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THE EFFECTS OF PHOTOGENERATED FREE CARRIERS AND MICROWAVE ELECTRON HEATING ON EXCITON DYNAMICS IN GaAs/AlGaAs QUANTUM WELLS

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Abstract—Modulation of the resonantly excited exciton photoluminescence of undoped GaAs/AlGaAs quantum wells is obtained in two ways: (a) by photoexcitation with an additional laser beam at above bandgap energies and (b) by microwave irradiation. At low temperatures the two types of modulation have an opposite effect on the PL lineshape. We show that photoexcitation generates separately localized electrons and holes, and their interaction with localized excitons accelerates exciton transfer from weakly into strongly localized states. The excess exciton energy is imparted to the localized electron (or hole). On the other hand, under microwave modulation, free hot electrons activate localized excitons into high energy states and the exciton spectral diffusion towards deeper states is slowed down.

INTRODUCTION

The photoluminescence (PL) band of undoped GaAs/AlGaAs quantum wells (QW) at low temperatures is due to radiative recombination of (e1:hh1)1S excitons. Its lineshape is affected by the rate of exciton terminalization between the localizing centers that are due to the interface potential fluctuations. The exciton transfers from QW regions where it is weakly localized (corresponding to the high energy part of the band), to regions of stronger localization[1]. The competition between this transfer rate and the exciton radiative recombination rate determines the exciton PL lineshape. The excess kinetic energy (between the high- and low-energy exciton states involved in the transfer) is usually taken up by acoustic phonons. Any additional processes that modify the exciton dynamics can thus be studied by monitoring the variations in the exciton PL lineshape. The aim of this study is to show that electron–exciton scattering processes have a pronounced effect on the exciton lineshape. We compare the effect on the lineshape of free electrons that are heated by microwave radiation with that due to localized electrons (or holes) that are generated by photoexcitation above the (e1-hh1) bandgap.

We introduce the modulation effects by showing the PL spectra of the (e1:hh1)1S exciton band observed in high quality, undoped GaAs/AlGaAs multiple quantum wells (MQW) at $T = 2$ K. Recently we found that an extremely weak above bandgap photoexcitation causes a strong modulation of the resonantly excited exciton PL band and a decrease in the LO-phonon resonant Raman scattering (RRS) intensity[2,3]. In these experiments the QW is irradiated by two lasers, both impinging on the same spot: a chopped laser beam with $E_{L_2} > E_{gap}$ and a cw laser with energy E_{L_1} resonant at various energies within the (e1:hh1)1S exciton band. The intensities of each laser vary in the range of 0.02–2 W cm^{-2}. A typical PL modulation effect is demonstrated in Fig. 1(a) where the PL spectrum is either resonantly excited (at E_{L_1} only, dashed line) or simultaneously excited by two laser beams (at E_{L_1} and E_{L_2}, solid line). The difference spectrum demonstrates that the PL intensity at low energies increases while it decreases at the PL peak. It is observed that the effect depends strongly on the chopping frequency of E_{L_2}.

In contrast, an opposite sign of the modulated PL spectrum is observed under microwave radiation heating[2,4]. This effect is due to hot electrons and it is demonstrated in Fig. 1(b) where the PL spectra, excited at $E_{L_2} = 1.96$ eV (dashed line) and under microwave radiation (solid line) are shown. Their difference spectrum is also presented in Fig. 1(b).

In this work we report on the temporal and spectral dependencies of these modulation effects and analyze them in terms of the exciton interaction with either localized electrons or hot electrons, respectively.

EXPERIMENT

Several undoped GaAs/AlGaAs MQWs were studied, with well widths ranging between 50 and 100 A and barrier widths of 200 or 100 A. They were all grown by molecular beam epitaxy on [001]-oriented GaAs substrates.

In order to study the temporal response of the excitons to electron–hole photogeneration, a pulsed He-Ne laser beam (a pulse width 0.2 ms is obtained by an acousto-optical modulator) is used for excitation and a photon counter with a scanned gate of 0.01 ms is used for analyzing the time-resolved PL kinetics.

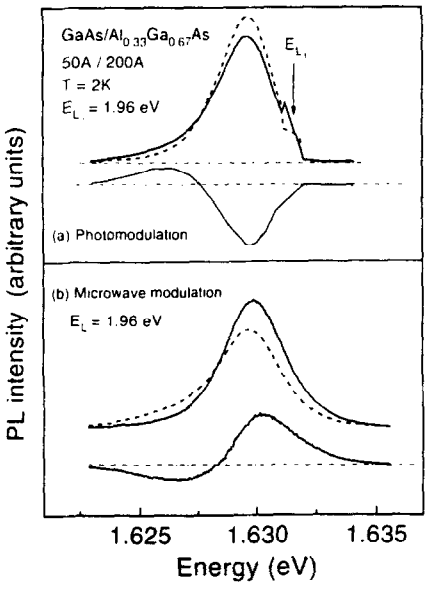

Fig. 1. The effects of above bandgap photoexcitation (a) and of microwave radiation (b) on the (e1:hh1)1S PL spectrum. (a) The PL spectrum is resonantly excited by E_{L_1} only (dashed line), and simultaneously by two laser beams (E_{L_1} and E_{L_2}) (solid line). Their difference is the photomodulated spectrum (lower curve). (b) The PL spectrum (dashed line), and that obtained under microwave radiation with power of 20 mW (solid line). Their difference is the microwave modulated spectrum (lower curve).

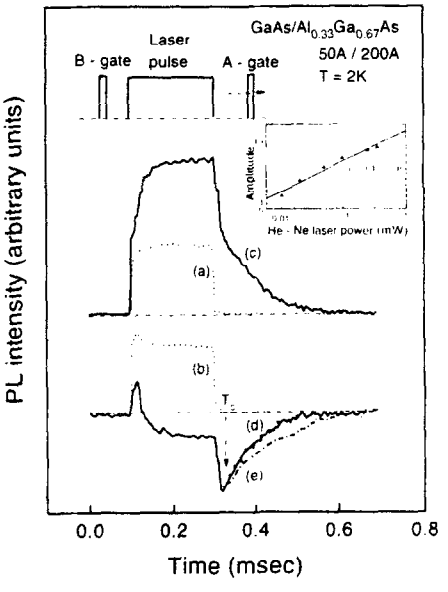

Fig. 2. The temporal behavior of the PL intensity monitored at two energies within the (e1:hh1)1S PL band: (a,c)—$E_m = 1.627$ eV; (b,d)—$E_m = 1.630$ eV; (a) and (b) are the response to the pulsed photoexcitation (at $E_{L_2} = 1.96$ eV only); (c) and (d) are the response when a cw resonant excitation (at $E_{L_1} = 1.634$ eV) is added. (e) PL decay when the cw resonant excitation intensity is 10 times less than that on curve (c). Insert: The dependence of the modulation amplitude when the gate is set at T_0 on the He-Ne laser power (I_{L_2}). α is an exponent of the approximate dependence.

Figure 2 shows the PL kinetics monitored at the low PL energy (a) and at the PL peak (b) under pulsed excitation at $E_{L_2} = 1.96$ eV. When a cw resonant excitation (at $E_{L_1} = 1.634$ eV) is added, the PL kinetics change. Curves c and d are obtained by taking the difference signals at the photon-counter gates A and B (shown schematically in Fig. 2). Gate A is scanned while B is fixed (before the He-Ne laser pulse turns on). Positive modulation (as shown in curve c) means that the PL intensity increases while a negative one (curve d) corresponds to an intensity decrease. Both curves show that the PL modulation effect decays a very long time after the He-Ne laser pulse terminates. The decay time varies with the cw resonant photoexcitation intensity. It decreases from 0.3 to 0.2 ms when the cw light intensity increases from 0.2 to 2 W cm^{-2} (curve e). The decay time is nearly independent of the above bandgap light intensity: the dots (marked on top of curve d) show the normalized PL decay at He-Ne laser intensity 10 times less than that used for curve d. The modulation efficiency of the cw resonantly excited PL increases with He-Ne laser light intensity: the amplitude of the negative signal measured at the gate position T_0 is shown in the inset of Fig. 2; the MPL amplitude $\propto I_{L_2}^{\alpha}$, where $\alpha = 0.45 \pm 0.05$.

Typical PL and PL excitation (PLE) spectra for two 50 A MQW samples are shown in Fig. 3(a, b) and for a 70 A MWQ—in Fig. 4(a). These QWs have different PL linewidths and Stokes shifts between the PL band peak and that of the PLE band. The PLE spectra are monitored at the peak or at the low energy tail of the PL band and are normalized so that the photoexcitation intensities in the exciton regions are equal. A clear difference between the PLE spectra

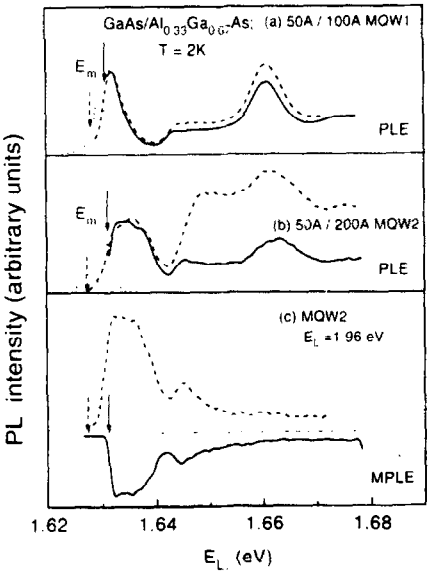

Fig. 3. (a), (b) PL (dots) and PLE spectra detected at the low PL side (dashed curve) and at the PL peak (solid curve) for MQW1 and MQW2. (c) MPLE spectra detected at the same energies as in (b).

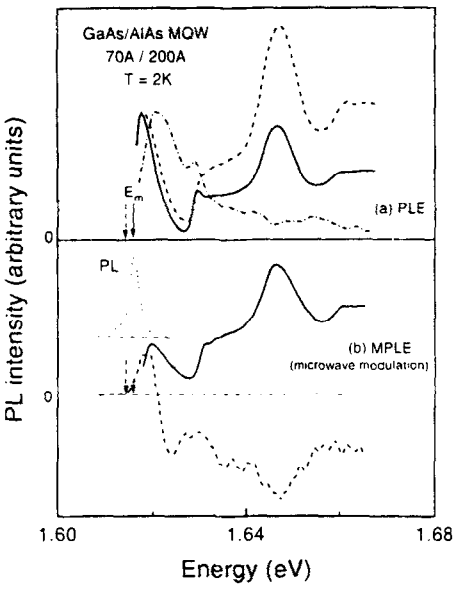

Fig. 4. (a) PLE spectra monitored at the exciton PL peak (solid), at the low exciton energy tail (dashed) and at the LO-phonon sideband (PSB) (dash dot). (b) Microwave modulated PLE spectra detected at the same positions of the exciton PL spectrum as on (a), respectively.

monitored at the high and the low PL energies is observed. When the excitation energy exceeds the (e1-hh1) bandgap, the PL intensity at the low energy side increases. This behavior is found in all QWs under study and it shows that the density of the strongly localized excitons increases sharply for $E_{L_1} > E_{gap}$. This result could be understood as an exciton mobility increase as the exciton kinetic energy increases or as due to the effect of free electron–hole pair generation on the exciton diffusion. In order to understand this we study how the photomodulation depends on the exciton energy.

In Ref.[3] we have shown that the photomodulation occurs only if the energy of the modulating excitation —E_{L_2}—exceeds the (e1-hh1) bandgap and that its efficiency depends on the energy of cw resonantly generated excitons by E_{L_1}. Figure 3(c) shows the dependencies of the modulation amplitude on the cw photoexcitation energy (MPLE spectra) monitored at the same PL energies, as in Fig. 3(b). These curves are measured at a fixed gate position after the He-Ne light pulse terminates by scanning E_{L_1}. As can be seen, an additional electron-pair generation affects mainly the resonantly excited excitons (in the spectral range of 1.63–1.64 eV) and it decreases as the monitored PL energy increases.

In high quality MQWs a LO-phonon sideband (PSB) is observed[5,6]. The modulation of the PSB luminescence reveals a temporal behavior different than that of the exciton PL. The photoinduced PSB luminescence intensity quenching can be characterized by two lifetimes: a fast response on additional photo-excitation (it is faster than our time resolution of 10^{-5} s), and a slow one which is approximately the same as that of exciton PL.

The PLE spectrum monitored at the PSB position is shown in Fig. 4(a) and is compared with that monitored at the exciton PL band peak. It is seen that the PLE spectrum peak of the PSB is located essentially below the bandgap and it shifts to higher energy in comparison to that of the exciton PL line. The photoexcitation efficiency of these PSB decreases above the band gap. We observed that the PL intensity of the entire PSB decreases under microwave radiation, as well as under photomodulation[3,6].

The hot electrons also induce an exciton PL spectral redistribution [see Fig. 1(b)]. The microwave radiation modulation has a decay time less than 10^{-5} s. The amplitude of the microwave induced modulation monitored at the PL peak band and in its low energy tail (MPLE spectra) are shown in Fig. 4(b). At the PL peak and at the high PL energies the modulation is positive and it strongly increases when the photo-excitation energy exceeds the band gap (since free electrons and holes are generated). The MPLE spectrum is more complicated when it is monitored at the low energy part of the PL spectrum [dashed curve in Fig. 4(b)]. There, the PL intensity increases when the photogeneration occurs directly into localized exciton states having energies that are close to the monitored energy (positive modulation) and it decreases when the photoexcitation is far from the monitored energy.

Finally we observe that the photo- and microwave modulation amplitude and decay time decrease as the temperature increases above 10 K and disappear above 20 K.

DISCUSSION

We observe that the exciton spectral diffusion within the inhomogeneously broadened (e1:hh1)1S band is affected by both photomodulation and microwave modulation. The model describing these phenomena is based on exciton–electron (or hole) scattering with a distinction between the electronic states involved in each case.

From the slow photomodulation decay we conclude that this phenomenon must involve separately localized electrons and holes since only such carriers can have long lifetimes. Also, the modulated PLE spectra demonstrate that the lower is the exciton mobility (in the low energy PLE spectral region) the stronger is the modulation effect. Therefore, the localized electrons (or holes) affect mainly the spectral diffusion of localized excitons in the spatially fluctuating interface potential. In our previous work[3] we suggested that a new channel for exciton transfer is induced by free carrier-exciton scattering that adds up to the phonon assisted exciton tunneling between the states that arise from interface roughness. Our present findings show that the interaction between localized

excitons and localized electrons results in an increase of the exciton spectral diffusion. The decrease of the modulation amplitude and decay time above 10 K means that this phenomenon is due to localization of the excitons and electrons (holes) in the interface fluctuating potential.

Under above bandgap illumination free electrons and holes are generated. While the majority of them are bound into excitons, a fraction of the unbound carriers migrate until they are localized by an interface potential fluctuation. In order to find deeper localized states, where the electron (hole) can live a time of the order of ms, this migration must last a sufficiently long time. Such a migration can occur only in high quality QWs that do not have a large density of nonradiative recombination centers. It is noted that in a simple model of interface monolayer islands having comparable dimensions to the exciton area, the exciton localization energy is higher than that of the localized electron (hole), since the effective electron mass is smaller than that of the exciton. Therefore, electron assisted exciton transfer from smaller islands into the larger ones is associated with exciton energy decrease.

The exciton PL lineshape at low temperatures is determined by the competition between the exciton radiative recombination and the exciton spectral diffusion rate. The exciton migration usually occurs by phonon-assisted tunneling between localized states. The interaction between a localized electron and an exciton can result in enhanced probability of tunneling or localization of the exciton in a site with a deeper localization energy. The excess exciton energy is imparted to the localized electron (hole) and it is ejected out of its localizing site. This results in a free electron (hole) that has a higher probability of recombination with other localized holes (electrons). As shown in Fig. 2 curve e, an increase in exciton density leads to a modulation decay time decrease. This can be the result of electron–exciton assisted hopping when the ejected mobile electrons accelerate the recombination with the localized holes.

The temporal behavior of the photomodulated PSB intensity can also be understood in this model. The PSB luminescence is due to the existence of localized electron–hole pairs similar to donor–acceptor pairs in bulk semiconductors[5]. Its PLE spectrum [Fig. 4(a)] is most intense at the energies above the (1e:hh)1S exciton band and below the (e1-hh1) gap; this suggests that the electron and hole in these pairs are localized closely enough to each other so that they are generated by the same photon (geminate pair[7]). This photogeneration process is similar to the resonant excitation of closely spaced donor–acceptor pairs that leads to selective excitation luminescence in semiconductors[7].

As mentioned above the PSB luminescence temporal behavior and its response to electron microwave heating are different of those of the exciton PL. The photoinduced quenching of the PSB luminescence is due to a decreased lifetime of the electron (hole) in closely localized pairs. This occurs in two processes: a fast one that is due to recombination of the electron (hole) of the pair with free holes (electrons) produced by the additional above bandgap photoexcitation. The other is a slow quenching channel of the PSB luminescence that is due to very long lived localized electrons (holes).

Microwave heated electrons dissociate these pairs and the entire PSB luminescence intensity decreases, unlike the PL spectral redistribution that is observed in the exciton PL band. We note that the radiative lifetime of a closely localized electron–hole pair observed by the PSB luminescence decay[5] is much shorter than that measured in this work by photomodulation that is due to more distant localized electrons and holes.

Figure 1(a) shows that the total PL intensity decreases under the additional photoexcitation. In our high quality MQWs the exciton redistribution towards the strongly localized states under additional photoexcitation leads to a decreased total PL intensity since the stronger localized excitons have a longer radiative lifetime[8] and the PL quantum yield decreases. In contrast, the microwave radiation modulation results in a total PL intensity increase [Fig. 1(b)]. The interaction of hot electrons with localized excitons activates them into weaker localized states. Therefore, their radiative lifetime decreases and the quantum yield increases.

Figure 1(b) shows that while the PL spectra are modified under microwave irradiation the Stokes shift between the PL and PLE spectra virtually does not change. Hence, the hot electron–exciton interaction does not result in a free exciton luminescence. Rather, it leads to exciton redistribution between localized states so that their spectral diffusion into deeply localized states is slowed down. However, the hot electrons can activate hopping of localized excitons to the deeper, more localized states when photogeneration occurs directly into localized states [dashed curve in Fig. 4(b)].

In conclusion we show that both photo- and microwave radiation modulations affect exciton migration in the MQW's structures. In the former case the basic mechanism is an interaction between localized excitons and localized electrons (or holes) and it results in enhanced exciton transfer into strongly localized states. In the latter case the excitons interact with electrons that are heated by the microwave field and are activated into weakly localized states. We also show that both distantly localized electrons and holes as well as closely localized electron–hole (geminate) pairs are thus observed in QW structures.

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