Impact ionization of excitons in GaAs

W. Bludau and E. Wagner
Max-Planck Institut für Festkörperforschung, 7 Stuttgart 80, Büsnauer Strasse 171, F. R. Germany
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We investigated the influence of externally applied electric fields on excitonic photoluminescence in GaAs. Direct experimental proof is obtained for impact ionization of free and bound excitons which was previously assumed from indirect evidence.

I. INTRODUCTION

Neutral shallow donors in semiconductors can suffer impact ionization by charge carriers accelerated in low electric fields.[1-6] Excitons resemble shallow donors in many respects, and thus impact ionization of excitons is also expected to be possible. Asnin *et al.*[7] concluded from the dependence of current-voltage (I-V) characteristics on illumination conditions in Ge that this exciton ionization is responsible for the observed breakdown, i.e., a rapid increase of the current I for applied field strengths exceeding a critical value. Yao *et al.*[8] later confirmed the essential feature of this interpretation.

These electrical measurements,[7,8] however, provide at best an indirect and integral way to investigate exciton impact ionization. In particular, they cannot differentiate between the individual contributions of free and bound excitons. Optical methods, on the other hand, e.g., measurements of luminescence intensities, yield more detailed information, since excitons are observed directly by their radiative decay. Difficult purity control of materials and experimental complications are probably the reasons why only few attempts have been made to perform investigations of this kind. Southgate *et al.*[9,10] mention a suppression of the exciton peak in GaAs at high electric field and at 77 K; a similar observation is presented by Moriya and Kushida.[11]

It is the purpose of this paper to exploit the present advanced state of the art in impurity control and optical detection techniques to elucidate exciton impact ionization. Direct evidence for ionization of free and bound excitons is obtained by observing field-dependent suppression of exciton recombination.

II. EXPERIMENTAL

The GaAs samples were layers grown by liquid-phase epitaxy on semi-insulating substrates. The specimens selected for the measurements were n-type with characters listed in Table I. The typical size of samples was 3×5 mm^2. Ohmic contacts were formed by alloying with Sn, causing the electric field to be parallel to the sample surface. The samples were mounted in a He-immersion cryostat and held at 1.8 K. Luminescence was excited with the 5145-Å line of an argon laser, with typical excitation power densities of 1 W/cm^2 on the illuminated spot. A single-photon counting technique was established with a 0.75-m grating SPEX monochromator and a cooled RCA 31 000 E photomultiplier tube. The wavelength resolution adjusted for spectra recording was better than 0.5 Å.

To check possible current-induced sample heating, we also used pulsed electric fields. By reducing the duty cycle we diminished the input electric power by 4 orders of magnitude compared to that of the dc case. Analysis of the temperature-dependent line shape[12] of the free-electron–acceptor-bound-hole transition, (e,A^0), assured us that no appreciable sample heating occurs even at the highest stationary fields applied. Thus thermal dissociation of excitons[13] can be excluded under the conditions used in our experiments. Determination of the field at the illuminated spot is somewhat ambiguous due to the influence of photocreated carriers on resistivity. We quote here the

TABLE I. Data of GaAs crystals investigated.

Sample	Temperature (K)	Mobility (cm^2/V sec)	Dopant Concentration (cm^{-3})
$E258$	300	7760	5.3×10^{13}
	77	136 000	6.0×10^{13}
$E286$	300	9300	6.0×10^{13}
	77	128 600	7.8×10^{13}
$E299$	300	7900	4.5×10^{13}
	77	127 000	4.5×10^{13}
$E312$	300	8460	1.0×10^{14}
	77	115 600	1.0×10^{15}
$E314$	300	6900	6.3×10^{12}
	77	178 000	1.0×10^{13}
$M67$	300	6160	2.0×10^{14}
	77	99 100	1.8×10^{14}

average field, i.e., the quotient of the applied voltage and the contact spacing.

III. EXPERIMENTAL RESULTS

Figure 1 shows spectra of exciton luminenscence recorded at different electric fields. The emission bands at 8180 and 8182 Å are known[14] to stem from the upper and lower polariton branch of the free exciton, X, respectively. The line at 8186.5 Å is due to the recombination of an exciton bound to a neutral donor (D^0, X).[15] The lines around 8195 Å originate from acceptor-bound excitons (A^0, X).[16] The line at 8190.8 Å—still ambiguous—is dedicated to either an exciton bound to an ionized donor (D^+, X),[13,17] or to the transition of a valence-band hole to a neutral donor (D^0, h).[14,18] We performed an analysis of the field dependence of this emission band and found[19] behavior analogous to the transition of free electrons to acceptor-bound holes.[20] This favors an interpretation that the line originates from the valence-band hole to neutral-donor recombination (D^0, h). We adopt this interpretation here.

The advantage of the experimental technique presented here in comparison with simple I-V measurements lies in the spectral resolution of luminescence suppression under electric fields. As can be seen in Fig. 1, all the emission bands are affected by increasing field strength. In order to obtain the luminescence as functions of field, the integral intensities of the individual lines were recorded. This is shown for one sample in Fig. 2. The results indicate that all lines are affected by the field (of some V/cm) and that the critical field strength at which impact ionization occurs depends on whether the exciton is free, donor or acceptor bound.

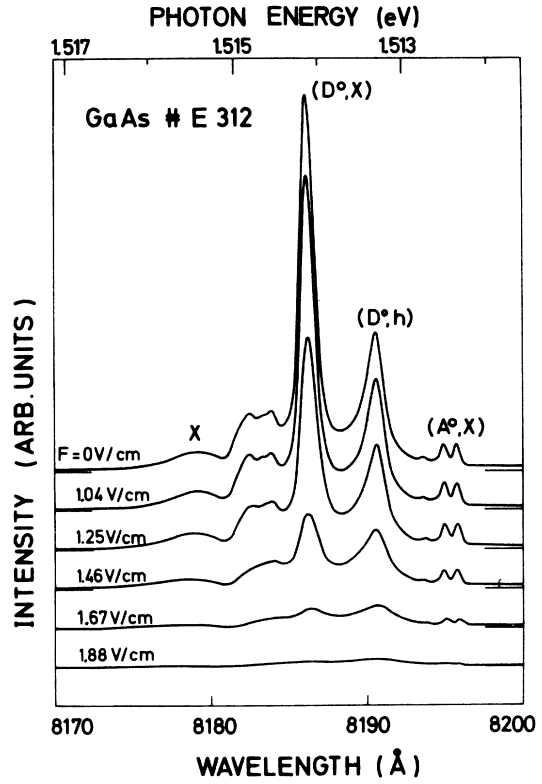

FIG. 1. Exciton luminescence spectra in GaAs. Parameter: applied electric field strength F. See text for nomenclature.

IV. DISCUSSION

A. Quenching mechanisms of exciton luminescence

As the experimental data of Figs. 1 and 2 show, luminescence intensities are quenched in electric fields. Quenching mechanisms which can cause this effect are (a) thermal dissociation, (b) ionization via Stark-effect, (c) field-induced enhancement of the nonradiative portion of exciton recombination, (e.g., Auger decay), (d) change in radiative lifetime, and (e) impact ionization by field-accelerated electrons. In the following we demonstrate that impact ionization must be the substantial mechanism for this quenching.

Thermal dissociation (a) was excluded already

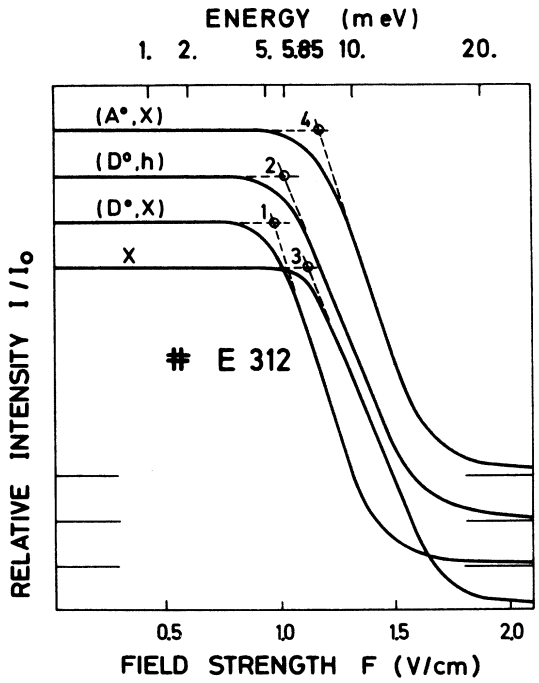

FIG. 2. Field dependence of the individual luminescence lines near the band edge of GaAs. I, I_0: intensity with and without field, respectively.

from experimental results. The influence of electric fields on wave functions and energy levels, (b), becomes important only at fields considerably higher[21] than those in our experiments (which were of the order of 1 to 10 V/cm). Blossey's calculations[21] indicate that spectra are only influenced by fields above 500 V/cm for GaAs. Thus, direct field ionization cannot explain the intensity suppression either. A shift in the ratio of radiative to nonradiative transition rate and changes in radiative exciton lifetime were checked by observing the radiative decay of conduction-band electrons. Figure 3 indicates unambiguously that suppression of exciton luminescence is accompanied by an increase in the (e,A^0) transition rate. This increase signifies an enhanced electron concentration in the conduction band.[22] To separate the contribution of impact ionization of excitons from that of donors, we recorded the I-V characteristics under *resonant* excitation of excitons. Experimental results are shown in Fig. 4. The breakdown without illumination is due to impact ionization of neutral donors.[5,6] Figure 4 reveals that resonant excitation of excitons leads to a current, i.e., an increase in free-electron density, at field strengths lower than those for donor ionization.

The generation of excitons is the only effect of resonant excitation. Accordingly the rise in electron concentration and the corresponding decrease in excitonic luminescence (see Fig. 3) cannot be explained by a change in the radiative-nonradiative transition ratio (c), nor by a diminished lifetime (d).

In conclusion, the impact ionization (e) of excitons by accelerated electrons is the only phenomenon consistent with all experimental results that is left to explain the suppression of exciton luminescence in electric fields.

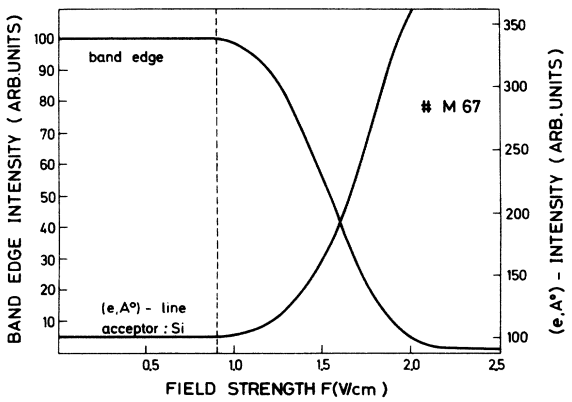

FIG. 3. Comparison of the field dependences of the total near-band-edge emission with the (e,A^0) luminescence.

B. Impact ionization of bound excitons

The exciton recombination intensity as a function of electric field (see Fig. 2) shows a distinct difference among the various types of excitons. The threshold fields are sample dependent, in as much as scattering mechanisms and capture processes influence the charge carrier mobility and thereby the drift energy gained from the field. The sequence in intensity suppression, however, was found to be the same in all the samples examined. Thus we have to conclude that there is a direct dependence between the free-carrier energy and the amount of energy needed for ionization of the free exciton or the exciton-impurity center complex. Impact ionization of (D^0,X) and (A^0,X) will be discussed first. For reference, Table II shows the decay channels to which donor- and acceptor-bound excitons can be dissociated by impact of an electron. The parameters required for these processes are the binding energies and ionization energies, respectively, since k momentum does not have to be conserved for bound excitons. Dissocia-

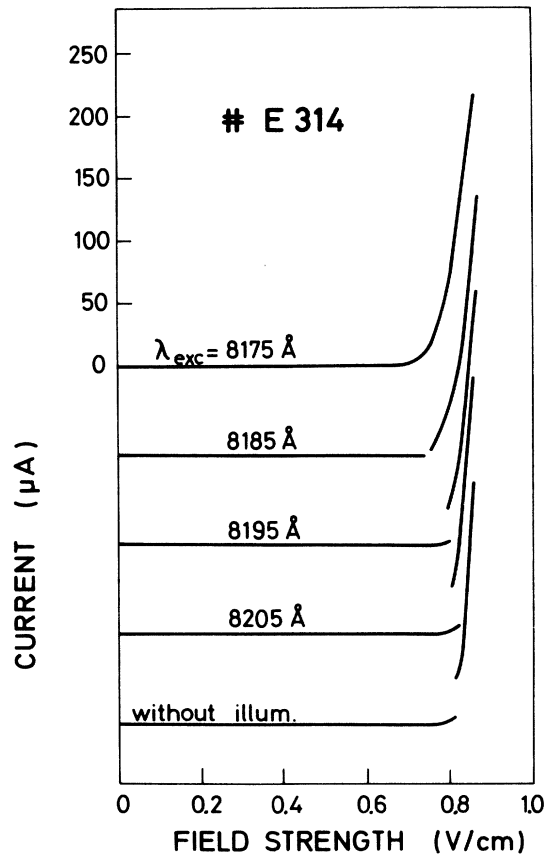

FIG. 4. I-V characteristics with resonant excitation at the cited wavelength λ_{exc}. For clarity, the curves are shifted versus each other.

tion paths labeled by (a) lead to a final state with a neutral impurity center and a free electron-hole pair; (b) labeled processes end in an ionized impurity and a free electron, while for channels designated by (c) the exciton is dissociated as a whole from the neutral impurity center.

Experimental results pertinent to discussion about dissociation mechanisms are

(i) the concentration of conduction-band electrons increases simultaneously with the suppression of excitonic decay (this is obtained from Figs. 3 and 4), indicating an impact process which results in free carriers.

(ii) Impact ionization of neutral donors [c.f. quenching of (D^0, h)] sets in between the threshold fields of ionization of donor- and of acceptor-bound excitons, respectively.

Using the well-known effective-mass donor binding energy of 5.85 meV in GaAs,[23] one is able to define an energy scale in Fig. 2 according to the relation between electron drift energy E_d and electric field F

$$E_d \sim F^2 . \tag{1}$$

For that scale, the donor binding energy is positioned at the field for which onset of donor impact ionization [c.f. (D^0, h) line] is observed.

The extrapolated points at which impact ionization sets in (open circles in Fig. 2) yield the critical drift energies ~5.4 meV and ~8 meV for (D^0, X) and (A^0, X) ionization, respectively. The ionization channel (c) of Table II is thereby excluded. The accuracy of the intensity curves shown in Fig. 2 is insufficient to discriminate between the dissociation processes (a) or (b) of Table II. Process (b), however, seems to be more likely under our conditions, since Coulomb interaction collisions show tendency towards producing charged final products.[24]

C. Impact ionization of free excitons

The only decay channel for free excitons, X, is the total dissociation into free electrons e and free holes h (excited states should not be considered), i.e.,

$$X \rightarrow e + h .$$

The binding energy of free excitons in GaAs is $E_X = 4.1$ meV.[25] In contrast to bound excitons, k momentum has to be conserved, thus giving a minimum collision energy of [Eq. (A3) in Ref. 26]

$$E_{\min} = (1 + 2\alpha/1 + \alpha) E_X \tag{2}$$

with $\alpha = m_e/m_h$, the quotient of effective electron and holes mass. With $m_e = 0.065 m_0$ and $m_h = 0.54 m_0$ (m_0: electron rest mass),[23] one obtains $E_{\min} = 4.6$ meV.

TABLE II. Dissociation channels and energies of bound excitons in GaAs. D^0, D^+: neutral, ionized donor, respectively; A^0, A^+: neutral, ionized acceptor, respectively; X: free exciton; e: conduction-band electron; h: valence-band hole. The various dissociation channels are described in Ref. (13); the data cited are taken from the measurements of Ref. (25).

Bound exciton	Dissociates in	Dissociation energy (meV)	
	$D^0 + e + h$	5.3	(a)
(D^0, X)	$(D^+, X) + e$	5.1	(b)
	$D^0 + X$	1.2	(c)
	$A^0 + e + h$	7.0	(a)
(A^0, X)	$(A^+, X) + e$	$\lesssim 7.0$	(b)
	$A^0 + X$	2.9	(c)

Experimental results in Fig. 2 and data from other samples reveal a discrepancy between the minimum energy of the colliding electron and the energy read from the scale of Fig. 2. We assume that regeneration of free excitons is responsible for the higher field needed to suppress free-exciton luminescence. Charge carriers released by impact ionization rapidly form free excitons, thus partly compensating the loss by ionization. This assumption was confirmed by finding even an increase in free-exciton luminescence in analogous measurements on an InP sample at electric fields sufficiently high for (D^0, X) ionization. We could not find this effect on our GaAs samples, but believe from the slight variation of the ionization onset for free excitons compared with the other bands that the relative rate of free-exciton regeneration is strongly sample-dependent.

V. CONCLUSIONS

We have shown by means of photoluminescence investigations directly that excitons in GaAs suffer dissociation by impact ionization if an electric field as low as several V/cm is applied to the sample. From these results, experimental proof was given for this process suggested earlier from indirect electrical measurements. The analysis of the exciton spectra revealed a distinct behavior of the different types of excitons, free, acceptor and donor bound. Comparison with the fields needed for donor ionization showed that in contrast to thermal dissociation, a bound exciton is not separated as a whole from the impurity center, but instead that charged particles (electrons and holes) are split off by collisions.

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