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Low-temperature absorption spectrum in GaAs in the presence of optical pumping

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Measurements of the absorption spectra of high purity GaAs in the presence of intense uniform optical pumping are described. With increasing excitation the resonant exciton absorption broadens and disappears, but the enhancement of the above bandgap absorption persists up to the highest intensities used. At these intensities the data can be fit by a calculation which includes band-gap renormalization and carrier heating resulting from intense optical pumping.

I. INTRODUCTION

It is well known that Coulomb interactions strongly influence the low-temperature optical-absorption spectrum in GaAs. These interactions result in a strong, sharp peak due to resonant absorption into the lowest bound state of the exciton and in an enhancement of the near-band-edge absorption coefficient.[1] In this paper we consider how these Coulomb effects are modified as the density of free carriers in the crystal is varied by optical pumping. With increasing excitation intensity the concentration of free excitons increases until exciton-exciton collisions broaden the excitonic resonance line. At sufficient density a Mott transition[2] to an electron-hole plasma occurs. At these densities the presence of free carriers not only leads to screening of excitons, but also leads to band-gap renormalization due to exchange and correlation effects between carriers.[3] In the presence of a relatively dense electron-hole plasma (EHP) the absorption spectrum will be modified to reflect this renormalization and the filling of the near-band-edge states[4] by the photoexcited carriers. The absorption spectrum is further modified as a result of carrier heating[5] that can result from intense optical pumping.

We have investigated these effects experimentally by measuring the transmission spectra of a thin (~0.5 μm) layer of GaAs as function of excitation intensity. The use of a thin layer permits us to resolve the sharp exciton resonance, to measure the transmission above the band gap and to uniformly pump the crystal. We find that the exciton absorption peak broadens and disappears at relatively low pump intensities; but that the excitonic enhancement of the above band-gap absorption coefficient persists to the highest intensity used. We find further that the chemical potential of the photoexcited plasma lies below the exciton energy (i.e., the photon energy at which exciton absorption peak occurs) by a few meV consistent with the requirement for bound electron-hole-liquid (EHL) formation.[6] However, the observation does not unambiguously imply the formation of EHL because of the carrier heating produced by the optical excitation. In Sec. III we will describe these measurements and discuss them in terms of available theories.

II. EXPERIMENTAL TECHNIQUES

The sample, a thin (~0.5 μm) crystal of GaAs sandwiched between two (~2 μm) layers of $Al_{0.12}Ga_{0.88}As$, was grown by molecular beam epitaxy on a GaAs substrate.[7] The substrate was etched away so that only the GaAs layer contributed to absorption of the pump radiation. For this configuration the excitation is nearly uniform in the GaAs crystal and changes in the absorption spectra are readily observed; however, we were unable to measure any net amplification[6] through the thin layer even at the highest excitation level used. A single broadband N_2 laser pumped dye laser was used to excite the crystal and measure the transmission of the sample in the spectral region of interest in the following manner. The bandwidth of the dye laser was approximately 150 Å; with its peak position adjusted to be at ~8050 Å, more than 99% of the incident photons were in the energy range where the crystal absorption was unaffected by intense pumping. The remaining (<1%) photons in the long-wavelength tail of this dye laser served as a probe for the transmission. This technique of pumping close to the band gap was chosen to minimize the carrier heating.[5]

The dye laser output was ~100 W peak with a pulse width ~5 nsec and pulse repetition frequency of ~30 Hz. It was focused to a spot ~100 μm in diameter; the intensity on the sample was controlled by means of calibrated neutral density filters in the laser beam. For all the measurements reported here, the sample was immersed in pumped He

at ~2 °K and the transmitted beam was detected by an intensified Si vidicon after being dispersed by a grating monochromator.

III. EXPERIMENTAL RESULTS AND DISCUSSION

In Fig. 1(a) the solid line shows the low-intensity absorption spectrum of our sample obtained by using a weak tungsten lamp. It is characterized by a sharp peak at the exciton energy (1.515 eV) and a relatively constant absorption above the band gap (1.519 eV).[8] The dashed line shows the theoretical prediction obtained by neglecting the Coulomb interactions.[1] The importance of these interactions are immediately apparent. We also show in Fig. 1(a) the absorption spectra obtained by using the dye laser at two different intensities. We see that the exciton absorption peak becomes broader and then disappears at intensity corresponding to 5×10^2 W/cm^2. A simple heat-capacity calculation indicates that the lattice temperature during the pulse is less than 10 °K. Since the exciton structure persists up to much higher temperatures for low excitation intensities,[1] it is reasonable to conclude that the observed reduction and broadening of the exciton resonance is due to either exciton-exciton interactions or screening of excitons due to free carriers. We estimate the free-carrier density to be in the mid 10^{15} range for this pump intensity.

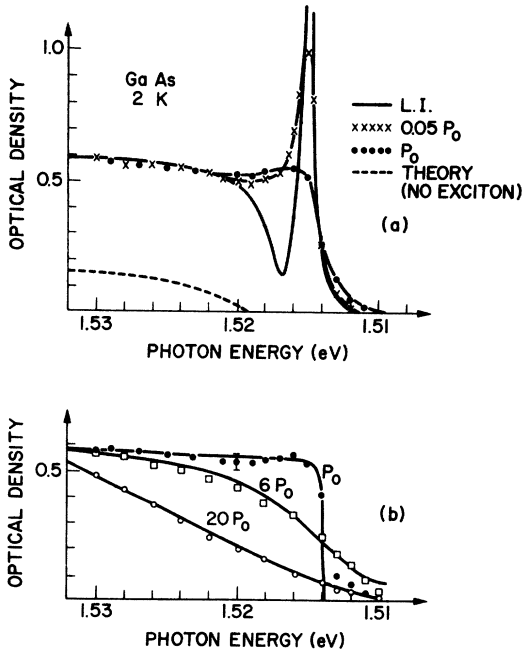

FIG. 1. (a) Optical density of GaAs determined by transmission measurements. The low intensity (L.I.) spectrum was measured with a tungsten lamp, while the remaining spectra were measured with a dye laser as described in the text. The dashed curve illustrates the absorption coefficient due to parabolic bands determined from the L.I. measurement by subtracting the exciton contribution to the total absorption coefficient. (b) Data for high excitation intensity. P_0 corresponds to incident intensity of $\approx5\times10^2$ W/cm^2. The solid lines are calculated as described in the text. Values of carrier density and temperature used in the calculation were, for P_0, $N=7\times10^{15}$ cm^{-3}, $T=20$ °K; for $6P_0$, $N=5\times10^{16}$ cm, $T=40$ °K; for $20P_0$, $N=1.5\times10^{17}$ cm^{-3}, $T=60$ °K.

It is important to point out that while the sharp peak associated with the exciton has been eliminated, the above-band-gap absorption has remained essentially unchanged up to these intensities, indicating that the excitonic enhancement to the absorption coefficient $\alpha(E)$ persists. While free-carrier screening might be expected to influence this enhancement also the following qualitative argument may explain our observation of no screening. Free-carrier screening is a dynamic response of the steady-state population of carriers to the creation of a new charge pair by photon absorption. A time on the order of an inverse plasma frequency ($\sim\omega_p^{-1}$) is required for the steady-state population to respond. However, when a photon of energy $h\nu$ is absorbed, the electrons and holes are created with kinetic energy $h\nu - E_g$, where E_g is the band-gap energy. If this kinetic energy is large the carriers move so rapidly that there is insufficient time to screen their interactions and the enhancement of $\alpha(E)$ due to Coulomb interactions persists. For the range of energy where $h\nu - E_g \simeq \hbar\omega_p$, the excitonic enhancement will be partially screened out. While this description qualitatively explains our observation, a quantitative comparison is not possible at present because no detailed theory describing the absorption line shape in the presence of dynamic screening, is available.

We now consider the absorption spectra obtained at even higher intensities. In Fig. 1(b) we show two such spectra and also reproduce, for reference, the spectrum with 500 W/cm^2 shown in Fig. 1(a). We note that $\alpha(E)$ varies quite gradually with E at these high intensities and that the onset of absorption occurs at energies lower than the exciton energy.

Since the resonant absorption due to excitons is screened the observed absorption spectrum results from the band-to-band transitions in the presence of the photoexcited plasma. The shape and the position of the spectrum are influenced by three factors: (i) dynamic screening processes as discussed above; (ii) band-gap renormalization; and (iii) heating of the carriers.

In the absence of photoexcited plasma and Coulomb enhancement, one expects a simple square-root absorption edge for parabolic bands (Fig. 2, curve 1). In the presence of a photoexcited plasma,

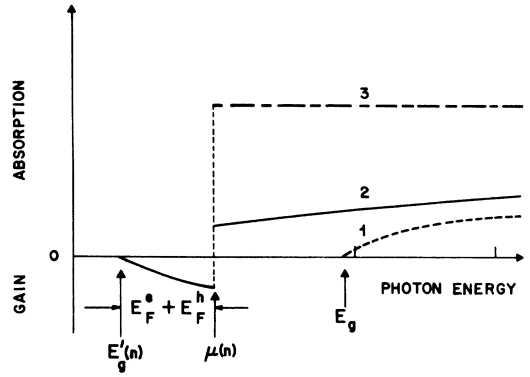

FIG. 2. Illustrative schematic of absorption coefficient at $T = 0\,^\circ\mathrm{K}$ for various conditions as discussed in the text.

the unperturbed band gap E_g is renormalized to E_g'. At $T = 0$, all the states up to the Fermi energies $E_F^e(n)$ and $E_F^h(n)$ are occupied by the photoexcited carriers. The chemical potential is given by $\mu(n) = E_g'(n) + E_F^e(n) + E_F^h(n)$ and determines the energy at which absorption begins. Transitions between occupied states result in negative absorption, or gain, for $E_g' < h\nu < \mu$ and the absorption edge occurs at $h\nu = \mu$.[6] Curve 2, Fig. 2 shows the expected spectrum if Coulomb enhancement of α is neglected. The influence of the carriers on the excitonic enhancement for transitions between occupied states (gain) has been discussed by Brinkman and Lee,[9] however as we point out above, their influence on transition between unoccupied states (absorption) has not been investigated theoretically. Examination of the low-intensity absorption spectrum in Fig. 1(a), suggests that for a pump intensity sufficient to screen the exciton resonance the range of photon energies over which there is significant reduction of the exciton enhancement is small so that it appears reasonable to assume at $T = 0$, $\alpha \simeq \alpha_0$ for $h\nu > \mu$ in the energy range of interest. This is consistent with the dynamic screening arguments presented above. The absorption spectrum for this case is illustrated schematically by curve 3, Fig. 2.

The effect of finite carrier temperature is to broaden the sharp demarcation between occupied and unoccupied states due to thermal excitation out of states near the Fermi energy. The absorption coefficient for $h\nu > \mu$ is simply given by a $(h\nu) = \alpha_0 (1 - f_e - f_h)$, where f_e (f_h) is the occupation probability for electrons (holes) so that $(1 - f_e - f_h)$ describes the band-filling effects at finite temperatures.[4] It can easily be shown that the transition from gain to loss will still occur at $h\nu = \mu$, but the transition will not be abrupt and the reduction of μ for $T > 0\,^\circ\mathrm{K}$ must be properly accounted for.

The chemical potential at $T = 0$ can be calculated directly from the theory of Brinkman and Rice[3] who determine the density dependence of the average energy per pair in the plasma, $\langle E \rangle$. We can write $\mu(n, 0) = \partial N \langle E \rangle / \partial N$, where N is the total number of particles in the system. The reduced band-gap is then given by $E_g'(n, 0) = \mu(n, 0) - E_F^e(n, 0) - E_F^h(n, 0)$. To estimate $\mu(n, T)$ we assume that $E_g'(n, T) = E_g'(n, 0)$ so that $\mu(n, T) = E_g'(n, 0) + E_F^e(n, T) + E_F^h(n, T)$. This approximation should be good for densities near the minimum in $\langle E \rangle$. In Fig. 3, we show $\mu(n, T)$ and $E_g'(n)$ calculated in this manner.

It is now a simple matter to calculate the position and the shape of the absorption spectrum if n and T are known. Unfortunately we do not have an independent measurement of the density n; also it is well known[5] that carrier temperature can be different from the helium-bath temperature under intense optical pumping conditions. We have therefore treated n and T as adjustable parameters in calculating the absorption spectra. The best fit curves, obtained for parameters indicated in the figure caption, are shown in Fig. 2(b) as solid lines. These results were obtained directly from the theory with no displacement of the energy scale. The calculation also predicts a small net gain for photon energies less than μ, but our measurements were not sensitive enough to detect this amplification.

The values of carrier density determined by this fit indicate that the carrier lifetime is short ($\sim 10^{-10}$ sec) consistent with the high threshold for lasers prepared by the molecular beam epitaxy method. Carrier temperatures are difficult to determine

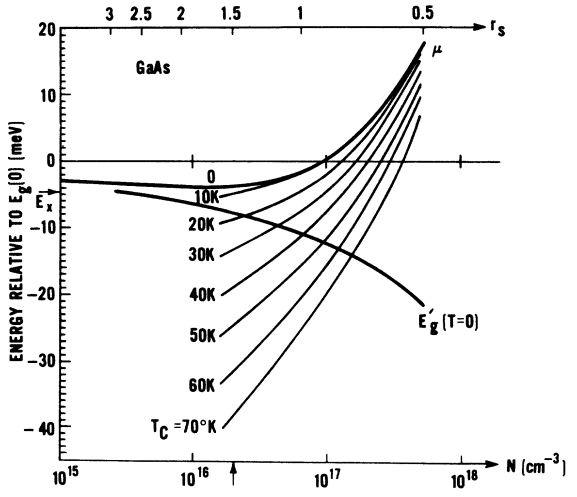

FIG. 3. Reduced band gap E_g' and carrier chemical potential μ for $T = 0\,^\circ\mathrm{K}$ as a function of carrier density. The influence of carrier temperature on chemical potential is also illustrated. The method of calculation is described in detail in the text.

independently, but the values obtained are consistent with what is expected for GaAs even for pumping close to the band edge.[5] With regard to the fitting procedure, we find that with the assumption that the Brinkman-Rice theory applies, a variation of either n or T by more than (10–15)% results in a noticeably poorer fit to the data. In fitting the data we found that carrier temperature principally influences the distribution of carriers at higher energy and therefore the shape of the curve; while the choice of carrier density influences the location of the chemical potential, or photon energy at the onset of absorption. Since in fitting the data no adjustment of the energy scale is made, we take the agreement between the measured onset of absorption and the calculated chemical potential as a measure of the accuracy of the carrier density determination.

Our determination of the carrier chemical potential ~5 meV below the free-exciton energy is consistent with recent reports of the direct measurement of gain spectra[6] and the previous measurements of gain by other methods.[10] In fact all such experimental determinations of the carrier chemical potential, for comparable excitation densities, find it lies below the free exciton in GaAs while the theory of Brinkman and Rice places it at the free-exciton energy for $T = 0\,^{\circ}\mathrm{K}$. The crucial question is does carrier heating account for this difference, or does the theory underestimate the plasma ground state energy sufficiently to allow the possibility of binding of the plasma phase? Since we have made use of the theory to fit our data, and find the data consistent with theory, we would conclude that theory is adequate as it now stands. However the possibility remains that the fit is not unique and an alternative calculation might provide an equivalent fit to the data. Such an interpretation based on gain spectra measurements has been suggested by Hilderbrand *et al.* The result of such a reformulation of the theory could be to interpret a significant fraction of the energy difference between the measured chemical potential and the free-exciton energy as binding energy of an electron-hole liquid. While it is impossible to definitely rule out such a possibility on the basis of our results, it is important to recognize that our measurement of significant reduction of above-band-gap absorption can only be related to carrier heating. And this heating is sufficient to account for the position of the chemical potential relative to the free exciton.

IV. SUMMARY

We have presented data on the absorption spectrum of a thin sample of GaAs in the presence of optical pumping at various intensities. We find that the exciton peak in the absorption spectrum disappears at relatively low intensities, but the enhancement of optical absorption above the band gap caused by Coulomb (excitonic) effect persists to very high intensisties. The spectra in the presence of intense pumping can be fit quantitatively to a model which includes band-gap renormalization according to the theory of Brinkman and Rice and carrier heating caused by optical pumping.

ACKNOWLEDGMENT

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[3]W. F. Brinkman and T. M. Rice, Phys. Rev. 7, B1508 (1973).

[4]The band filling by photoexcited carriers shifts the absorption edge and has been called the dynamic Burstein shift. See, for example, Jagdeep Shah, C. Lin, and R. F. Leheny, Solid State Commun. 18, 1035 (1976).

[5]See, for example, Jagdeep Shah, C. Lin, R. F. Leheny, and A. E. DiGiovanni, Solid State Commun. 18, 487 (1976), and references cited therein.

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