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High-density excitonic state in two-dimensional multiquantum wells*

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ABSTRACT

A sharp photoluminescence spectral feature has been observed in AlGaAs/GaAs multi-quantum-well structures under high intensity, resonant excitation at the ground state exciton. This feature, which appears below the exciton ground state, emerges from a cold dense system of excitons, but prior to the break-up of excitons into an electron-hole plasma. A collective excitonic state is speculated.

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When excitons in semiconductors are present in sufficiently high density, their ultimate break-up into an electron-hole Fermi system is inevitable. However, the transitory stages between a neutral excitonic gas phase which is essentially single particle state and an electron-hole metallic phase, a many body state, is not well understood, experimentally or theoretically[1,2]. Much less understood is the problem in two dimensions. In this communication, we report the observation of luminescence spectral features from quasi-two-dimensional (Q2D) semiconductor heterostructures under high intensity optical excitation. The observed luminescence spectra bear the characteristics of many body phenomena that appear to be more complex than the two extreme phases mentioned. Using resonant excitation method to generate dense exciton systems, we studied the evolution of luminescence as function of excitation intensity. The emergence of these luminescence lines is described and their nature will be discussed.

The samples are undoped ($N_{A,D} < 10^{15}$ cm^{-3}) $Al_{0.3}Ga_{0.7}As$-GaAs multi-quantum well (MQW) structures grown by molecular-beam epitaxy. Excitation was performed with an optically pumped tunable dye laser of 8 nsec pulse duration. Samples were mounted in a dewar whose temperature was varied from 1.8 to over 100 K, and in a magnetic field up to 10 tesla. The incident light was normal to the sample, and focused to a spot ≈ 100 μm. The luminescence light was detected in two configurations, either normal to or parallel with the MQW plane. In the first configuration, the luminescence is essentially spontaneous, while in the latter some stimulated emission effect may occur due to a longer active length (≈100 μm). Thus, care was taken to ensure proper experimental observation and interpretation of various luminescence spectra. Light emanating from the entire excited spot was collected, and spectra were somewhat inhomogeneous. The excitation intensity I_{ex} was varied with calibrated neutral density filters. The ab-

absolute values of I_{ex} were determined to within a factor of two, but the relative intensities were within 10%. Luminescence was analyzed with a spectrometer equipped with a GaAs cathode photomultiplier.

Luminescence spectra of emission normal to the MQW layer plane are shown in Fig. 1 for sample 1. The sample was nominally a 146 Å MQW structure as depicted in the inset. The low intensity absorption spectrum shows the two lowest exciton states, the heavy hole and the light hole exciton (Fig. 1 (a')). The large linewidths of these two lines are due to inhomogeneous strains in the thin sample for absorption. Luminescence spectra at low excitation intensity (trace (a)) show the heavy hole exciton recombination (labeled X_h) as a narrower high energy peak, and other extrinsic structures at lower energy. These characteristic properties of MQW structures under low intensity excitation have been well established[3]. High intensity studies, from 10^4 to 10^6 W/cm^2 were performed using near or on resonant excitation techniques, i.e., the excitation energy $h\nu_{ex}$ was close to the ground state (heavy hole) exciton energy. Under this condition, excitons are created with very little kinetic energy. When I_{ex} exceeds 10^4 W/cm^2 (Fig. 1, traces (b)), a new feature, labeled X', emerges at about 6 meV below the heavy hole exciton. The X' linewidth is comparable to or narrower than those of the exciton states. As I_{ex} is further increased, the X' intensity grows much faster than that of X_h, and X' energy red-shifts slightly, but not very significantly. At an intensity about 30 times the threshold value at which X' emerges, a second structure appears (Fig. 2). This second structure, which rapidly overwhelms X' at still higher I_{ex}, can be presumed to be due to an electron-hole plasma (EHP), since an EHP is the ultimate limit of a high density electron-hole system. Lineshape analysis of the second structure in terms of the EHP theory yields qualitatively the expected behavior and, thus, renders support to this interpretation. The same process was observed in the luminesin the parallel configuration. The gain factor across the excited spot was measured and found to be small for $I_{ex} \leq 2\times10^5$ w/cm^2. The luminescence in this configuration is then essentially spontaneous. However, at higher I_{ex}, where the EHP emission appears, there is significant difference between the spectra of the two configurations obtained under identical excitation conditions, thus indicating the effect of stimulated emission on the EHP recombination. At issue in this communication is the nature of X'.

It is important to establish that X' is a different entity from the EHP. Besides the evidence from the luminescence spectra, excitation spectroscopy also contributes evidence for this distinction, as well as suggests the close connection between X' and exciton. The luminescence detected at X' displays a strong resonance with a sharp low energy edge as the excitation energy $h\nu_{ex}$ is near the heavy hole exciton (Fig. 3). This sharp edge is coincident with the low intensity CW absorption of exciton (Fig. 1 (a')). Although the bleaching of this exciton resonance eventually occurs at very high I_{ex}, it is clear that at the excitation level where X' begins to form, excitons still exist as a defined energy level of the semiconductor. This aspect is a further distinction between X' and EHP. At the electron-hole density where an EHP is formed, excitonic structure is completely bleached out[1,2]. Absorption at the intensity for EHP creation should not show strong excitonic feature. The dependence of X' on $h\nu_{ex}$ also suggests a connection between X' and exciton. As $h\nu_{ex}$ exceeds the exciton energy, X' appears to shift toward lower energy (Fig. 3, trace (b)). However, although evidences were not as obvious as in luminescence spectra, the apparent redshift of X' was probably not real, but due to the emergence of EHP in lieu of X' for $h\nu_{ex}$ larger than a certain value, at a constant I_{ex}. In summary, resonant excitation was found to be the crucial condition for the observation of a sharp and distinct X', as well as the discernment of a gradual appearance of the

IP recombination close to X'. This indicates that a cold, dense system of excitons is required for the appearance of X'.

For increasing lattice temperature T_L (up to 45 K), the intensity of X' decreases while that of X_h increases or remains relatively unchanged. This relationship between X' and X_h intensities vs. T_L is also dependent on I_{ex}. This may suggest that X' and X_h represent two different phases of a dense excitonic system, in which a temperature shift causes an increase in one population at the expense of the other.

The above experimental results are applicable to a number of samples. Two samples with 145 and 63 Å well width yield the most suggestive data. Table I briefly summarizes the measured characteristics of the experimental results. There are samples which failed to exhibit this kind of below-exciton emission, instead, broad band emission extending toward higher energy, i.e., Burstein shift, was observed. We believe that in these samples, optically generated carriers fail to completely relax in energy within their radiative recombination lifetime to form a many body state.

Phenomenologically, somewhat analogous luminescence effects were observed for highly excited bulk GaAs[4,5]. A structure labeled A or P by various authors appeared to be similar to X', although the latter seems to be more pronounced. The A/P structure has been interpreted [5] as an exciton-exciton (or excitonic polariton) scattering effect. For this case, this model would involve two heavy hole excitons undergoing a collision which leaves a light hole exciton and a photon which becomes X'. However, this model does not appear to be satisfactory to account for X'. A serious objection is based on the Zeeman behavior of various states, shown in Fig. 4. The light hole exciton states split into two resolvable states with opposite polarization. Yet no such corresponding

...ssing of X', as stipulated by the conservation of energy, was observed. ... if X' energy could be arbitrarily correlated within a linewidth to either ...nch of X_g, the deviation (Fig. 4) is too systematic to accept the hypo...sis.

In conclusion, high intensity resonant excitation on MQW structures pro...es a luminescence feature arising from a cold dense exciton gas, prior to the ...mation of degenerate electron-hole metallic phases. There have been theoret...l studies[6] which predict a first order Mott transition for a 2D Coulomb ..., directly from a neutral phase to a plasma phase. The picture from the cur...t experimental work appears to be more complicated than this theoretical ...el. There appears to be an intermediate state between an exciton gas and an .... X' could represent a collective state formed from a system of 2D interact... excitons. A possible boson-boson interaction, i.e., excitons exchanging ...l or virtual acoustic phonons at certain exciton critical density is a hy...hetical consideration. Such systems of interacting excitons appear to form a ...und state prior to the formation of a degenerate electron-hole phase at much ...her densities.

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Table I. Energy (E) and linewidth (Γ) in unit of meV of X', the heavy hole exciton X_h and the light hole exciton X_ℓ for two samples. Measurements are by optical methods.

	X'		X_h		X_ℓ	
h (Å)	E	Γ	E	Γ	E	Γ
5	1527.5±1	2	1533.5±0.5	1.8	1540.0±0.5	2.5
[illegible]	1570±2	5	1582±2	5	1597±8*	10

ad and complex structure was observed

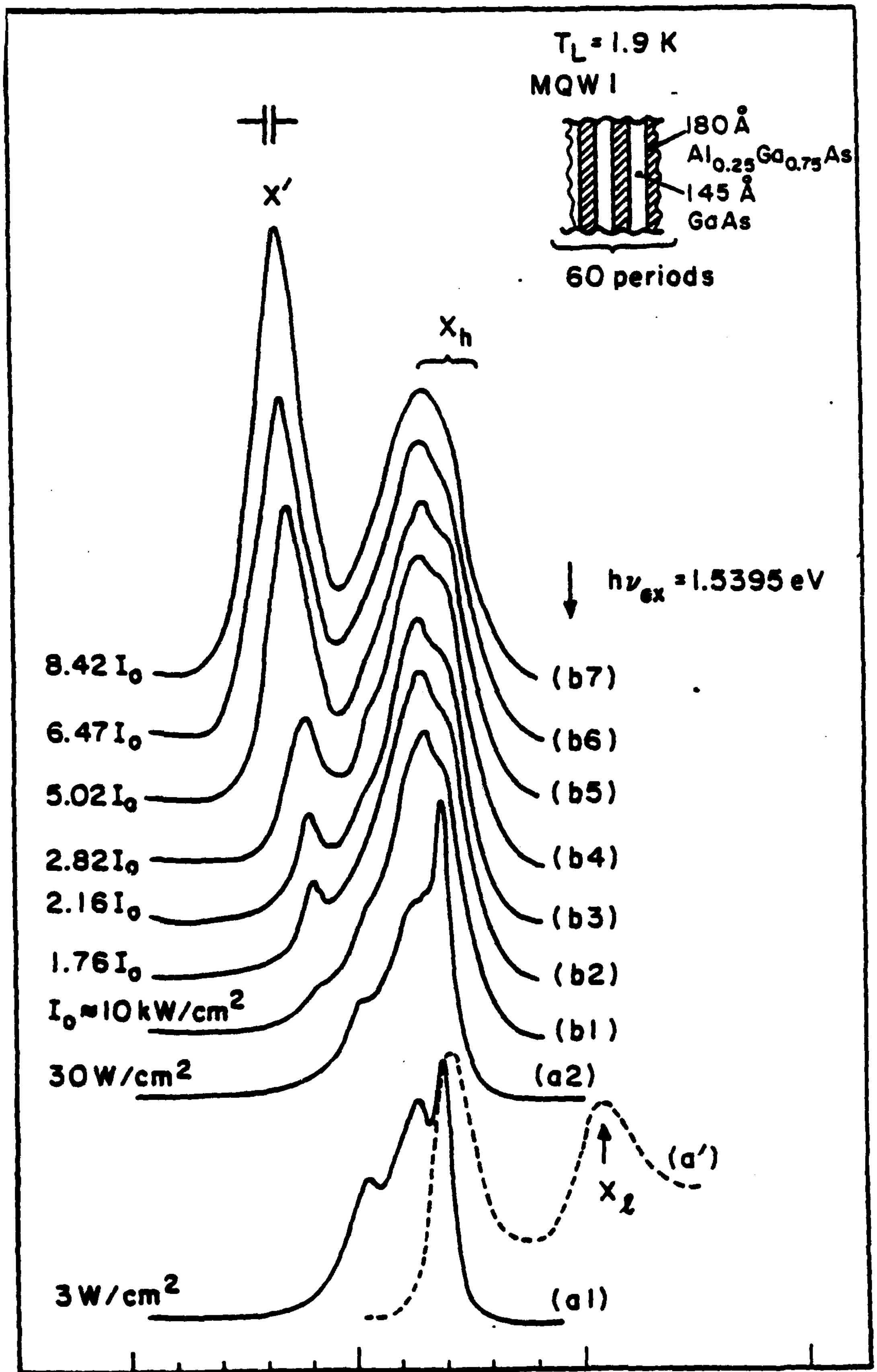
FIGURE CAPTION

(a'): absorption spectrum of MQW 1 at low intensity. X_h and X_ℓ are the heavy hole and the light hole exciton, respectively. (a1): and (a2): luminescence spectra of emission normal to the MQW layer plane (normal configuration) obtained with low intensity, CW 6328 Å laser excitation. The highest energy peak is the heavy hole exciton, and lower energy structures are extrinsic which saturate as I_{ex} is increased. (b): normal configuration luminescence spectra obtained with pulsed dye laser excitation at 1.5395 eV (near resonance excitation), showing X' emergence. (b) were smoothed; some structures are probably not real since they are well within the noise level.

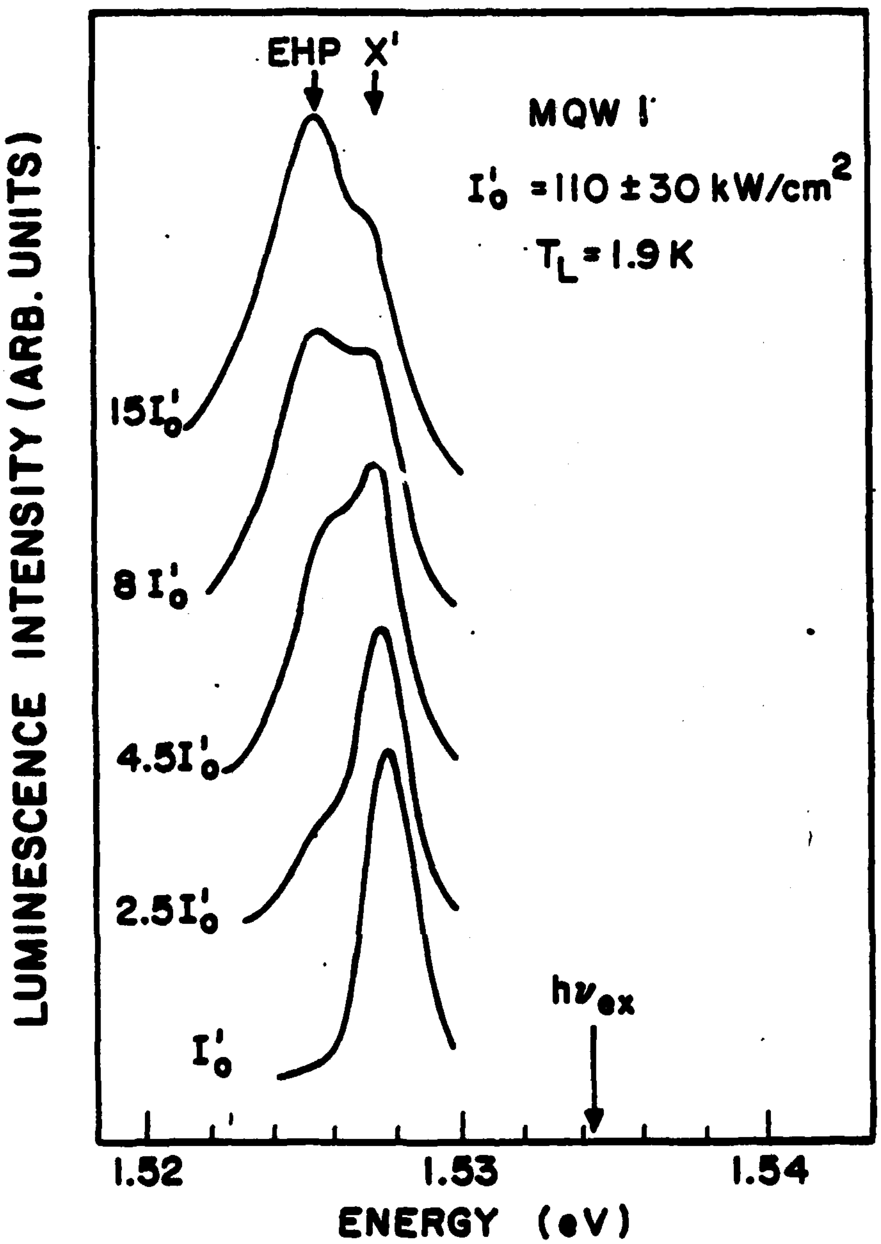
Normal configuration luminescence spectra at still higher excitation intensities where a second structure, identified as due to an EHP, appears and overwhelms X'.

(a) and (b): normal configuration luminescence spectra with $\hbar\nu_{ex}$=1.5395 eV (near resonance) and 1.92 eV (off resonance), respectively. $I_{ex} \approx 30\pm10$ kW/cm^2. The structure in (b) may be due to an EHP, thus different from that in (a), which is X'. (c) and (d): excitation spectra, showing X' normal configuration luminescence intensity, collected within the indicated band (1.5 meV wide) as a function of $\hbar\nu_{ex}$. The resonance at the heavy hole exciton X_h is also indicated by an arrow. (d) is vertically shifted for the sake of clarity.

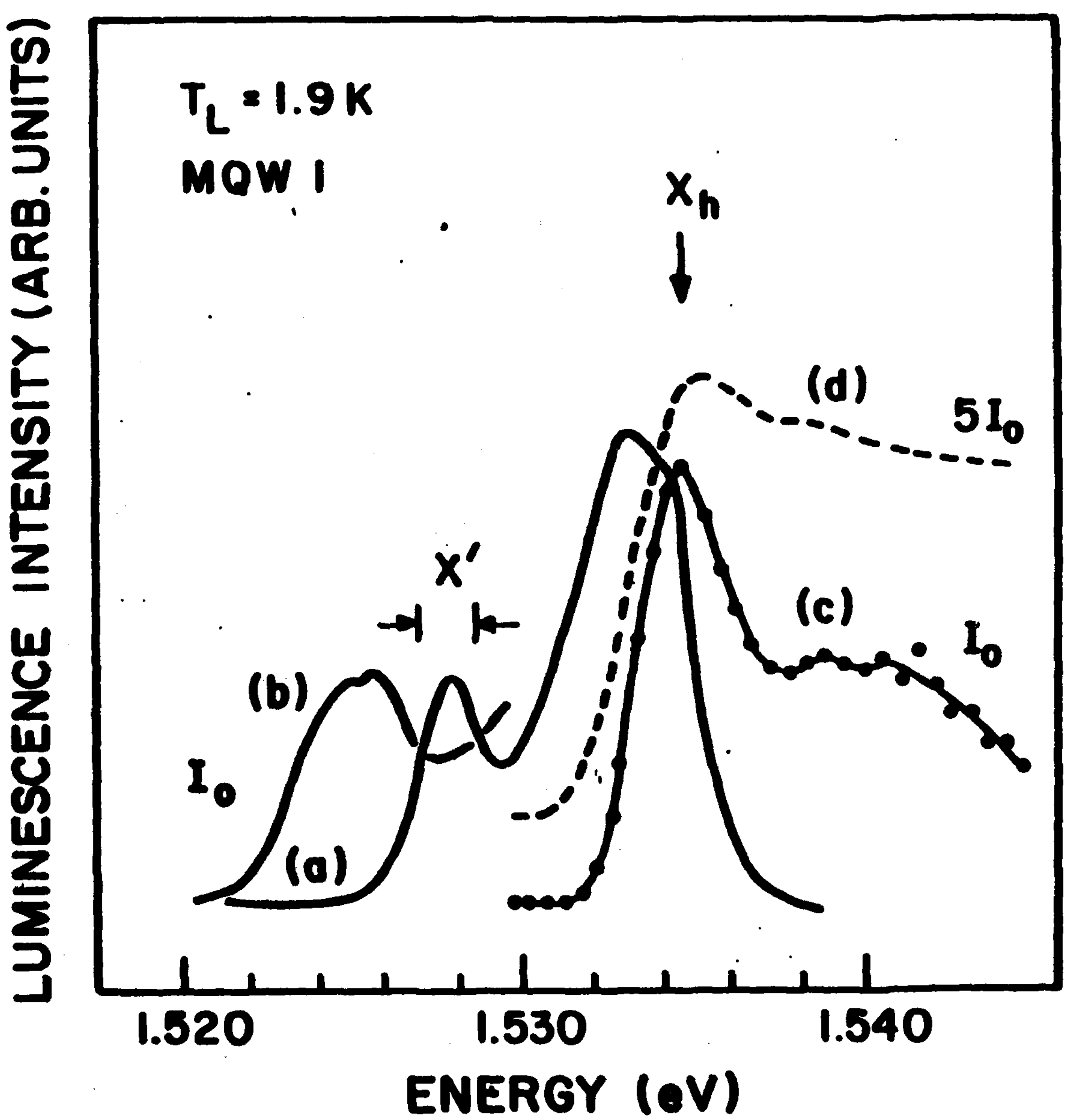
Zeeman shifts of X', the heavy hole exciton X_h and light hole exciton X_ℓ in MQW 1. Data on X_ℓ were obtained via low intensity excitation spectroscopy. The labels ±3/2,±1/2 represent spin quantum numbers of the hole. (a) is $2h\nu(X_h)-h\nu(X')$. According to the excitonic scattering model, conservation of energy requires (a) coincide with at least one of the two X_ℓ branches. But the deviation is clearly systematic. (b) is the theoretical calculation of free e-h pair energy.



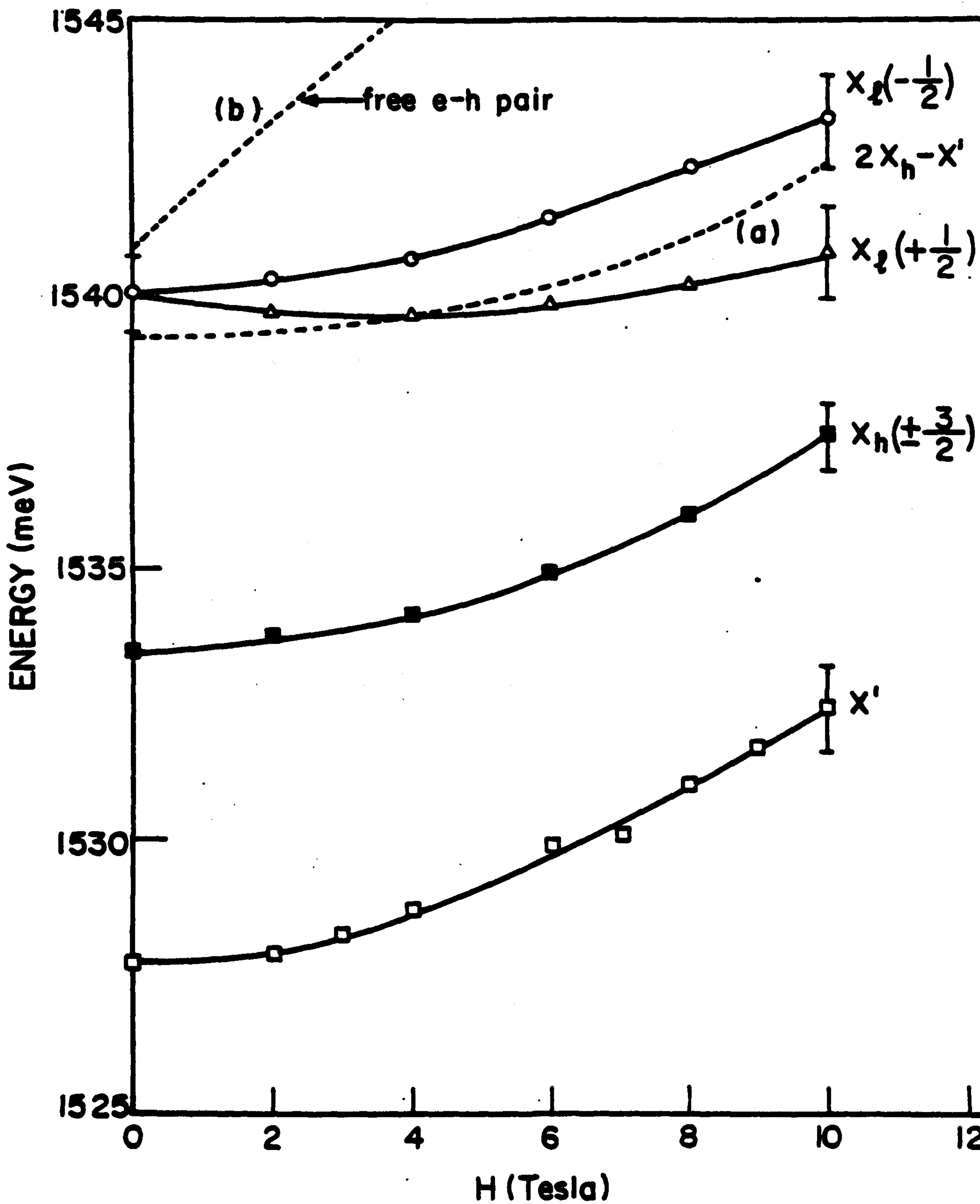
T_L = 1.9 K
MQW I
180 Å
$Al_{0.25}Ga_{0.75}As$
145 Å
GaAs
60 periods
X′
X_h
$h\nu_{ex}$ = 1.5395 eV
8.42 I_0
6.47 I_0
5.02 I_0
2.82 I_0
2.16 I_0
1.76 I_0
I_0 ≈ 10 kW/cm²
30 W/cm²
3 W/cm²
(b7)
(b6)
(b5)
(b4)
(b3)
(b2)
(b1)
(a2)
(a′)
X_ℓ
(a1)
1.520
1.530
1.540
1.550
ENERGY (eV)



EHP
X'
MQW I
I'o =110 ± 30 kW/cm2
TL = 1.9 K
15I'o
8 I'o
4.5I'o
2.5I'o
I'o
hνex
LUMINESCENCE INTENSITY (ARB. UNITS)
1.52
1.53
1.54
ENERGY (eV)



LUMINESCENCE INTENSITY (ARB. UNITS)
T_L = 1.9 K
MQW I
X_h
X′
(a)
(b)
(c)
(d)
I_o
I_o
5I_o
1.520
1.530
1.540
ENERGY (eV)



1545
1540
1535
1530
1525
0
2
4
6
8
10
12
ENERGY (meV)
H (Tesla)
(b)
free e-h pair
$X_\ell(-\frac{1}{2})$
$2X_h - X'$
(a)
$X_\ell(+\frac{1}{2})$
$X_h(\pm\frac{3}{2})$
X'

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