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Evidence for excitonic decay of excess charge carriers in high quality GaAs quantum wells at room temperature

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A comparative study is presented of the room-temperature decay of laser excited microwave photoconductivity and of cathodoluminescence of high quality GaAs multiple quantum wells grown by molecular beam epitaxy and of high quality GaAs liquid phase epitaxial layers. The results from both experiments are in *quantitative* agreement and prove that carrier recombination in multiple quantum wells occurs via excitonic decay channels at excess carrier densities less than 10^{17} cm^{-3}. In contrast, band-band recombination prevails in three-dimensional material.

A comparative study of microwave conductivity (μPC) transients and cathodoluminescence (CL) transients of GaAs quantum wells (QW's) at *room temperature* is presented. These methods complement each other: free charge carriers give rise to the microwave signal, excitons will be shown to cause the luminescence signal. Our results demonstrate conclusively *that it is the excitonic decay path which dominates the room-temperature recombination of the two-dimensional electron-hole gas* and thus the decay of both signals at carrier concentrations is less than 10^{17} cm^{-3}.

Thus one of the most controversial issues[1–10] of the rapidly developing field of semiconductor microstructures is answered. This answer is of great importance for many quantum well applications.[11–14] At low temperature the recombination of excess charge carriers in bulk semiconductors like GaAs proceeds via impurity related processes.[15] CL transients and time-delayed spectra reveal a stepwise relaxation and recombination. In both *n*-type and *p*-type samples the rapid capture of free holes is followed by a much slower capture of long-lived free electrons.[15] At room temperature, band-to-band recombination is the dominant decay mechanism.

Recent *low-temperature* CL and photoluminescence (PL) experiments on undoped[1–3,16] and doped[1,2] *p*-type QW's have shown that excitonic recombination is the dominant recombination process for well widths $\leqslant 11$ nm. Increasing structurally induced localization leads to an increase of the radiative recombination rate of excitonic processes as compared to extrinsic ones.[1–3,17] Impurity and trap states are bypassed by the excitons. At room temperature band-to-band recombination[7,10] or excitonic recombination[1,2] was inferred to dominate the emission spectrum. Other authors claim that nonradiative recombination due to defects in the barriers[6,9] or through interface states[4,5,8] governs the carrier decay. Our experiments are designed to distinguish between these possibilities.

The samples used in the present study were grown by molecular beam epitaxy (MBE)[18] in a commercial ultrahigh vacuum system equipped with precise control of effusion cell temperatures and beam shutter operations. The QW layers are either high purity undoped *p* type ($p \approx 10^{14}$ cm^{-3}) or uniformly Be doped ($p \approx 10^{16}$ cm^{-3}). Each quantum well system consists of up to 60 GaAs layers, sandwiched between 17.6-nm-thick $Ga_{0.6}Al_{0.4}As$ layers. The width of the GaAs layers, L_z, was varied in these samples between 5.2 and 11.3 nm. The 4-K mobility of modulation-doped GaAs/GaAlAs heterostructures grown under similar conditions as the MQW layers by MBE is close to 10^6 cm^2 V^{-1} s^{-1} without, and up to 1.5×10^6 cm^2 V^{-1} s^{-1} under illumination,[18] indicating excellent interface quality. Much thicker layers grown by liquid phase epitaxy (LPE) are investigated additionally. We will concentrate here on the results obtained from an undoped $L_z = 11.3$ nm MQW and a 40-μm-thick layer grown by liquid phase epitaxy. The latter layer is of high purity with a mobility of 174 000 cm^2 V^{-1} s^{-1} and a carrier concentration of $n = 1.6\times10^{13}$ cm^{-3} at 77 K.

It is suspected that the first interface between GaAs grown on top of GaAlAs is of poor quality showing high interface recombination velocities and low mobility in inverted two-dimensional electron gas structures. In MQW samples like the ones used in our investigation having 60 GaAs layers the first one or two GaAs quantum wells act in the same way as prelayers which were shown to improve the interface quality.[19] Dawson *et al.*[9] reported for MQW samples grown in a home made MBE system equipped with oil diffusion pumps a nonsaturating learning curve of the interface recombination velocities ($S_1 + S_2$) with a lowest value of ($S_1 + S_2$) = 20 cm/s. If we assume for our high quality QW's ($S_1 + S_2$) = 10 cm/s, the interface recombination time $\tau_s = L_z/(S_1 + S_2) = 113$ ns for a $L_z = 11.3$ nm sample. This time is much larger than any of the time constants which are relevant to our study.

The μPC experiments are performed at 30 GHz using an apparatus described elsewhere.[20] No contacts need to be used. A frequency-doubled pulsed Nd:YAG laser was used as the excitation source. The full width at half-maximum of one pulse is 12 ns. Thus decay times much shorter than 15 ns are not detectable. The deposited energies per pulse were up

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to 50 μJ cm^{-2}. The number of e-h pairs generated per unit volume can be calculated to be $\delta n, \delta p \geqslant 1.3\times10^{18}$ cm^{-3} (illuminated surface: 0.1 cm^2, penetration depth $\leqslant 10^{-4}$ cm). Allowing for an initial rapid decay by one order of magnitude which escapes observation, we still end up at δn, $\delta p \geqslant 1.3\times10^{17}$ cm^3. This number is three to four orders of magnitude larger than the room-temperature carrier concentration of our samples at thermal equilibrium: we are in the high excitation regime.

Figure 1(a) shows the transients of the photoconductivity of the LPE layer for two different excitation intensities on a semilogarithmic scale. The decay is *nonexponential*. This result is to be expected because electron-hole recombination is known to dominate.[21] Figure 1(b) shows the photoconductivity transients of the MQW sample for three different energy densities of the exciting laser pulse equivalent to a calculated initial e-h pair density of 1.3×10^{17}, 1.3×10^{16}, and 1.3×10^{15} cm^{-3}, respectively. The decay is unambiguously exponential and the deconvoluted decay time $\tau \approx 25$ ns is independent of excitation intensity! This key result can only be understood if a monomolecular decay process is dominating. We note that an increase of the excitation density by a factor of 10 leads to an increase of the μPC signal by a factor that is less than 10. The increase of the signal becomes successively smaller with increasing excitation. CL transients and spectra presented now [see Fig. 2(b)] show that this sublinear increase in μPC signal can be explained by the onset of additional excitonic decay channels which lead to shorter effective lifetimes.

A newly developed CL technique[15] is employed here. All transients are taken from a quasi-equilibrium excited state which is populated by a 1-μs-long electron pulse having rise and decay times less than 200 ps. Time delayed spectra show that it takes only a few nanoseconds after the onset of excitation to reach this quasi-equilibrium state. Figure 2(a) shows three room-temperature CL transients of the MQW sample for three different excitation intensities. For the highest excitation intensity of 1.8 kW/cm^2 we estimate a carrier density $\delta n, \delta p > 5\times10^{16}$ cm^{-3}, which is again much larger than the thermal equilibrium density. A detailed mathematical analysis reveals that the decay is *multiexponential* for all three excitation intensities. The analysis also shows that three decay times contribute, and that these are the same for all transients, independently of the initial density. Only the relative weight of each decay time varies with excitation: processes with shorter decay time are increasingly favored at higher excitation. The full lines represent excellent fits to the data, the parameters used for the fits are shown in Fig. 2(a). Remarkably enough, just one decay time, $\tau_1 = 15$ ns, governs the decay after approximately 20

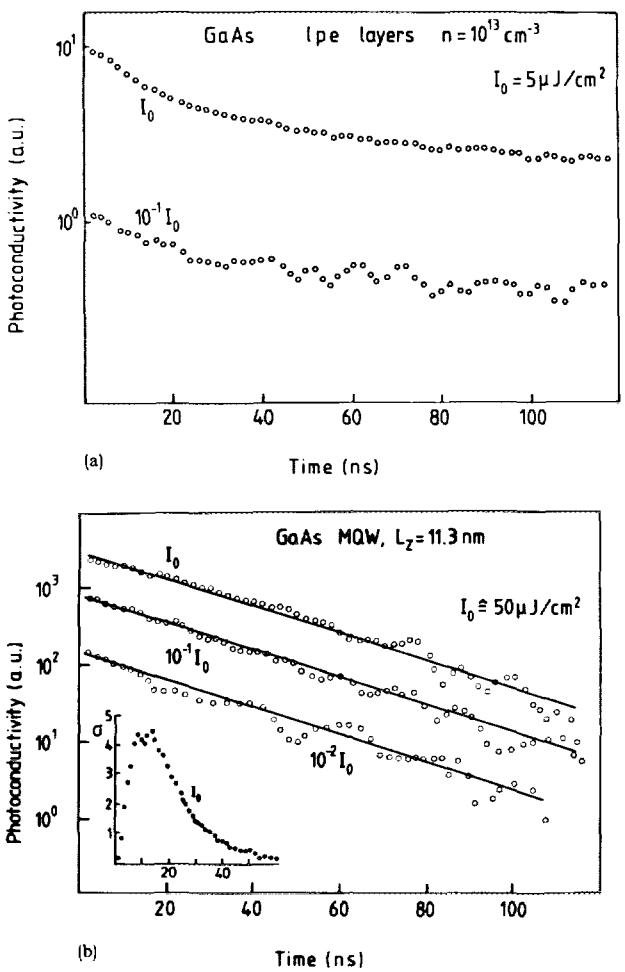

FIG. 1. (a) Photoconductivity transients of a 40-μm-thick high-purity, $n \approx 10^{13}$ cm^{-3}, LPE GaAs layer after excitation with a frequency-doubled YAG laser for two different excitation intensities. (b) Photoconductivity transients of a GaAs MQW sample with $L_z = 11.3$ nm after excitation with a frequency-doubled YAG laser for three different excitation intensities.

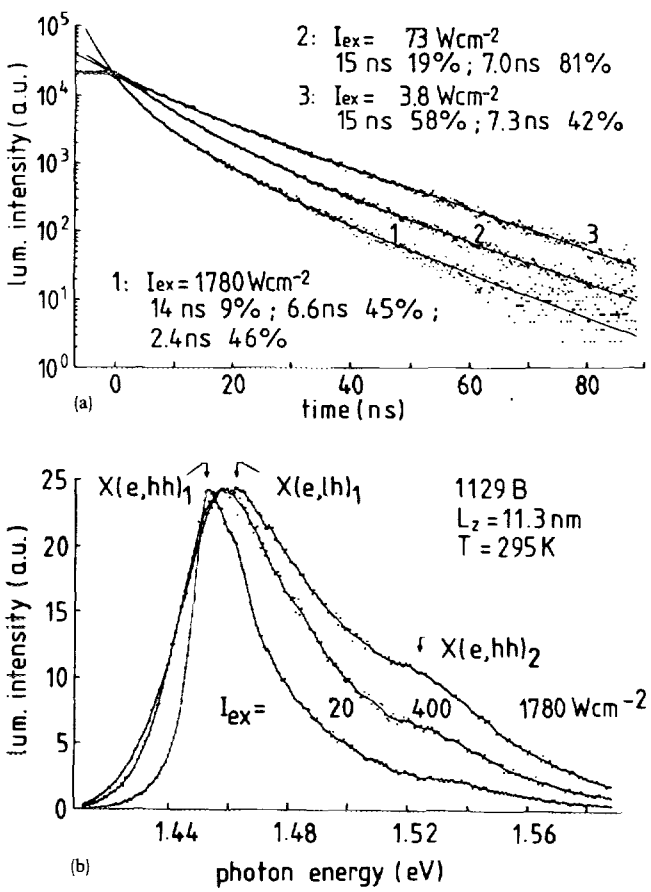

FIG. 2. Cathodoluminescence decay of $L_z = 11.3$ nm GaAs MQW for (a) three different excitations and CL spectra for (b) the same excitation intensities. The full lines present fits to the data points.

ns and all three decay curves are parallel to each other after that time resembling very much the data of Fig. 1(b). This decay time is in good agreement with the decay time of 20 ns derived from μPC, taking into account the slight increase of τ_1 due to the inherent time jitter of the latter experiment.

We will now show that the CL and μPC results taken together conclusively prove the dominance of excitonic recombination over band-to-band recombination in the range of carrier densities discussed here. Let us assume that the thermal equilibrium densities of electrons n_0 and holes p_0 are increased by *e-h* generation process. $\delta n = \delta p$ excess carriers are generated. If excitation is switched off at $t = t_0$ these carriers decay at a rate

$$\delta\dot{n}(t) = -\alpha[(n_0 + p_0)\delta n(t) + \delta n^2(t)], \quad (1)$$

where α is the bimolecular recombination constant.

If $\delta n_0 = \delta n(t_0) \gg (n_0 + p_0)$—this is the high excitation limit—we can neglect the first term in Eq. (1). After integration we get

$$\delta n(t) = \delta n_0/(1 + \delta n_0 \alpha t). \quad (2)$$

The luminescence intensity I_1 is proportional to $\delta\dot{n}(t)$ whereas the μPC is proportional to $\delta n(t)$. The variation of both quantities with time is nonexponential. Precisely such a nonexponential decay is observed for the μPC of the LPE layer [Fig. 1(a)].

If $\delta n_0 \ll (n_0 + p_0)$—this is the extreme low excitation limit— we can neglect the second term in Eq. (1) and get after integration

$$\delta n(t) = \delta n_0 \exp(-t/\tau), \quad (3)$$

with

$$\tau = [\alpha(n_0 + p_0)]^{-1} \approx [\alpha n_0]^{-1}$$

if $n_0 \gg p_0$ (n-type material). An exponential decay with a time-independent decay time results. The decay time depends on the background majority-carrier concentration. Our estimates show that we are *well in the high excitation regime*. Figure 1(a) presents a clear confirmation of that. Thus the observation of an exponential decay of the luminescence and the photoconductivity from the quantum wells with excitation intensity independent decay time(s) cannot be explained by band-to-band recombination but only by an excitonic (monomolecular) process which obeys a law similar to Eq. (3). The dominating recombination mechanism of the free carriers is apparently rapid exciton formation. Therefore, the excitonic lifetime also governs the photoconductivity decay.

Figure 2(b) visualizes why we observe a *multiexponential decay* of the cathodoluminescence at high excitation. Steady state CL spectra are displayed for the same excitation intensities as in Fig. 2(a). Higher lying excitonic levels are increasingly populated with increasing intensity. At the same time some broadening particularly at the high-energy side occurs. This pretty unstructured broadening is attributed to the onset of band-band recombination. A Mott transition might explain this change. This transition might be assisted by scattering from higher exciton levels [$X(e,lh)_1$, $X(e,hh)_2$] to the continuum of the $X(e,hh)_1$ state.

We would like to conclude by emphasizing that we have presented for the first time data on the transient decay of laser excited microwave conductivity of high quality MQW's, monitoring the decay of the *excess free carrier* population in the range 10^{14}–10^{17} cm^{-3}. These data show unambiguously that still at room temperature the free carriers decay via an excitonic channel. Free electrons and holes form excitons on a subnanosecond time scale and recombine subsequently. The results of microwave experiments agree qualitatively and quantitatively with results from time-resolved cathodoluminescence experiments which monitor directly the excitonic decay in great detail. Room-temperature excitonic recombination is unique to two-dimensional GaAs systems as proved by a comparative study of high quality LPE GaAs layers.

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