Effects of microwave electric fields on the luminescence of *n*- and *p*-type GaAs

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(Received 26 November 1996)

The effects of microwave electric fields on the luminescence spectra of GaAs with varying levels of *n*- and *p*-type doping are investigated. The interaction between the microwaves and the luminescence processes can be divided into two different cases. In the first case, the energy that the microwaves impart to the free carriers (both majority and minority) in the material heats the crystal lattice, with the carriers and phonons in thermal equilibrium, and the effect on the luminescence is a manifestation of the temperature dependence of the luminescence. In the second case the microwaves affect the luminescence through an increase of the energy of the free carriers participating in the luminescence processes independent of the temperature of the crystal lattice. The two cases are experimentally distinguished for our experimental conditions using the fact that the lattice-heating effects have long transient times compared to those changes in the luminescence not due to lattice heating. The magnitude of the lattice heating is found to increase with the total number of free carriers present in the system since the rate of lattice heating per free carrier is more or less constant. The magnitude of the lattice heating is in good quantitative agreement with theoretical predictions for the energy relaxation rates of the free carriers and the heat transfer properties of GaAs in an environment of cold helium gas. Models for the effects that are not due to the lattice heating are also in agreement with our experimental results. [S0163-1829(97)06943-9]

INTRODUCTION

A number of modulation spectroscopies have been developed that rely upon the effect of an external perturbation on the luminescence spectrum of a sample. These are used to gain information about the properties of the sample beyond that which is contained in the original luminescence spectrum (e.g., optically detected magnetic resonance,[1] and thermally modulated luminescence[2]). Applying microwave electric fields as the perturbation has the potential advantage of producing a large effect on the luminescence (as large as a 70% change in luminescent intensity in this study) with minimal sample preparation. In addition this change in the luminescence can have a spectral dependence which can give insight into the nature of the luminescence.[3] In order to understand microwave modulated photoluminescence (MMPL), a modulation spectroscopy based on the effect of microwave electric fields on the luminescence, the effects of the microwave electric fields must be understood in some detail. In this study we use the model system of doped, molecular-beam-epitaxy (MBE) grown GaAs, where the properties of the low-temperature luminescence have been extensively studied.[4]

The microwave electric fields accelerate the free carriers present in the material as the luminescence is taking place. This acceleration of the free carriers can alter the luminescence in two basic ways: through heating of the lattice or through some process that is essentially independent of the lattice temperature. There are at least two possible sources for the latter type of effect.[5] First, if the carriers being accelerated are involved in the luminescent processes, they can change the relative intensities of these processes through the different dependences of the capture cross sections on the carrier energy.[6] Since in any real material nonradiative recombination is also a competing process, this effect can also change dramatically the integrated intensity of the luminescence, or the intensity of any single process. A second possible explanation for the changes in the luminescence due to the presence of the microwave electric fields is impact ionization by the accelerated free carriers of shallow donors or of excitons localized at impurities or interfacial steps.[6,7] Impact ionization might manifest itself spectroscopically as the enhancement of band-acceptor recombination at the expense of donor-acceptor-pair recombination or as the enhancement of the free excitonic emission at the expense of that of excitons localized at impurities.[6,7]

Effects on the luminescence due to heating of the lattice can be experimentally distinguished by the inefficient cooling of samples in an environment of helium gas in the proximity of liquid helium. Inefficient sample cooling leads to measurable transients after the microwaves have been turned off for any change in the luminescence caused by microwave heating of the lattice. We have previously established the identification of slow processes in our system as lattice-heating processes with experiments done in different thermal environments; the magnitudes of the slow processes decrease significantly when the sample is immersed in superfluid helium while the fast processes are not strongly affected.[8] Effects of lattice heating by the microwaves have been observed in a number of samples where the concentration of

free carriers is expected to be relatively high either due to doping, as in this study, or due to long carrier lifetimes which lead to high concentrations of photoinjected carriers.[8,9]

EXPERIMENT

The GaAs samples were grown by MBE on semi-insulating GaAs substrates. As will be discussed later, the use of substrates with low free carrier concentrations is required to ensure all measured effects are due to properties of the epilayers. The n- and p-type doping levels ranged from approximately 10^{15}–10^{20} cm^{-3}. Silicon and beryllium were used as the n- and p-type dopants, respectively. Epilayer thicknesses ranged from 1 to 8 μm. Typical sample dimensions used were $\sim$3 mm$^2\times$0.5 mm thick (including the substrate).

The samples were mounted with rubber cement on a Teflon holder and placed in the electric field maximum of a 16-GHz TE_{001} microwave cavity. Typical microwave powers used in this study were $\sim$4 mW which, in the cavity used, generate electric fields $\sim$10 V/cm in a direction parallel to the surface of the sample.[10] A cavity Q of approximately 1000 has been measured for this cavity with a GaAs sample inside. Samples of similar size and similar (semi-insulating) substrates have been used to minimize the change in cavity Q from sample to sample. Since the maximum value of the microwave electric field applied to the sample is proportional to the square root of the cavity Q, variations in cavity Q are not expected to be a major source of error in the analysis of the experiment.[10] The samples were kept in an environment of helium gas at an ambient temperature of approximately 5 K. It is this inefficient method of cooling that allows us to measure transients in effects caused by microwave heating of the sample.[8] Luminescence is excited with a cw argon-ion laser (5145 Å) while the sample is subjected to chopped microwave electric fields. Laser powers of 10 mW, corresponding to intensities of 300 mW/cm^2, were typical. The change in the luminescence due to the presence of the microwave electric fields is measured at the phase and frequency of the chopping of the microwaves using lock-in detection. The luminescence is dispersed with a grating spectrometer and detected with a photomultiplier.

The lattice-heating effects are separated from those changes in the luminescence not due to lattice heating by varying the chopping frequency of the microwaves. As the reciprocal of the microwave chopping frequency becomes short compared to the time scales of the transients of the lattice heating effects, the signal being detected, in this case the change in the luminescence intensity caused by sample heating, decreases in amplitude. Assuming the rise time of the signal is short compared to the characteristic time of the decay, the detected amplitude should be proportional to the inverse of the chopping frequency for values of the chopping frequency much greater than the inverse of the characteristic time of the decay. For the experimental conditions and typical sample dimensions used in this study, this decrease in signal amplitude is observed for frequencies of 10 Hz or greater.

In addition to the aforementioned use of semi-insulating substrates, it is appropriate to discuss some other precautions taken in the design of the apparatus and analysis of the data. The Teflon holder on which the samples were placed slides through two small holes in the microwave cavity such that the position of the sample in the cavity is fixed in one direction. The shape of the holder and specific location of the sample on the holder fix the position of the sample in a second direction as well as remove the rotational degrees of freedom. The position of the sample in the third direction is fixed by the small hole used for optical access to the cavity. The sample carrier concentrations and the epilayer thicknesses were measured by the growers[11] and are used in the analysis of the data.

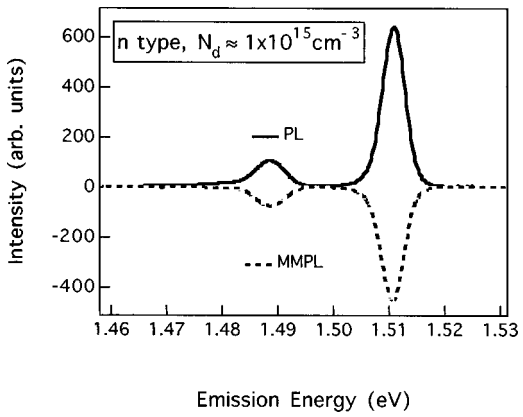

FIG. 1. Typical PL and MMPL spectra. The MMPL spectrum is the difference between the PL with the microwaves on and with the microwaves off. The fact that the MMPL is qualitatively the negative of the PL indicates that the effect of the microwaves is to enhance nonradiative recombination at the expense of all of the near-gap luminescence processes.

RESULTS AND DISCUSSION

A typical PL spectrum as well as an MMPL (difference) spectrum for a relatively lightly doped n-type sample are shown in Fig. 1. The PL shows two features, the higher energy of the two due to excitonic recombination and the lower energy process being associated with recombination through an acceptor level. As is expected,[12] the excitonic emission becomes weaker compared to the emission associated with impurity levels as the impurity concentration increases. For heavily doped material, the total near-gap PL efficiency also decreases drastically as previously reported; this behavior has been associated with self-compensating defects in the material.[13] The MMPL spectrum is qualitatively the negative of the PL spectrum, indicating that the microwaves enhance competing nonradiative recombination, or, in principle, radiative recombination outside the spectral range shown, at the expense of near-gap luminescence. The MMPL signal shown here is relatively fast, indicating that it is not related to a heating of the crystal lattice. Qualitatively, the effects that are related to a heating of the lattice also quench the luminescence and lead to a similar MMPL line shape. Therefore, one cannot necessarily differentiate between these two effects based on MMPL line shape alone.

Another major difference between the lattice-heating and not-lattice-heating effects, in addition to their speed in the experimental design used here, is the magnitude of each ef-

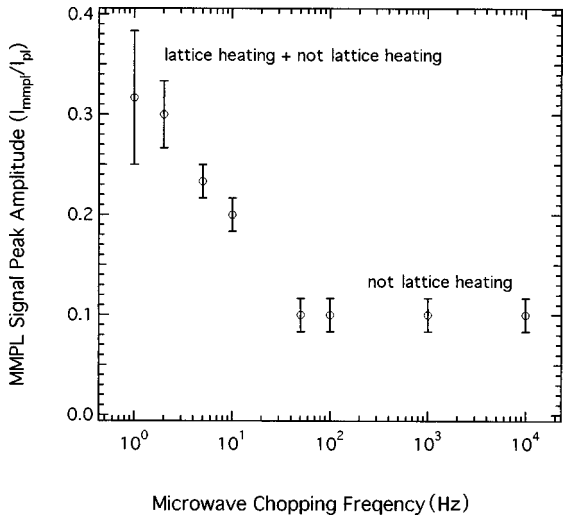

FIG. 2. Chopping frequency dependence of the MMPL signal. The component of the signal due to lattice heating decreases significantly between 1 and 100 Hz, while the component not related to lattice heating is independent of chopping frequency up to at least 10 kHz

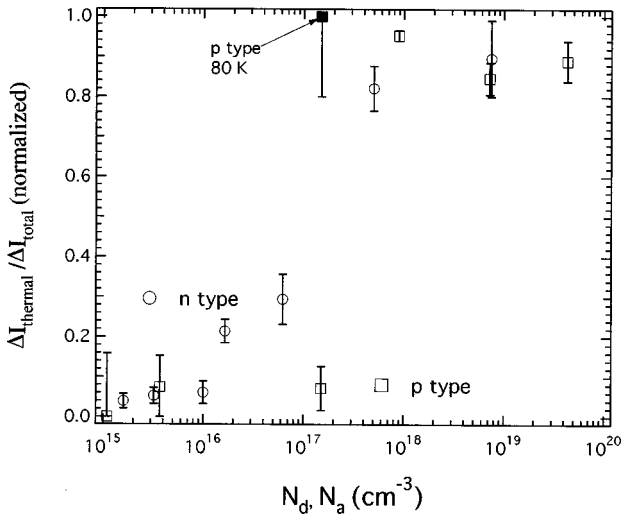

FIG. 3. The magnitude of the change in luminescence due to the lattice heating relative to the total change in luminescence (lattice heating+not lattice heating) caused by the microwaves. The points have been adjusted to take variations in samples thicknesses into account (see text). Normalizing the magnitude of the lattice-heating effects to the magnitude of the total effect of the microwaves reduces the influence of changes in effective power.

fect. For a sample in which the not-lattice-heating effects dominate, these effects are typically a factor of five greater, in terms of the change in the peak intensity of the luminescence, than the lattice-heating MMPL signals observed, for a sample where this effect dominates.

A typical dependence of the MMPL signal (peak amplitude) on the microwave chopping frequency is shown in Fig. 2. The signal contains two components that can be resolved by changing the microwave chopping frequency. One component, which dominates below about 10 Hz and which is due to lattice heating, decreases with increasing chopping frequency. The second component, which is not related to lattice heating, remains independent of the chopping frequency up to 10 kHz.

The lattice-heating component of the MMPL signal, relative to the total MMPL signal, is shown in Fig. 3 as a function of the concentration of impurity atoms present in the samples. The numbers are normalized to account for differences in epilayer thickness. The dependence of lattice heating on epilayer thickness will be discussed in the next section. The general trend is an increase in the lattice-heating component of the signal as the concentration of impurities, and hence of free carriers, is increased. The gross effect demonstrated here is an increase in the total amount of heat transferred to the lattice as the concentration of free carriers increases due to a nearly constant rate of energy transfer to the lattice per free carrier. This is the reason for using semi-insulating substrates; low carrier concentration in the substrates ensures that lattice-heating effects are due to free carriers present in the epilayers.

The magnitude of the microwave lattice heating, and hence the amplitude of the slow component of the MMPL signal, will be related to the total number of free carriers present in the system. At typical sample temperatures during the experiments described here (5–10 K) the concentration of free carriers (both photo- and thermally generated) is not expected to be equal to the concentration of dopants. In fact, at low dopant concentrations, particularly in p-type material, the carriers are expected to be ''frozen out'' at impurities, and free-carrier concentrations are expected to be low. At high dopant concentrations the wave functions of carriers bound to impurities are expected to overlap causing a banding of the impurity levels which results in free-carrier concentrations very close to the dopant concentrations. An estimate for when this Mott transition occurs is when[14]

$$aN^{1/3} > 1, \tag{1}$$

where N is the concentration of the dopant and ''a'' is the Bohr radius for the impurity within the semiconductor. Empirically, this transition is seen at dopant concentrations for which the left hand factor in Eq. (1) is around 0.25.[14,15] For GaAs this corresponds to $N_D \sim 1\times10^{16}$ cm^{-3} for shallow donors and $N_A \sim 1\times10^{18}$ cm^{-3} for shallow acceptors. Hence the range in doping concentrations used here was chosen to include both the ''freeze-out'' and banded regimes, for both n- and p-type samples. The transition between the two regimes is clearly seen in the data for the p-type samples (open squares) shown in Fig. 3. The sample with $N_A = 1.5\times10^{17}$ cm^{-3} has a dramatically smaller lattice-heating component than the sample with $N_A = 1\times10^{18}$ cm^{-3}. In addition, the lattice-heating component of the MMPL signal of the former is significantly lower than that of the two n-type samples, $N_D = 7\times10^{16}$ cm^{-3} and $N_D = 5\times10^{17}$ cm^{-3}. These observations from these four samples indicate that the lattice-heating component of the MMPL interaction is related to the concentration of free carriers and that the carriers are banded for the samples $N_D = 5\times10^{17}$ cm^{-3}, $N_D = 7\times10^{16}$ cm^{-3}, and $N_A = 1\times10^{18}$ cm^{-3}, and ''frozen out'' for the sample $N_A = 1.5\times10^{17}$ cm^{-3}, in agreement with the estimates made above.

Additionally, the lattice-heating component increased for the sample with $N_A = 1.5\times10^{17}$ cm^{-3} when the experiment was performed in an ambient of cold nitrogen gas (see Fig. 3,

solid square), where the concentration of free carriers is expected to be greater than at 5–10 K. If the acceptors are assumed not to be banded at all, the free-carrier concentration at 5 K is expected to be equal to the concentration of photogenerated carriers (essentially no acceptors are ionized at 5 K), while at 80 K the concentration of free carriers (holes) is calculated[16] to be approximately the same as the concentration of ionized acceptors, 2×10^{16} cm^{-3}. This concentration of holes is expected to produce a large amount of lattice heating (similar to 10^{17}–10^{18} cm^{-3} of electrons). The actual magnitude of the steady-state temperature increase is also larger due to the less efficient heat transfer into the environment of gaseous nitrogen compared with helium.

CALCULATION OF POWER DELIVERED TO THE CRYSTAL LATTICE

Assuming we can define a carrier temperature, T_c, to describe the kinetic energy of the carriers as accelerated in the microwave electric field, the rate at which heat is delivered to the lattice by the free carriers present is given by[17]

$$P_{\rm in}=N_c\left[\frac{k(T_c-T_l)}{\tau}\right], \tag{2}$$

where N_c is the total number of free carriers, T_l is the lattice temperature, and k is the Boltzmann constant. The quantity in the square brackets is the rate of energy loss per free carrier. The dependence of the energy relaxation time, τ, on the carrier temperature, lattice temperature, and number of carriers is suppressed in Eq. (2) for simplicity. Theoretical predictions for relaxation rates, which take these factors into account, will be used for the final numerical analysis.[17] Where Eq. (2) is valid, up to a saturation in the amount of power delivered to the sample by the microwaves, the power absorbed by the sample is proportional to the number of free carriers present.

Assuming Newton's law of cooling,[18] the rate at which heat is dissipated from the sample to the gaseous helium environment is given by

$$P_{\rm out}=\kappa A(T_l-T_{\rm He}), \tag{3}$$

where κ is the heat transfer coefficient between GaAs and helium gas, A is the surface area of the GaAs, and $T_{\rm He}$ is the temperature of the helium gas surrounding the sample.

The lattice temperature increase after the microwaves have been on for a long time is then given by the steady-state condition $P_{\rm in}=P_{\rm out}$,

$$T_l-T_{\rm He}=\frac{N_c k(T_c-T_l)}{\kappa A\tau}. \tag{4}$$

It is clear from Eq. (4) that the steady-state temperature increase is proportional to the ratio of the total number of free carriers present in the system divided by the surface area. Neglecting the exposed surfaces on the sides of the sample compared to the growth surface as well as the number of free carriers present in the (semi-insulating) substrates, this ratio is equal to the carrier concentration in the epilayer,

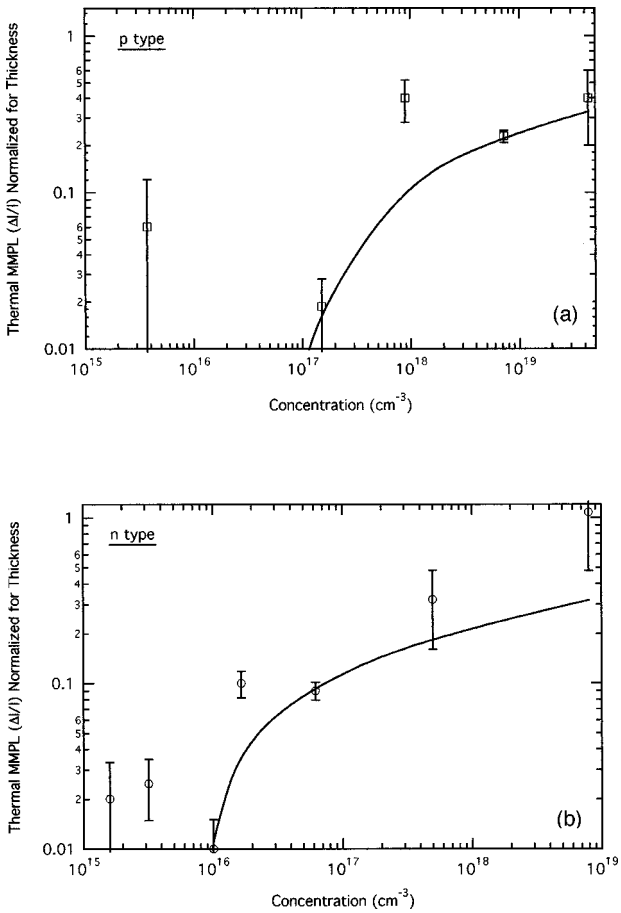

FIG. 4. Fits of the lattice-heating component of the MMPL. The slope of the curves is a measure of the screening of the carrier-phonon scattering (i.e., a reduction in the per carrier energy relaxation rate resulting in sublinear behavior) as the total number of free carriers is increased.

$$n_{e,h}=\frac{N_{e,h}}{At}, \tag{5}$$

times the thickness of the epilayer, t. Hence

$$T_l-T_{\rm He}\cong\frac{n_c t}{\kappa}\left[\frac{k(T_c-T_l)}{\tau}\right]. \tag{6}$$

As previously discussed, the typical microwave electric fields to which the sample is subjected are around 10 V/cm. For a typical scattering length at low temperature (approximately the mean distance between impurities)[19] the free carriers in the material are accelerated to 1 meV by these fields, i.e., T_c exceeds T_l by approximately 10 K. At this energy (1 meV), the rate of energy relaxation [the quantity in square brackets in Eqs. (2) and (6)] is about 1 meV/nsec for holes and is expected to be one to two orders of magnitude less for electrons.[17,20] The coefficient of heat transfer for the samples in helium gas is expected to be 25 W/m^{2}K from both the literature and measurements of the thermal transients in our system.[8] For low electron concentrations (10^{15}–10^{16} cm^{-3}), these values agree well with a steady-state temperature increase of 1–2 K that is estimated from a comparison of the lattice-heating component of the MMPL and the measured temperature dependence of the PL. For typical sample di-

mensions of 1 μm thick by 2 mm^2 this increase corresponds to a value of P_{in} (and P_{out} at steady state) of approximately 0.5 mW.

Note that since the relaxation rates are expected to be much greater for holes than electrons,[17] it is difficult to separate the effects of minority and majority carriers in the more heavily doped n-type samples. However, at low doping levels in the n-type samples any acceptors present are expected to be ''frozen out'' and the ''turn on'' in the lattice-heating component as the donor concentration is increased is due to an increase in the number of free electrons.

Fits of the lattice-heating component of the MMPL to linear functions of the logarithms of the dopant concentrations are shown in Figs. 4(a) and 4(b). The fits are made only on the points where the carriers are expected to be banded (above 10^{16} cm^{-3} for donors and 10^{17} cm^{-3} for acceptors) as discussed in the previous section. Below this level, the magnitude of the lattice-heating component becomes small, in agreement with the number of free carriers becoming small compared to the concentration of impurities (at the onset of carrier freezeout). Because there is no measurable lattice-heating effect in the highest-purity p-type sample, substrate effects are expected to be unimportant for all samples. The fact that in the banded regime of doping concentrations the magnitude of the lattice-heating component is clearly sublinear in the number of carriers, in apparent disagreement with Eq. (6), could result from a number of factors. First, screening of the electron-phonon interaction with increasing free-carrier concentration is expected to reduce the energy relaxation rate by an order of magnitude over the concentration range studied here.[17] Also the saturation of the magnitude of the lattice-heating component is probably due to a fundamental limit on the rate of heating set by the total microwave power. This conclusion is supported by the fact that the saturation is at approximately the same level for n- and p-type samples despite the fact that the energy relaxation rates for electrons and holes are expected to be different, as discussed previously.

CONCLUSIONS

The most significant contribution of this work is the identification of the factors that influence the relative magnitudes of the lattice-heating and not-lattice-heating effects on the PL of GaAs subjected to microwave electric fields. The rate at which heat is delivered to the lattice is proportional to the total number of free carriers present. The actual magnitude of the change in the PL due to lattice heating is determined by this rate, geometrical factors, and the coefficient of heat transfer between the sample and its environment (here helium gas). These factors determine the steady-state temperature increase of the sample required to transfer heat to the environment at the same rate as heat is delivered to the lattice by the relaxation of the free carriers. The measured amplitude of the lattice-heating component of the MMPL signal is in good agreement with the theoretically predicted values for the free-carrier energy relaxation rates in GaAs. Given that the heat transfer coefficients, the free-carrier energy relaxation rates, and the dimensions of the sample are known, the magnitude of the lattice-heating component of an MMPL signal can be used to determine the number of free carriers present during the experiment. Conversely, MMPL could also be used as a tool to measure free-carrier energy relaxation rates.

ACKNOWLEDGMENTS

The authors wish to thank B. R. Bennet, D. S. Katzer, and B. V. Shanabrook at the Naval Research Laboratory for supplying us with well characterized samples. This research was supported by the National Renewable Energy Laboratory under Contract No. XAD3121142 and by the Office of Naval Research under Contract No. N000149410941.

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