] and correspond, in the low-doping limit, to the onset of the exciton and continuum absorption, respectively. The exponents α_1 and α_2 are in agreement with Hopfields's rule of thumb[13] and can be written in terms of the number of electrons required to screen the hole potential, as determined by the Friedel sum rule. Therefore, even for low doping concentrations ($k_F a_0 \ll 1$), the exciton shows up in

absorption spectra as an infrared power-law singularity with an exponent close to 1, accompanied by a weak secondary threshold at the Fermi level. In the high-doping limit, the remnant of the atomic exciton at the Fermi level (the Mahan exciton) is unstable. Instead, one finds an infrared power-law singularity, which has the same origin as the soft x-ray singularity in metals[14,15] first discussed in the pioneering work of Mahan.

In principle, in contrast to three dimensions, in two dimensions one always has a bound state. However, in the high-doping limit it must have negligible spectral weight and perturbation theory (on the level of summing of parquet diagrams) should become valid. Thus, in two and three dimensions there is a single power-law singularity, the nature of which changes continuously from the exciton dressed with an infinite number of charge- and spin-density-wave excitations in the low-doping limit to the usual Fermi-level singularity in the high-doping limit.

In the case of a finite hole mass, the recoil of the hole will smear the singularity over a frequency range $(m_e/m_h)\epsilon_F$, where m_e and m_h are the electron and hole effective masses. At high doping concentrations, whenever this effective bandwidth is much larger than the Coulomb energy (as determined by the binding energy of the Mahan exciton), the singularity is completely removed. At low frequencies, the absorption spectrum is then determined by indirect transitions.[11] Even at low doping concentrations there will be a Stokes shift between emission and absorption. This shift increases with increasing doping and and changes continuously into the usual Burstein-Moss shift in the high-doping limit. Exactly this behavior, up to now unexplained, has been reported in Ref. 1 and can also be seen in Fig. 1.

We argue that the many-body effects described above may also explain the circular-polarization anomaly. We offer two possible explanations for this effect. The first relies on the fact that in the absorption process, in the presence of a strong exchange interaction between the conduction electrons and the $n=2$ hh, the Fermi sea instantaneously builds up a collective moment parallel to the moment of the hole and of the same magnitude. This is analogous to the electron-spin polarization between two successive impurity-spin flips in the Kondo effect. In our case, however, the hole moment decays on a much faster time scale than the Fermi-sea polarization.[16] Provided that the moment of the photoexcited electron relaxes together with that of the hole, the net polarization of the luminescence is then due to the polarization of the Fermi sea and is opposite to that of the pump beam. This then resolves the polarization anomaly.

This mechanism requires a large exchange between the $n=2$ hh and the $n=1$ electrons, and an even larger one between the photoexcited $n=2$ electron and the $n=2$ hh. It is clear, however, that for the structures studied here the usual exchange matrix element calculated from one-particle states is small. A sufficiently large exchange interaction could only arise from many-body effects; the irreducible exchange interaction between the $n=2$ hh and the $n=1$ Fermi sea involves, for example, virtual intermediate-state interactions between the $n=1$ lh and the conduction electrons. In the case of an infinite hole mass, these intermediate-state processes are singular in the same way as the (final-state interaction) processes discussed above for absorption. (Similar effects were studied by Nozières and Abrahams in the context of Raman scattering in metals.[17]) Also, because of the symmetry of the subband wave functions, the effective exchange between the $n=2$ hh and the $n=2$ electron due to the same mechanism would be indeed larger. Since the temperature at which the polarization anomaly goes away is approximately 20 K, the required exchange can be as small as 1 meV. This would be 1 order of magnitude smaller than the direct Coulomb energy in the low-doping limit as determined by the atomic exciton binding energy. However, for high doping, exchange is strongly enhanced by ferromagnetic fluctuations (corresponding to the Stoner enhancement in three dimensions) and can become of the same order of magnitude as the "screened" direct interaction.

A second possibility for an understanding of the polarization anomaly relies on the fact that particle-hole pair excitations of the Fermi sea couple (i) different hole states in momentum space within the same subband, and (ii) different subbands. Even in process (i) the states possess different symmetries because of the band mixing and thus the "shakeup" of the Fermi sea increases the symmetry breaking in QWS. Moreover, since the $n=1$ lh and the $n=2$ hh bands are very close in energy, a strong many-body hybridization [process (ii)] is to be expected for these two bands in the course of the absorption process. With increasing doping, the $n=2$ final hole state may therefore change from heavy-hole to light-hole character. Process (i) was first invoked by Sham[3] to explain the occurrence of an unexpectedly strong polarization component normal to the layer in the emission spectrum of heavily doped quantum wells.

Preliminary data show that there is no polarization anomaly at all in p-type samples and also no anomaly for excitation at the $n=3$ hh transition in n-type samples. However, the shakeup in absorption should lead to similar effects in p- and n-type samples for excitation at the $n=2$ hh transition while the shakeup in emission does not depend on the excitation frequency. Thus, the experimental findings mentioned above appear to rule out a large contribution to the anomaly from shakeup in both absorption and emission (as long

as these processes are taken as independent). Finally, we note that all experimental observations are consistent with the collective nature of the Fermi-sea polarization implied by our first mechanism. In particular, it explains the weak Hanle effect mentioned above. More importantly, in p-type materials this mechanism is absent as a result of the short spin-relaxation time of the holes in the Fermi sea. All that remains is the uncertainty in the order of magnitude of the exchange interaction between conduction electrons and the $n=2$ hh.

We have also calculated the absorption spectrum in the high-doping limit for frequencies close to the indirect absorption edge. As has been discussed above, a finite hole mass will smear the Fermi-level singularity completely in that limit. The lowest optical (Auger-type) transitions, from the top of the $n=1$ heavy-hole band to the Fermi level, are then assisted by particle-hole pair excitations with almost no energy and with momenta close to the Fermi momentum. Note that away from the Fermi level the quasiparticle picture for the electrons breaks down and thus electron-electron interactions have to be included and, moreover, treated on the same footing as the electron-hole interactions. By summing the six diagrams second order in the interactions, we obtain

$$I(\omega)\sim[(1+m_e/m_h)U(k_F,0)+J(k_F,0)]^2[\rho(\omega-\omega_I)/(m_e\epsilon_F/m_h)]^3,$$

where ω_I is the indirect threshold and ρ the electron density of states; $U(k,\omega)$ and $J(k,\omega)$ are the effective direct and electron-hole exchange interactions. We note that as a result of the so-called "exchange hole" all exchange contributions are exactly canceled by the corresponding direct contributions.

A detailed comparison between theory and experiment will involve extensive calculations which would incorporate the actual band structure. We note that inclusion of the effects of a finite hole mass is a difficult task; this is mainly because for frequencies away from the indirect threshold the problem can never be treated by simple perturbation theory, irrespective of the value of m_e/m_h.[11] Anyway, we expect that a great deal of the new qualitative physics we discussed will survive a more complete treatment.

We would like to thank A. Pinczuk and, especially, L. J. Sham for helpful discussions. Two of us (A.E.R. and S.S.-R.) would like to thank J. C. Phillips for stimulating their interest in the polarization anomaly. The high-quality quantum-well structures studied were provided by A. C. Gossard and W. Wiegmann.

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