Photoluminescence dynamics in GaAs along an optically induced Mott transition*

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We present a detailed experimental study of the effects of the optically induced transition from the excitonic, insulating regime, to the plasma, metallic regime, on the spectra and on the photoluminescence dynamics of GaAs. The transition is rather abrupt and presents a Mott-like behavior. The critical temperature, of 49 K, corresponds to the exciton binding energy. Through the study of the characteristics of the photoluminescence dynamics, the critical density for the transition has been obtained with unprecedented resolution. © *2007 American Institute of Physics*.
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I. INTRODUCTION

In a direct-gap, intrinsic semiconductor, like GaAs, the photoluminescence (PL) characteristics can be divided into two well known regimes: (i) the excitonic, insulating regime, at low temperatures (T) and carrier densities (n); and (ii) the plasma, metallic regime, at high T and/or n. Time-resolved optical spectroscopies have provided invaluable information on the interplay between the coupled free-carrier and excitons in photoexcited semiconductors. Most of the studies deal either with the regime (i) or (ii). However, the PL dynamics in the intermediate range, where a Mott transition[1] between the excitonic regime and the conducting electron-hole plasma phase should take place, has not been investigated in detail until recently in bulk III–V semiconductors.[2] Driven by T or decreasing interparticle distance, excitonic states unbound and a plasma of electron-hole pairs dominates the spectra. This transition has been also studied in quantum wells (QWs) by means of time-resolved broadband terahertz spectroscopy[3,4] and PL.[5] In the electron-hole plasma regime, time-resolved studies have concentrated in the thermalization[6] and cooling mechanisms of the hot photocreated carriers, but little attention has been paid to the processes responsible for the onset of the luminescence, characterized by its rise time, τ_r. Considering now the excitonic regime, it is established that in QWs, after photocreation of electron-hole pairs, the exciton formation,[7] and its relaxation to the bottom of the band result in PL time evolutions with rise times up to several hundreds of picoseconds (ps) long. The excitation-density dependence of τ_r is strongly influenced by the sample characteristics. Thus, the literature provides a wide spectrum of experimental data with rise times increasing[8] or decreasing,[9] when raising the excitation density. On the other hand, in bulk III–V samples, these time-resolved studies are scarce[10] and establish that the free-exciton PL rise time is strongly influenced by trapping in localization centers. Recently the origin of the light emission at the energy of excitonic resonances in semiconductors has been questioned,[11] and the contribution of excitons and free carriers to the PL has been the subject of intense debate.[12,13] The question is about the existence of an uncontroversial excitonic signature and how to distinguish between recombination arising from unbound electrons and holes or bound excitons. The theory shows that a sharp PL line at the exciton energy does not necessarily involve the population of excitons but can be caused by Coulomb correlation contributions to the photon-assisted recombination of free electron and hole pairs.[14] Here we present a study of the exciton/electron-hole plasma PL dynamics in bulk GaAs in a wide range of lattice temperatures and excitation densities after a pulsed nonresonant excitation. We concentrate on the onset of the luminescence and demonstrate that the rise time contains direct information on the existence of free carriers and/or excitons. The excitation-power dependence of τ_r for different lattice temperatures presents a behavior typical of a metal-to-insulator transition.

II. SAMPLES AND EXPERIMENT

We have used undoped GaAs epilayers, grown by molecular beam epitaxy, 2.5 μm thick, sandwiched between two thin AlAs layers. The samples, mounted on a cold finger cryostat, were nonresonantly photoexcited (1.631 eV) with 2 ps long pulses from a $Ti:Al_2O_3$ laser. The PL was energy and time resolved with a synchroscan streak camera coupled to a spectrometer (time/energy resolution better than 15 ps/0.3 meV). The estimated density of the photogenerated carriers is correct within a factor of 2 (the relative uncertainty in the density when comparing two excitation densities is below

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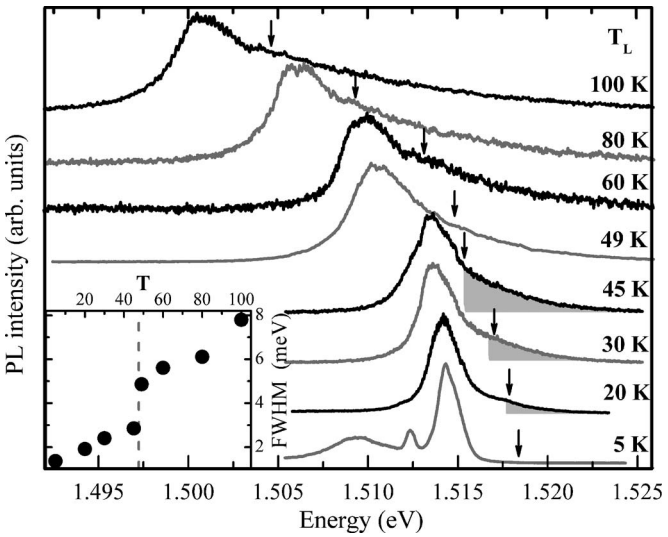

FIG. 1. PL spectra recorded 1.8 ns after an initial pulsed excitation density of 0.75×10^{15} cm^{-3} for different lattice temperatures. Arrows indicate the energy position of the band gap. The shadowed regions show the electron-hole pair luminescence. Inset: FWHM of the whole luminescence band.

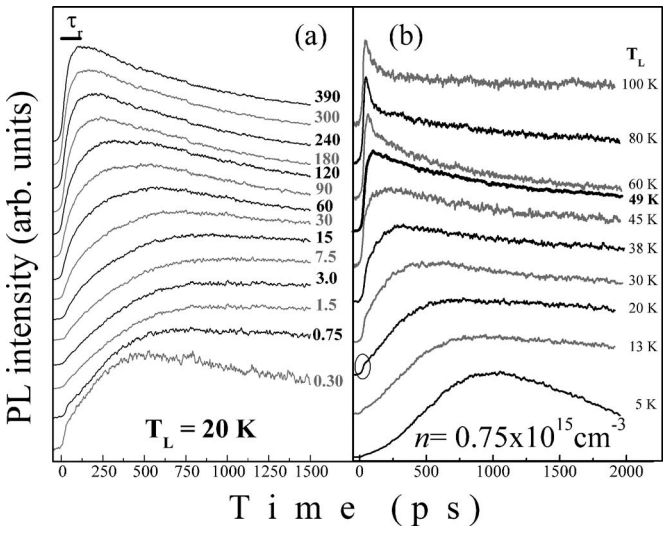

FIG. 2. (a) Time evolution traces at the spectral maximum for T_L=20 K. The numbers on the right show the excitation density for each trace in units of 10^{15} cm^{-3}. (b) Time traces for different lattice temperatures (T_L) at an excitation density of 0.75×10^{15} cm^{-3}.

2%). We have selected the central part of the excitation spot to ensure that we are dealing with an homogenous excitation density.

III. RESULTS AND DISCUSSION

The first indication of the different contributions, either from excitons or electron-hole pairs, to the light emission can be obtained from the T dependence of the PL, as demonstrated in Fig. 1, which depicts spectra recorded 1.8 ns after the excitation at different lattice temperatures for a low excitation density of 0.75×10^{15} carriers/cm^3. At 5 K the spectrum displays the characteristic excitonic emission (1.512–1.516 eV range) and electron-acceptor recombination structures at lower energies.[10] For temperatures up to a critical temperature, T_c=49 K, the spectra are dominated by the excitonic emission. In the range 5 K$\leq T<$49 K the emission from electron-hole pairs becomes apparent (shaded regions) at the band gap energy (indicated by arrows), and its relative intensity increases. In this temperature range, the full width at half maximum (FWHM) of the PL band also increases with temperature (a factor of 2.2 from 5 to 45 K), as it is shown in the inset of Fig. 1. For $T\geq$49 K the spectra present a much wider overall linewidth. The spectrum at 49 K undergoes an abrupt shift toward lower energies and it is significantly wider than that at 45 K. The abrupt shift and the broadening at T_c demonstrate that there are two kinds of spectra belonging to two different regimes. There is also a strong reduction of the binding energy with increasing temperature,[2] indicative of the exciton ionization at T_c.

Low temperature, 20 K, PL time-evolution traces at the energy of the spectral maximum are shown in Fig. 2(a) at different excitation densities n. Following the usual practice in the literature, we employ the time for the PL to reach its maximum intensity, defined as τ_r, to analyze the initial emission dynamics. τ_r is shown for the uppermost curve in Fig. 2(a) with a horizontal bar. At high excitation densities ($n>150\times10^{15}$ cm^{-3}) the system behaves like an electron-hole plasma due to the effective carrier screening and the initial carrier temperatures are much larger than the lattice temperature (T_L), therefore these traces are quite similar for all temperatures.[2] On the other hand, at low excitation densities the PL time evolution presents very different features at different T's, as easily seen in Fig. 2(b). For temperatures up to 45 K the PL onset is characterized by two distinct features: (i) a fast initial component (enclosed by a circle in the trace of 20 K) and (ii) a subsequent slower rise. The interplay between the exciton and electron-hole pair emissions is responsible for the shape of the time evolution of the onset of the PL at the free-exciton energy.

At low excitation densities [Fig. 2(b)], the slow component fully dominates the onset of the PL at the lowest T_L (5 K). Under these conditions the PL is mostly arising from excitonic recombination.[12,13] The long τ_r reflects the slow phonon-assisted exciton relaxation from states of large momentum k, where electrons and holes were bound to form excitons, to the radiatively active states at k=0.[15] The fast component has been previously observed in GaAs and tentatively attributed either to the emission of free electron-hole pairs[16] or to a rapid exciton formation mediated by longitudinal optical (LO)-phonon interactions.[17,18] Although the fast component could be attributed to an enhanced exciton formation due to high carrier temperatures,[18] our results do not favor this mechanism since on one hand the fast component is absent at the lowest temperature and LO-phonon emission is independent of lattice temperature, and on the other hand, at low excitation powers the carrier temperature is low. Therefore, we attribute this fast component to the recombination of unbound electron-hole pairs. With increasing T_L the fast component becomes more important: the fraction of excitons present in the system at short times is reduced, in agreement with computational results.[19] For $T_L>T_c$=49 K, the fast initial component fully dominates the rise time. For those values of T_L excitons are ionized (as $k_BT_L>$4.2 meV,

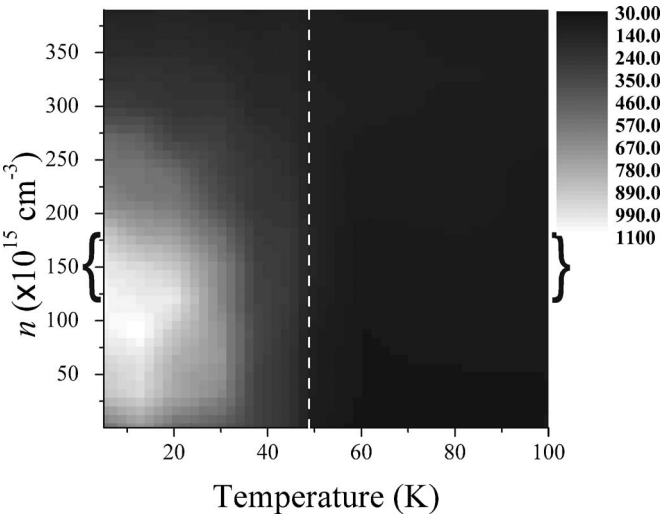

FIG. 3. Gray-scale map of the rise PL time (τ_r) as a function of temperature and carrier density (in units of 10^{15} cm^{-3}). The scale in picoseconds is indicated at the right-hand side bar. The white line marks T_c and the brackets encompass the critical density, n_c, region.

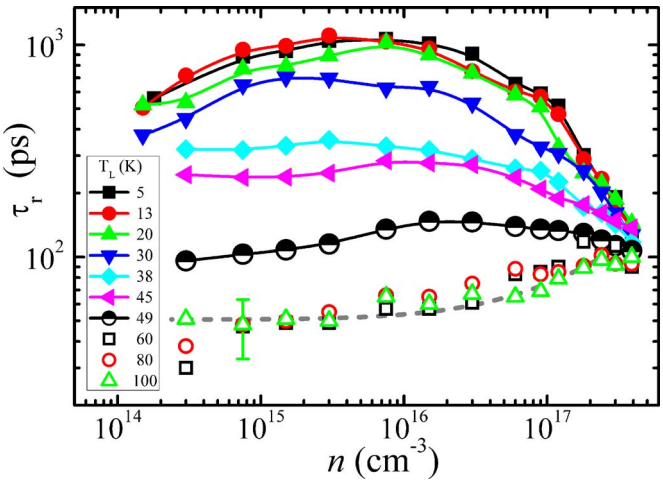

FIG. 4. (Color online) Rise time, τ_r, as a function of excitation density for different lattice temperatures. Solid symbols correspond to the excitonic/insulating phase; open symbols correspond to the electron-hole/metallic phase. The dashed line is a guide to the eye.

the exciton binding energy) and all the luminescence arises from the recombination of Coulomb-correlated electron-hole pairs (see Fig. 1).

Let us now discuss the behavior of the fast rising component of the PL at a given T_L for different excitation powers. For low T_L, Fig. 2(a), where exciton formation is not inhibited by thermal ionization, the fraction of electron-hole pairs that bind to form excitons increases with increasing excitation density.[13,17] This is also borne out by our experiments: the electron-hole recombination (fast component) is overcome by the slow excitonic component when the excitation density is increased ($0.3\times10^{15}<n<3.0\times10^{15}$ cm^{-3}). Screening between carriers becomes significant at higher densities and inhibits the binding of electron-hole pairs into excitons[19]; electron-hole pair recombination is again important and the dynamics accelerate. At the highest densities ($n>150\times10^{15}$ cm^{-3}) the emission occurs mainly from electron-hole pair recombination. These effects are clearly seen in Fig. 3 which presents a two-dimensional gray-scale map of τ_r as a function of temperature and carrier density: the lower left corner illustrates the growing importance of excitonic recombination with increasing density at low temperatures until electron-hole pair emission dominates (upper left corner). This figure also visibly reveals that at $T_L>T_c=49$ K electron-hole recombination is the origin of the light emission independently of carrier density.

There is another feature seen in Fig. 2(b) that is worthwhile discussing. For $T_L>49$ K the fast component of the onset of the PL is followed by an initial fast decay, which becomes more evident with increasing T_L. A detailed analysis at $T_L=80$ K obtains that this fast initial drop is more important at low excitation densities,[2] being completely absent for $n>50\times10^{15}$ cm^{-3}. We interpret this fast decay as a consequence of a warming of the electron-hole plasma. At the lowest excitation densities, the fast subpicosecond thermalization in conjunction with efficient LO-phonon assisted relaxation, results in thermalized carrier populations with initial temperatures close but slightly below T_L. The warming of the carriers to T_L changes the carrier distributions, in particular resulting in a depletion of the states at the energy of the maximum of the PL band. This effect has been observed for excitons in GaAs QWs.[20] For $n>50\times10^{15}$ cm^{-3}, an analysis of the high energy tail of the PL yields carrier temperatures above T_L, therefore the depletion does not occur and the initial drop is absent from the time traces. Similar arguments about the lattice and carrier temperatures explain why the fast drop is lacking at low temperatures and low carrier densities [Fig. 2(b)].

It is clear from the previous discussions that a critical temperature $T_c=49$ K can be identified, which sets a boundary in the spectral (Fig. 1) and dynamic (Fig. 2) behavior of carriers in the system. The interplay between the excitonic (slow component) and electron-hole pair (fast component) recombination is summarized in Fig. 4 for different T_L and excitation densities. The curves can be classified in two groups, corresponding to $T_L<T_c$ (solid symbols) and $T_L>T_c$ (open symbols), around the curve at 49 K. The τ_r dependence on n of these groups shows certain symmetry with respect to the $T_L=T_c$ curve, with negative (positive) curvature for $T_L<T_c$ ($T_L>T_c$). This symmetric behavior is characteristic of a metal-to-insulator transition,[21] being in our case T_L the order parameter having a critical value of 49 K (in transport experiments the order parameter is the resistivity[21]). The first group of curves, $T_L<49$ K, corresponds to the excitonic insulating phase in which the onset of the PL is dominated by the slow component and τ_r is governed by the exciton relaxation. For $n<3\times10^{15}$ cm^{-3} and T_L up to 30 K, the monotonic increase of τ_r with density is related to the trapping of free excitons in bound states.[10] For higher excitation densities, the steady decrease of τ_r with increasing density arises from the fast relaxation of excitons induced by exciton-exciton scattering. The second group, $T_L>49$ K, corresponds to the metallic phase, with the onset of the PL dominated by the fast component (electron-hole pair recombination). τ_r, which increases monotonically with increasing n, is determined by the thermalization and cooling of carriers.[2]

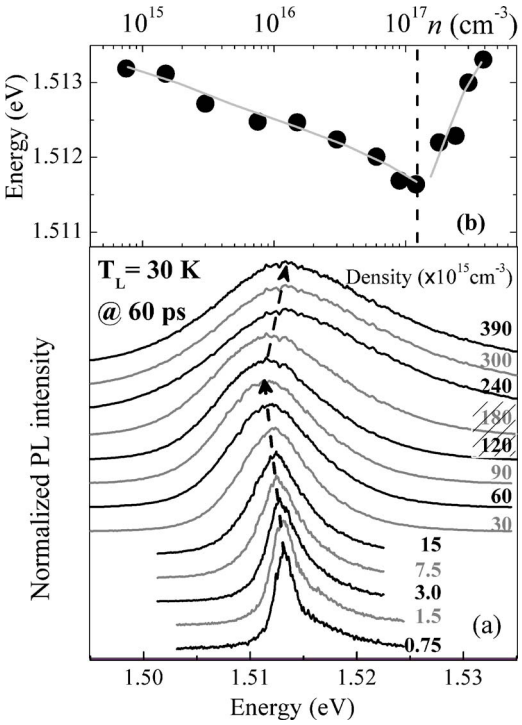

FIG. 5. (a) PL spectra at $T_L=30$ K, 60 ps after the pulse arrival, for different carrier densities (shown on the right in units of 10^{15} cm^{-3}). The arrows are a guide to the eye. (b) Energies of the PL peak as a function of carrier density. The vertical dashed line marks the boundary between the insulating and metallic phase.

Only at the highest excitation densities, in the region where all the curves tend to approach a common value of $\tau_r \approx 100$ ps, the rise time is essentially characterized by electron-hole recombination for any lattice temperature, due to the effective carrier screening, as discussed earlier. Plotting the rise-time dependence on T_L for several excitation densities (not shown, see Ref. 2 for more details), there is a temperature (49 K, the critical temperature) for which τ_r is nearly independent of the excitation density. Additionally, the curves reverse their order when crossing this temperature. These two facts are similar to what is found in resistivity studies around the metal-to-insulator transition in two dimensional high mobility semiconductors.[21]

A close inspection of Fig. 3 reveals that for $n \geq 180 \times 10^{15}$ cm^{-3} the rise-time dependence on T_L has a behavior much less dependent on excitation density than that obtained at lower densities. We attribute this fact to the dominance of the electron-hole plasma (metallic state) contribution to the light emission at high n and short times. Therefore, we are able to ascertain that the excitation density range $120-180 \times 10^{15}$ cm^{-3} establishes a phase boundary in the characteristics of the onset of the PL, similar to the $T_L=49$ K boundary discussed earlier.

This boundary is not only seen in the PL dynamics but also in the spectral characteristics of the emission. Figure 5(a) shows PL spectra at a lattice temperature of 30 K, obtained 60 ps after the arrival of the excitation pulse, for different carrier densities ranging from 0.75×10^{15} cm^{-3} up to 390×10^{15} cm^{-3}. Apart from the broadening of the lines with increasing carrier density, the energy position of the peak of the emission shows a nonmonotonous dependence on n: it redshifts up to a density of $n_c=120-180 \times 10^{15}$ cm^{-3} and blueshifts for larger densities, as compiled in Fig. 5(b). These shifts are due to manybody effects, such as band gap renormalization that redshifts the emission and to the Moss–Burstein effect that produces a blueshift, which compete with each other, but the latter distinctly dominates above n_c with the carriers filling the bands and displacing the center of gravity of the emission toward higher energies. This filling of the bands points to a predominant free carrier contribution to the spectra.[22] Our observations are similar to those reported by Guillet *et al.* in quantum wires,[23] although the redshift is absent in their case probably due to a different weight of the band gap renormalization and Moss–Burstein shift contributions in one dimensional systems as compared with those in bulk materials. Summing up, both, the dynamics and the spectral characteristics change markedly at n_c; the metal-to-insulator transition in the system is set by this density range (densities varying by a factor 1.5), in a much more abrupt fashion than the observed Mott transition in recent experiments in QWs,[4] which takes place over an order of magnitude in excitation densities. The transition densities we find are about five times greater than theoretical calculations[24] that yield a Mott density for GaAs of 28.1×10^{15} cm^{-3} at $T_L=0$.

IV. SUMMARY

We have established that time-resolved photoluminescence, with picosecond excitation pulses, in order to obtain a satisfactory compromise between time and energy resolution, is an invaluable tool to study the role of free carries and excitons in the emission and gives an unambiguous signature of the origin of the PL at the energy of the exciton in bulk semiconductors. We have demonstrated that the rivalry between exciton and electron-hole pair recombination and relaxation determines the spectral and carrier-dynamics properties of bulk GaAs at various lattice temperatures and excitation densities. Both contributions to the PL coexist and cannot be separated at low temperatures (below 49 K) and low excitation densities (below $120-180 \times 10^{15}$ cm^{-3}), as evidenced by the concurrence of a fast and a slower rise time at the onset of the photoluminescence. We have presented evidence of a continuous but rather abrupt metal-to-insulator transition in the rise-time characteristics, at a lattice temperature of 49 K. This temperature is very close to that associated to the exciton binding energy in bulk GaAs (48.7 K, 4.2 meV, respectively). This fact suggests that the transition is ruled by exciton-phonon interaction rather than by exciton-exciton scattering. Similarly, an excitation density of $120-180 \times 10^{15}$ cm^{-3} sets a transition in the rise-time dependence on lattice temperature, which is about five times greater than a theoretical obtained Mott transition density.

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