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20. Abstract

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PHOTOLUMINESCENCE STUDY OF CONFINED ELECTRON

HOLE PLASMA IN $Ga_xIn_{1-x}As$ HETEROSTRUCTURES

M. Gal, J.M. Viner and P.C. Taylor
Department of Physics
University of Utah
Salt Lake City, UT 84112

and

C.P. Kuo, R.M. Cohen and G.B. Stringfellow
Department of Materials Science and Engineering
University of Utah
Salt Lake City, UT 84112

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ABSTRACT

A low energy photoluminescence (PL) emission is observed in $Ga_xIn_{1-x}As$ heterostructures. The emission originates from an electron-hole plasma (EHP) confined in a 500 Å $Ga_yIn_{1-y}As$ layer between the InP substrate and a wider band-gap $Ga_xIn_{1-x}As$ layer. A line shape analysis of the EHP emission yields electronic temperatures which essentially coincide with the bath temperature. Linear polarization of the PL was observed which indicates a degree of strain in the confining layer. Studies in a magnetic field indicate that the carrier transport in the heterostructure studied is via free carriers and not via excitons.

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INTRODUCTION

$Ga_{.47}In_{.53}As$ lattice matched to InP is an important semiconducting compound, due to its large potential for a number of opto-electronic and high speed electronic applications. The main focus of recent spectroscopic work in these compounds has involved the study of excitonic and impurity related phenomena.[1-5] The binding energy of the free exciton has been determined from photoluminescence (PL) measurements, as well as binding energies for several acceptors.[1] We report a PL process which is due to an electron-hole plasma (EHP) confined to a thin $Ga_yIn_{1-y}As$ layer which is between an InP (larger band-gap) substrate and a larger band-gap layer of $Ga_xIn_{1-x}As$. This identification is a result of analyses of the intensity and the temperature dependences of the PL, the effect of a magnetic field on the heterostructure and the polarization properties of the emitted light.

The $Ga_xIn_{1-x}As$ layers were grown by organometallic vapor phase epitaxy (OMVPE). Because the carriers are confined to a thin layer one can observe PL from an EHP at much lower pumping intensities than is commonly the case for three dimensional samples. In addition, the observation of efficient PL from an EHP implies that competing non-radiative processes at the interfaces are unimportant. This observation thus provides a graphic demonstration of the high interfacial quality which can be achieved by the OMVPE growth technique.

In highly excited indirect-gap semiconductors, such as Ge and Si, a luminescence line, which appears at slightly lower energy than that of the free exciton, has been identified as an emission from an electron-hole liquid.[6] This situation is in contrast to direct gap materials, where the very short carrier lifetime usually prevents phase separation between the free exciton gas and the electron-hole liquid. At high intensities the excitons dissociate into an EHP, composed of free electrons and holes.

In a bulk semiconductor, substantial EHP density only occurs at very high excitation intensities due to the short lifetime and the rapid diffusion of the carriers out of the photoexcited volume. Typical excitation intensities in a bulk semiconductor can reach the kW or even the MW range.[7] By confining the carriers in a potential well, we have observed EHP luminescence at several orders of magnitude lower pump intensities and thus we were able to use C.W. excitation instead of the usual pulsed excitation. As a result, our measured spectra were independent of the time evolution of the exciton-EHP system that often complicates the description of this phenomenon.[7]

EXPERIMENTAL DETAILS

The $Ga_xIn_{1-x}As$ layers studied were grown by the OMVPE technique in an atmospheric pressure, horizontal reactor[8] using trimethylindium (TMIn), trimethylgallium (TMGa) and arsine (AsH_3) in a flowing stream of Pd purified H_2. The details are described in ref. 8. The flow rates of the reactants were: H_2, 2 liters/minute, AsH_3 (10% in H_2), 60 sccm, and TMGa (held at -11.5°C), 0.9 sccm, TMIn (held at 10°C), 125 sccm. In order to obtain the best combination of morphology and electrical and optical properties an initial 500 Å layer of $Ga_yIn_{1-y}As$ is grown (1 minute at a rate of 0.05 μm/min.) directly on the semiinsulating InP substrate at a temperature of 540°C.[8] The temperature was then raised to 650°C for the growth of a 1-1.5 μm layer of $Ga_xIn_{1-x}As$. Since the distribution coefficient is somewhat temperature dependent,[9] the TMIn flow rate was increased from 110 sccm for the 540°C layer to 125 sccm for the 650°C layer. However, the composition of the 540°C layer may still be somewhat In rich due to an anticipated lag in the arrival of the TMGa to the system because of its extremely slow flow rate. An attempt was made to alleviate this problem by turning the TMGa flow from

the vent to the reactor 5 seconds before the TMIn flow was switched into the reactor. Five seconds is the delay time calculated from the volume of the line between the TMGa source and the reactor and the TMGa flow rate. In spite of this precaution, the initial $Ga_yIn_{1-y}As$ layer probably has a variable composition with $y < x$ for much of the layer.

The results were obtained on several samples, the structure of which is shown in Fig. 1a. The composition of the layer grown directly onto the InP substrate was[10] approximately $y = 0.43$ ($Eg = 0.77$ eV), while for the top layer $x = 0.47$ ($Eg = 0.81$ eV). The InP substrate was semi-insulating except for one sample for which an n^+ substrate was employed.

Photoluminescence was excited by the 5145 Å line of an Ar^+ ion laser, the intensity of which was changed by using calibrated neutral density filters. The excitation density varied from 20 mW/cm^2 to 20 W/cm^2. The emitted light was dispersed by a Jobin-Yvon HR 320 grating monochromator and detected by a liquid nitrogen cooled Ge detector. The resulting signal was amplified with the usual lock-in technique and stored in a LeCroy 3500 signal averaging computer.

The sample was either immersed in superfluid helium or was in contact with the He gas. The temperature could be varied between 2 K and 80 K. In some cases, a magnetic field was applied to the sample using a superconducting magnet.

RESULTS AND DISCUSSION

For an unintentionally doped n-type sample at $T = 2$ K, Fig. 2b shows a typical PL spectrum excited at the lowest power densities ($P_1 = 20$ mW/cm^2). At these power levels only the emission from the top layer is observed. The spectrum of Fig. 2b shows an exciton line at $h\nu = 0.82$ eV, and an impurity-

related line lower in energy by about 20 meV. The excitonic nature of the dominant high-energy line is supported by the observed excitation and temperature dependence. The dependence of the emission intensity on the excitation intensity is very close to linear, while the temperature dependence shows the thermal dissociation of the exciton with a thermal activation energy equal to the binding energy of the exciton. These results are similar to the findings of Goetz et al.[1] Using similar arguments, it was previously shown,[1] that the lower energy broader line is due to a donor-to-acceptor transition. In these undoped OMVPE samples, the acceptor is probably zinc.[1]

The PL spectra change dramatically upon increasing the pump intensity. Figure 2a shows the result of changing the excitation intensity by a factor of 100. A new emission band appears at lower energies, which at the highest excitation intensities dominates the emission. The dependence of this new band on excitation power can be seen in Fig. 3., where the logarithm of the emission intensity is plotted as a function of wavelength for different pumping intensities. As can be seen from this figure, the peak of the emission moves to higher energy (lower wavelength) with increasing excitation intensity, while at the same time, the low energy "tail" of this emission changes very little with excitation intensity. Although the exact functional relation between the emission and excitation varied from sample to sample, the integrated emission intensity is strongly superlinear.

The temperature dependence of this new band was studied between 2 K and 80 K. The integrated intensity varied only slightly with temperature over this range (see Fig. 4), with some samples showing an increase in the emission intensity as the temperature was raised.

In light of the observations noted above, we believe that this new emission band is due to an EHP confined between the two higher band-gap

materials. This identification implies that the interface between the InP
substrate and the thin layer, and between the thin layer and the thicker
$Ga_{0.47}In_{0.53}As$ top layer are of high quality (i.e., with a low interface
recombination velocity). If this were not the case, then one would expect
competing surface-related processes to dominate the recombination. For a
semi-insulating InP substrate, and undoped, slightly n type InGaAs layers,
the heterostructure would look schematically like that shown in Fig. 1b.
(The effects of band-bending have been neglected in the figure for these
undoped layers.) Since the absorption coefficient is approximately 10^5 cm^{-1}
at the energy of excitation (hν = 2.41 eV),[11] the nominal penetration
depth of the laser light is approximately d $\approx$ 0.1 μm. Thus, at low pumping
intensities only the top layer is excited, and this excitation condition will
give rise to the exciton and impurity-related emission bands characteristic
of the top layer. At higher pumping intensities, an increasing number of
photogenerated carriers diffuses from the top layer and gets collected in
the potential well formed by the conduction band discontinuity. Since the
width of the potential well is only ~ 500 Å, the carrier _density_ becomes
substantial even at relatively low pumping power levels.

In order to evaluate the plasma density and plasma temperature of the
confined EHP, we have fitted the measured luminescence band to the expression
which describes radiative recombination from an EHP in the absence of momentum
conservation:[12]

$$I(\hbar\omega) = I_0 \int_0^{\hbar\omega'} D(E) \, D(\hbar\omega' - E) \left[1 + \exp(E_f^h - E^h)/kT \right]^{-1}$$

$$\left[1 + \exp[(\hbar\omega' - E - E_f^e)/kT] \right]^{-1} dE \tag{1}$$

where D(E) and D($\hbar\omega$ - E) are the (3 dimensional) density of states in the

conduction and valence bands, and $E_f^{e,h}$ is the electron (hole) quasi-Fermi energy determined from

$$N = c \int_0^\infty x^{1/2} \left[\exp[x - E_f)/kT] + 1 \right]^{-1} dx \tag{2}$$

In Eq. 2, N is the carrier density, E_g' is the renormalized band-gap, that includes the electron-hole exchange and correlation effects and $\hbar\omega' = \hbar\omega - E_g'$.

Initially, the PL spectrum at 10 K was fit with reasonable values of the three fitting parameters (plasma density, plasma temperature and renormalized band gap). The argeement between the model and experiment is shown in Fig. 5. The fits at elevated temperatures (see Fig. 5) were then obtained by adjusting only the renormalized gap to match the slight temperature dependence of the low energy onset of the PL. These small changes in the renormalized gap affect only the position in energy of the PL and not its width or shape. In addition, a small decrease in the renormalized gap with temperature is to be expected primarily because of the temperature dependence of the actual gap.

This model describes well the experimentally observed emission, except for the low energy "tail" region. It is interesting to note, that the plasma temperature used in fitting agrees closely with the bath temperature for all samples and temperatures studied. In most highly excited semiconductors, when the excitation energy is well above the band gap, the electronic temperature is found to increase above the bath temperature.[7] This increase is due to the heating of the plasma (either directly or indirectly) by the hot electrons (and holes) that are created by the exciting photon. In our case however, the energy difference between the laser photon and the band-gap of GaInAs is dissipated in the top layer and subsequently by the He, and thus the carriers captured into the potential-well are "cool." The excess

energy of the electrons and holes forming the EHP is only ⌐f the order of 50
meV, which is not enough to heat up substantially the electronic system.

The discrepancy between the experiment and the theory, which is observable
on the low energy tail of the emission curves, can be the result of several
phenomena. Band-gap fluctuations created by alloy compositional fluctuations
are likely candidates for causing a low energy tail in the emission spectra,
as is the case for other alloy systems.[13],[14] In our system an additional
source may be the band gap variation produced by spatially-varying strain
which is known to change the band gap.[15] The existence of strain in some of
our samples was confirmed by observing a degree of linear polarization of the
emission while for the same samples the exciton luminescence (of the
substrate was unpolarized (see Fig. 6). The degree of linear polarization
varied from sample to sample, and no attempt was made to correlate the degree
of polarization with other sample parameters.

We can demonstrate that the observed emission is indeed from the
quantum well, and not from an as yet unidentified emission of the top layer,
though not by externally hindering the drift of the carriers toward the lower
gap layers. An effective way of reducing the number of carriers arriving at
the potential well can be achieved by applying a magnetic field parallel to
the sample layers, i.e., normal to the diffusion path of the carriers. As a
result, the diffusion length is reduced by the magnetic field:[16]

$$L_D(H) = \frac{L_D(0)}{\omega_c \tau} \tag{3}$$

where ω_c is the cyclotron frequency and τ is the mean time between scattering
events. At a field value of H = 2.5 Tesla, using $\tau = 10^{-13}$ sec[17] we find that
$L_D(H)$ is reduced by a factor of 3 or 4 relative to the field free case. Thus

in a magnetic field we expect the EHP luminescence intensity to decrease and shift to lower energy, and the exciton luminescence of the top layer to increase.

Our results, conducted in a magnetic field of 2.5 T parallel to the layer structure, confirm this model, as can be seen in Fig. 7. At this value of the magnetic field, the EHP luminescence was decreased by approximately 40%, while the exciton luminescence was found to increase. We did not attempt a quantitative anaysis because the effect of the magnetic field on the radiative transitions (Zeeman effect) is not known for this heterostructure. It is interesting to note, however, that our results imply that the carrier transport through the heterojunction is mainly due to free electrons and holes as opposed to exciton transport, which would be insensitive to the magnetic field. In a similar experiment, exciton transport was observed in GaAlAs-GaAs heterostructures by Lee et al.[18]

When an n^+ InP substrate is employed instead of a semi-insulating substrate the conduction band discontinuities are greatly reduced[19] and neglecting band bending, the potential well for electrons becomes negligible. This lack of confinement of the electrons is reflected in the luminescence spectrum. At low excitation intensities PL is observed from the excitonic and impurity-related processes in the top layer (Fig. 8c). At higher excitation intensities, the emission characteristic of the lower band-gap layer appears (see Fig. 8b and 8a). In this case, however, no EHP emission is observed (at least in the excitation intensity range studied) but instead the PL consists of the exciton and impurity emissions characteristic of the lower band gap layer. The identification of these emissions was confirmed by their respective temperature and excitation-intensity dependences.

Finally, when a new line was introduced to provide additional H_2 flow just past the TMGa bubbler, the intensity of the EHP emission was decreased by several orders of magnitude. This supports the conclusion that the EHP emission is, indeed, due to the presence of a layer having a lower Ga content next to the substrate.

SUMMARY

We have observed EHP emission from a 500 Å thick layer of $Ga_yIn_{1-y}As$ confined between InP and $Ga_{0.47}In_{0.53}As$ layers. Since the emission was observable even at low power levels (> 300 mW/cm^2), we conclude that the collection efficiency of the potential well was substantial, and thus the heterojunction interface was of high quality.

A line shape analysis of the EHP emission yields electronic temperatures which essentially coincide with the bath temperatures. Linear polarization was observed which indicates a degree of strain in the layer. This strain may also expain the slight discrepancy at low energies between the theoretical and the experimentally-observed line shapes. Studies in a magnetic field indicate that the carrier transport in the top layer ($Ga_{0.47}In_{0.53}As$) is via free carriers and not via excitons.
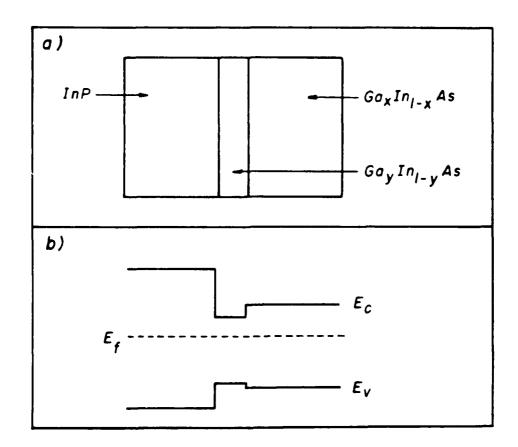
ACKNOWLEDGMENTS

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11. Since the absorption coefficint at $h\nu$ = 2.40 eV in both GaAs and InAs is approximately 10^5 cm^{-1}, we believe it is reasonable to assume the same value for $Ga_{0.49}In_{0.51}As$.

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FIGURE CAPTIONS

Fig. 1.   Structure of typical samples. Dimensions are given in the text.

Fig. 2.   Typical PL spectra under two different excitation intensities:
           (a) excitation intensity I_0 = 2 W/cm^2, (b) I_0 = 20 mW/cm^2.

Fig. 3.   Low energy emission lineshape as a function of excitation intensity.

Fig. 4.   Temperature dependence of the exciton luminescence ($I_{exciton}$) and
           the low-energy PL band (I_{EHP}). The curves are only aids to the eye.

Fig. 5.   Luminescence spectra at constant excitation intensity for different
           temperatures. The circles represent the experimental points, while
           the solid curves are the calculated EHP lineshapes using parameters
           given in the figure and described in the text.

Fig. 6.   Polarization dependence of the EHP lineshape.

Fig. 7.   The effect of a magnetic field on the EHP emission intensity. See
           text for details.

Fig. 8.   PL spectra under three different excitation intensities for samples
           grown on n$^+$ substrate.
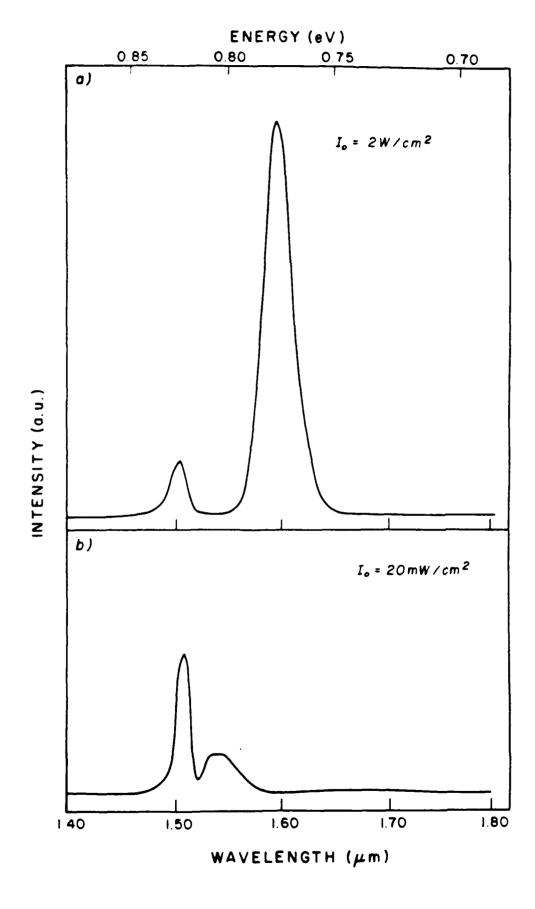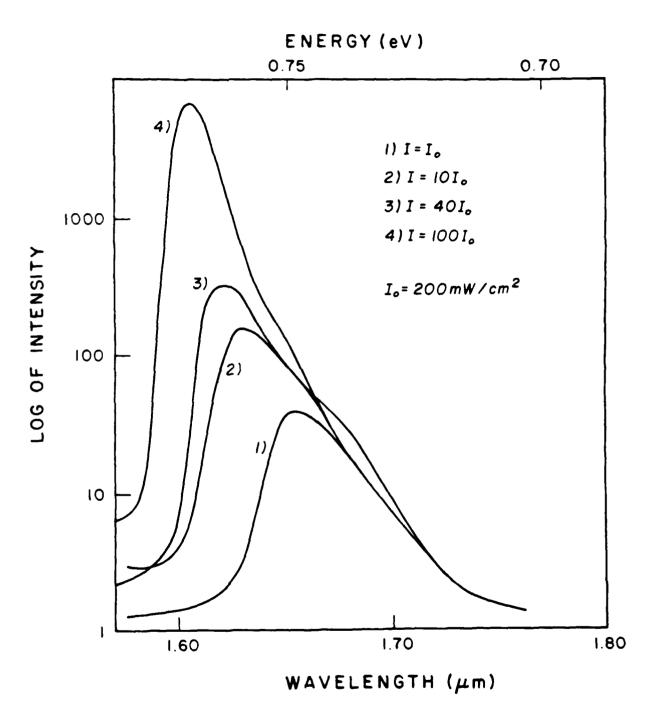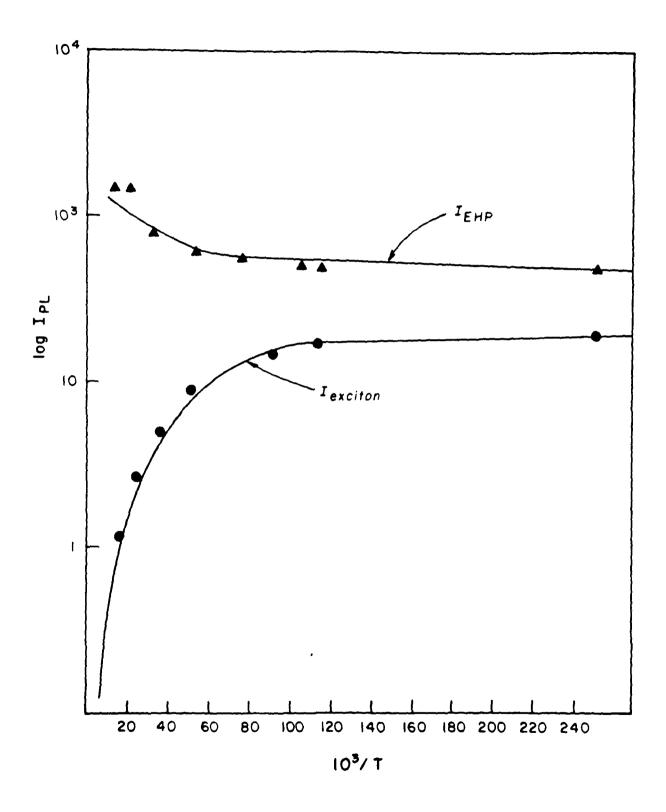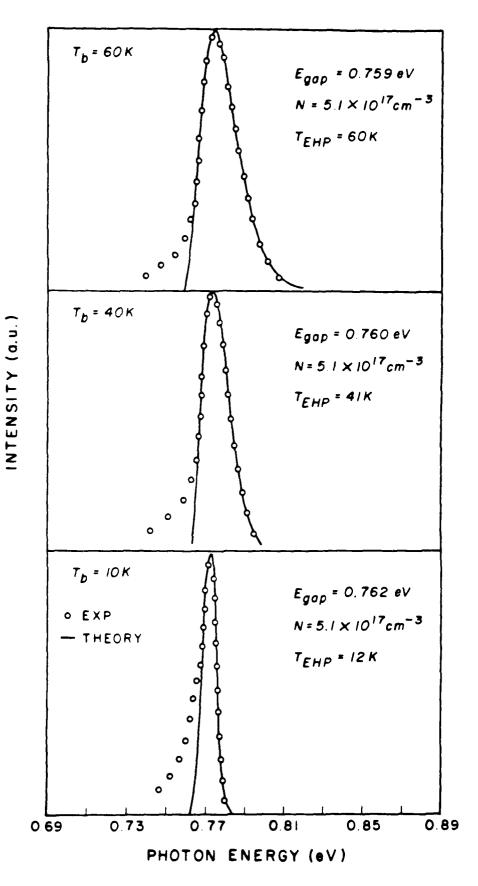
a)

InP → $Ga_xIn_{1-x}As$

$Ga_yIn_{1-y}As$

b)

E_c

E_f

E_v

Fig. 1.

Fig. 2.

Fig. 3.

Fig. 1.

$T_b = 60K$

$E_{gap} = 0.759\, eV$

$N = 5.1 \times 10^{17} cm^{-3}$

$T_{EHP} = 60K$

$T_b = 40K$

$E_{gap} = 0.760\, eV$

$N = 5.1 \times 10^{17} cm^{-3}$

$T_{EHP} = 41K$

$T_b = 10K$

○ EXP

— THEORY

$E_{gap} = 0.762\, eV$

$N = 5.1 \times 10^{17} cm^{-3}$

$T_{EHP} = 12K$

INTENSITY (a.u.)

PHOTON ENERGY (eV)

0.69   0.73   0.77   0.81   0.85   0.89

ENERGY (eV)

INTENSITY (a.u.)

WAVELENGTH (μm)

E ∥ layer

E ⊥ layer

ENERGY (eV)

WAVELENGTH (μm)

Fig. 7.

a) $T_b = 4.8\,K$
$I_o = 20\,W/cm^2$

b) $T_b = 4.5\,K$
$I_o = 1\,W/cm^2$

c) $T_b = 3.9\,K$
$I_o = 30\,mW/cm^2$

INTENSITY (a.u.)

PHOTON ENERGY (eV)

0.69   0.75   0.79   0.81   0.85   0.89

Fig. 5.

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