SLAC-PUB-5731
WISC-EX-92-322
February 1992
(A)

Electron Spin Polarization in Photoemission from Strained GaAs grown on GaAsP*

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Submitted to *Physical Review Letters*

* This work was supported in part by the U. S. Department of Energy under contract numbers DE-AC03-76SF00515 (SLAC), and DE-AC02-76ER00881 (UW).

ABSTRACT

Spin-polarized electron photoemission has been investigated for strained GaAs epitaxially grown on a $GaAs_{1-x}P_x$ buffer. The lattice mismatched heterostructure results in a highly strained epitaxial layer and significant enhancement of electron-spin polarization is observed. The effect of epitaxial layer strain is studied for a variety of samples with epitaxial layer thicknesses varying from 0.1 μm to 0.3 μm and the phosphorous concentration x varying from 0.21 to 0.28. Electron spin polarization as high as 90% has been observed. The 0.3 μm-thick sample, well in excess of theoretical estimates for the critical thickness for pseudomorphic growth, reaches an electron-spin polarization of 80%, demonstrating a significant persistence of lattice strain.

PACS numbers: 29.25.Bx, 29.75.+x, 73.60.Br, 79.60.Eq

Recently electron spin polarization in excess of 70% has been observed in photoemission from a thin strained epitaxial InGaAs layer grown on a GaAs substrate.[1] The observed polarization enhancement is a result of strain in the InGaAs layer due to a small (0.9%) lattice mismatch of the epitaxial layer relative to the GaAs substrate. Strain induces a valence band splitting which permits optical excitation of a single band transition, thus leading potentially to 100% polarization of the photoemitted electrons. A similar polarization enhancement was subsequently observed in photoemission from epitaxial GaAs grown on a thick GaAsP buffer layer.[2] Although these pioneering experiments have demonstrated strain enhanced electron spin polarization, there has been no systematic investigation of the conditions required to optimize the heterostructure parameters for maximum polarization and high quantum efficiency.

This letter reports the first systematic study of the effects of strain on electron polarization using samples with a heterojunction of GaAs epitaxially grown on a GaAsP buffer layer. Samples with varying epitaxial layer thickness and varying buffer layer phosphorous concentration were used to cover a range of strains, and the electron spin polarization and quantum efficiency were measured for each sample as a function of excitation photon energy.

The theory relating strain to band structure has been discussed extensively in the literature.[3] The band structure of the strained layer is altered such that the heavy-hole and light-hole valance bands are no longer degenerate in energy at the Γ point, and the energy splitting is then proportional to the strain. A suitably thin epitaxial layer of GaAs grown on a GaAsP substrate incorporates a biaxial compressive strain in the plane of the interface and a tensile strain along

the growth direction. The strain dependent energy levels of the heavy-hole (HH) and light-hole (LH) bands relative to the conduction band (C) are given by:[4]

$$E^{C,HH} = E_0 + \delta E_H - \delta E_S,$$

$$E^{C,LH} = E_0 + \delta E_H + \delta E_S - (\delta E_S)^2/2\Delta_0 + \cdots$$

where E_0 is the direct band gap of fully relaxed GaAs and Δ_0 is the spin orbit splitting. The quantities δE_H and δE_S represent the hydrostatic shift of the center of gravity of the $P_{3/2}$ multiplet and the linear splitting of the $P_{3/2}$ multiplet respectively, and are given in terms of the biaxial strain ϵ parallel to the interface by:

$$\delta E_H = 2a[(C_{11} - C_{12})/C_{11}]\epsilon$$

$$\delta E_S = b[(C_{11} + 2C_{12})/C_{11}]\epsilon$$

where the parameters a and b are the interband hydrostatic pressure and uniaxial deformation potentials, respectively, and the C_{ij} are the elastic-stiffness constants appropriate to the GaAs crystal structure. Since the biaxial strain ϵ is compressive for the present structures, the effect of strain is to increase the band gap energy of GaAs and to remove the degeneracy of the heavy-hole and light-hole levels such that $E^{C,HH} < E^{C,LH}$. In the present experiment the lattice mismatch, and hence the strain, is determined by the phosphorous concentration of the buffer layer on which the epitaxial layer is grown.

When a lattice mismatched layer is grown on a substrate, the misfit between the layers is accommodated by elastic strain in the epitaxial layer and pseudomorphic growth takes place. However, if the epitaxial layer exceeds a characteristic critical thickness, the stored elastic strain in the epitaxial layer is relieved by producing misfit dislocations. Theoretical considerations based on thermodynamic equilibrium arguments at the heterostructure interface have been used to calculate the equilibrium critical thickness,[5] and it has been experimentally confirmed that the dislocation density in the structure increases rapidly when the critical thickness is exceeded.[6] However, there is experimental evidence indicating that the lattice strain is preserved to thicknesses beyond the critical thickness, although with a significant epitaxial layer dislocation density. Significant strain relief can be observed only above a second critical thickness (the non-equilibrium critical thickness) which is roughly an order of magnitude greater than the equilibrium critical thickness.[7] Several models have been proposed to explain the persistence of lattice strain.[8]

The samples for the present experiment were grown by the Spire Corporation[9] using Metal-Organic-Chemical-Vapor-Deposition (MOCVD). A 0.25 μm thick p-type GaAs buffer layer was grown on a (100) p-type GaAs substrate oriented two degrees towards (110) direction. In order to produce a strain relieved $GaAs_{1-x}P_x$ layer on GaAs, a 2.5 μm-thick $GaAs_{1-x}P_x$ layer was grown with an increasing phosphorous fraction from 0 to x to accommodate the lattice mismatch, followed by an additional 2.5 μm-thick $GaAs_{1-x}P_x$ layer with a fixed phosphorous fraction. The lattice mismatched GaAs epitaxial surface layer was then grown on this buffer. The epitaxial surface layers were p-type doped with zinc to a value of $4\text{-}6\times10^{18}\text{cm}^{-3}$. In order to preserve an atomically clean surface the samples were anodized to form

an oxide layer of about 100 Å on the GaAs surface.[10] The oxide layer was later removed as described below.

The samples were analyzed with X-ray diffractometers at the Spire Corporation and in the Department of Materials Science and Engineering at the University of Wisconsin. Asymmetric (115) reflections were used to measure the phosphorous fraction of the $GaAs_{1-x}P_x$ buffer layer, and (004) reflections were used to determine the epitaxial layer strain as described in Ref. 1. Table 1 summarizes the parameters for the five samples studied: the phosphorous fraction, the GaAs epitaxial layer thickness, the lattice mismatch, the measured strain, the calculated gap energies[11] of the heavy and light-hole bands relative to the conduction band, and the equilibrium critical thickness (h_c^{eq}). The measured strains represent the most probable value inferred from the X-ray analysis. The phosphorous fraction was chosen so that the energy splitting between heavy and light-hole bands was greater than 50 meV. The GaAs epitaxial layer thicknesses were chosen to be both greater and less than the non-equilibrium critical thickness experimentally observed by Orders and Usher.[7]

The electron spin-polarization was measured by Mott scattering at 65 keV in a system described elsewhere.[12] Prior to installation in the system, the sample was degreased sequentially in boiling solutions of trichloroethylene, acetone, and methanol. The protective oxide layer was then removed in ammonium hydroxide, and the sample was rinsed in distilled water and methanol. After the sample installation the gun was baked at 220°C for about 80 hours and at 150°C for 24 hours to achieve the necessary ultrahigh vacuum. During the bake, the sample was maintained at about 270°C by a resistive heater at the back of the cathode

support structure. The final pressure during the subsequent polarization measurements was less than 10^{-10} Torr. The cathode was activated to obtain a negative electron affinity surface using cesium and nitrogen trifluoride. Prior to activation, the cathodes were heat-treated typically for 2 hours at 450°C. This relatively low temperature was used to protect the strained layer structure. For bulk GaAs, temperatures in the range of 600-650°C are normally used.

Fig. 1 shows the measured spin polarization (data points) and cathode quantum efficiency (solid curve) at room temperature as a function of excitation photon energy for sample 3. The experimental uncertainty shown in the figure is the statistical error; for most points $\delta P_e < 1\%$. There is an overall absolute uncertainty $\delta P_e/P_e$ of 5% in the polarization. The band gap energy of GaAsP and the calculated heavy-hole band gap energy expected for this sample are shown by the arrows in the figure. In the energy region greater than about 1.7 eV, both GaAsP and GaAs layers contribute to the photoemission, resulting in an increase of the quantum efficiency and a small decrease in the polarization. In the energy region smaller than 1.7 eV, the photoemission from GaAsP diminishes sharply as the excitation photon energy decreases, and the major contribution to the photoemission can come only from the GaAs layer. The polarization is observed to increase sharply at about 1.54 eV reaching 90% at about 1.46 eV. The sharp polarization enhancement at 1.54 and the maximum polarization at 1.46 eV correspond to the expected gap energies $E^{C,LH}$ and $E^{C,HH}$, respectively. The photocurrent lifetime was also measured for this sample at a wavelength of 825 nm yielding an effective lifetime of 168 hours.

Fig. 2 shows the measured electron spin polarization for all the samples at

room temperature. The polarization of samples 1, 2, and 3 shows a systematic shift of the location of the polarization enhancement toward higher energy as the phosphorous fraction is increased. This shift is consistent with the expected change in the energy gap $E^{C,LH}$ due to increased strain. The maximum polarization at the photon energy corresponding to the gap energy $E^{C,HH}$ increases from 82% to 90% as the phosphorous fraction is increased, most likely due to a more selective excitation of the heavy-hole band, since the heavy-hole light-hole splitting increases from 50 meV to 67 meV for these samples.

Although the buffer layers for samples 4 and 5 have the same phosphorous fraction as sample 2, the polarization of these samples shows a systematically different behavior. The polarization enhancement shifts toward lower energy as the GaAs thickness is increased from 0.15 μm (sample 2) to 0.20 μm(sample 4) and to 0.30 μm (sample 5). For sample 5, photoemission is observed beyond the expected band gap energy $E^{C,HH}$. This is a strong indication that the lattice is partially relaxed and that the heavy and light-hole band energies are merging towards the values expected for relaxed GaAs. However, even though there is considerable relaxation of the strain, the maximum polarization of both samples reaches more than 80%. Since the epitaxial layer of sample 5 is about 30 times thicker than the equilibrium critical thickness, this high polarization demonstrates a significant persistence of lattice strain.

Fig. 3 shows the measured quantum efficiency as a function of excitation photon energy for all the samples. As expected, the quantum efficiency increases with increasing GaAs epitaxial layer thickness. However, the gain in quantum efficiency in the high polarization region is offset by a commensurate decrease in polarization

due to the increased relaxation of the sample strain. For the present samples, the highest quantum efficiency that corresponds to at least 80% polarization is 0.13%, measured for sample 4. The sample to sample reproducibility of the quantum efficiency demonstrates that the anodization and stripping procedure reliably results in a clean surface.

A strained epitaxial layer thicker than the equilibrium critical thickness is metastable, and lattice strain may relax under the heat-treatment required for cathode activation. Such a strain relaxation from annealing has been reported for strained heterostructures by J. M. Baribeau et al. in Ref. 7. To study the effect of heating on sample polarization, sample 5 (0.30 μm epitaxial layer thickness) was heat-cleaned at successively higher temperatures up to 570°C, the cathode activated, and the spin-polarization remeasured. The observed photon energy dependence of the polarization enhancement was reproducible and the difference in maximum polarization was only $\approx 5\%$, indicating that the sample strain was stable over this temperature range of heat cleaning even though significant strain relief was already present at room temperature. However, it was observed that the sample 5 polarization decreased from a maximum of 80% to 62% as the excitation photon energy was decreased, approaching the polarization expected for fully relaxed GaAs.

Heavy doping produces significant potential fluctuations in the conduction and valence bands (band tailing effect).[13] The band gap energy smearing due to the band tailing for the present samples is estimated to be about 30 meV at room temperature. A smearing of this magnitude is not negligible in comparison to the energy splitting between the heavy and light-hole bands, and may limit the

maximum polarization. To study the possible effect of this smearing on the spin polarization, the polarization of samples 3 and 4 was also measured at liquid nitrogen temperature (77°K), where band tailing effects should be significantly smaller. While no significant change in maximum polarization was observed, the energy shift of the polarization enhancement was consistent with the increased band gap energy obtained at 77°K.

In conclusion, electron spin polarization as high as 90% has been observed for strained GaAs grown epitaxially on a thick GaAsP buffer layer. The effect of epitaxial layer thickness on polarization has been systematically studied, and a polarization enhancement due to strain is observed for epitaxial layer thicknesses considerably greater than the theoretical expectation for the equilibrium critical thickness. The strain enhanced electron spin polarization measured at room temperature was stable with respect to temperature cycling of the samples, and did not significantly change even though heat cleaning temperatures were varied from 400-570°C. The polarization was also measured at 77°K, and the value of the maximum polarization was unchanged. The highest quantum efficiency that corresponds to at least 80% polarization was 0.13%.

We would like to thank Dr. S. Vernon of the Spire Corporation for discussions of the sample growths conditions. We thank Prof. R. J. Matyi of the University of Wisconsin for many discussions and the X-ray diffraction analysis of the samples. P. D. Moran provided skilled help in the X-ray analysis. We would like to thank R. Mair for assistance in the measurements and sample characterization. We thank Profs. L. W. Anderson and J. E. Lawler of the University of Wisconsin for the loan of a laser, and G. J. Collet of SLAC for skillful technical support. This work

was supported in part by the U. S. Department of Energy under contract numbers DE-AC03-76SF00515 (SLAC), and DE-AC02-76ER00881 (UW).

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13. See Ref. 11 p. 131.

Table 1. Strained GaAs samples

Sample	1	2	3	4	5
x	0.210	0.243	0.279	0.244	0.238
GaAs thickness (μm)	0.11	0.15	0.11	0.20	0.30
Lattice mismatch (%)	0.76	0.88	1.01	0.88	0.86
Measured strain (%)	76	85	87	81	61
$E^{C,HH}$ (eV)	1.46	1.47	1.48	1.47	1.46
$E^{C,LH}$ (eV)	1.51	1.52	1.54	1.52	1.52
h_c^{eq} (Å)	133	110	92	110	113

FIGURE CAPTIONS

Fig. 1 Electron spin-polarization as a function of excitation photon energy (data points), and cathode quantum efficiency as a function of excitation photon energy (solid curve) for sample 3. The band gap energies of GaAsP (dashed arrow) and the calculated heavy-hole band gap energy (solid arrow) are indicated in the figure.

Fig. 2 Electron spin-polarization as a function of excitation photon energy for all the samples; sample 1 (cross), sample 2 (diamond), sample 3 (open circle), sample 4 (square), and sample 5 (solid circle).

Fig. 3 Cathode quantum efficiency as a function excitation energy for all the samples: sample 1 (long dash), sample 2 (dotdash), sample 3 (solid), sample 4 (short dash), and sample 5 (dotted).

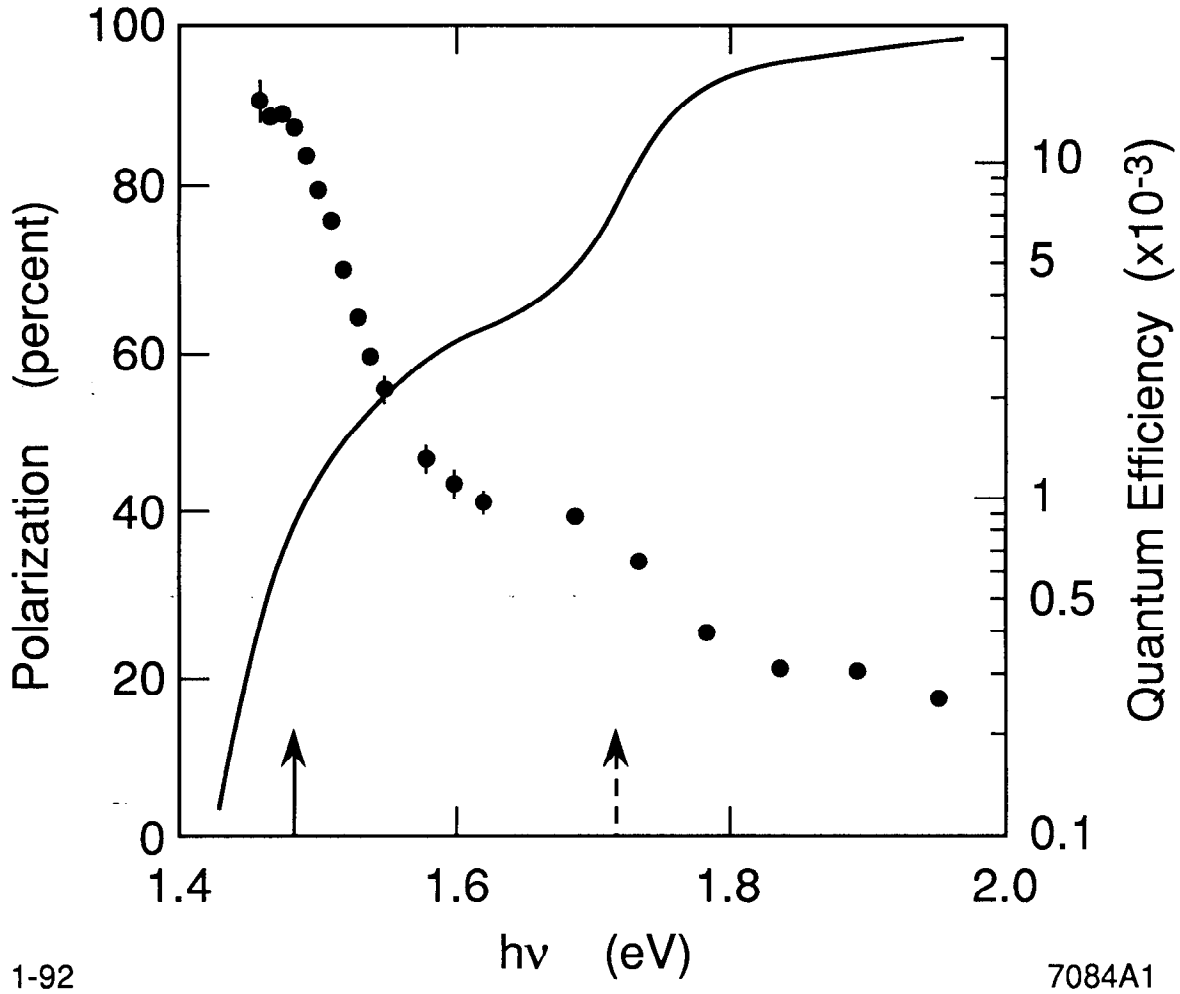
100
80
60
40
20
0
Polarization (percent)
10
5
1
0.5
0.1
Quantum Efficiency (x10-3)
1.4
1.6
1.8
2.0
hν (eV)
1-92
7084A1

Fig. 1

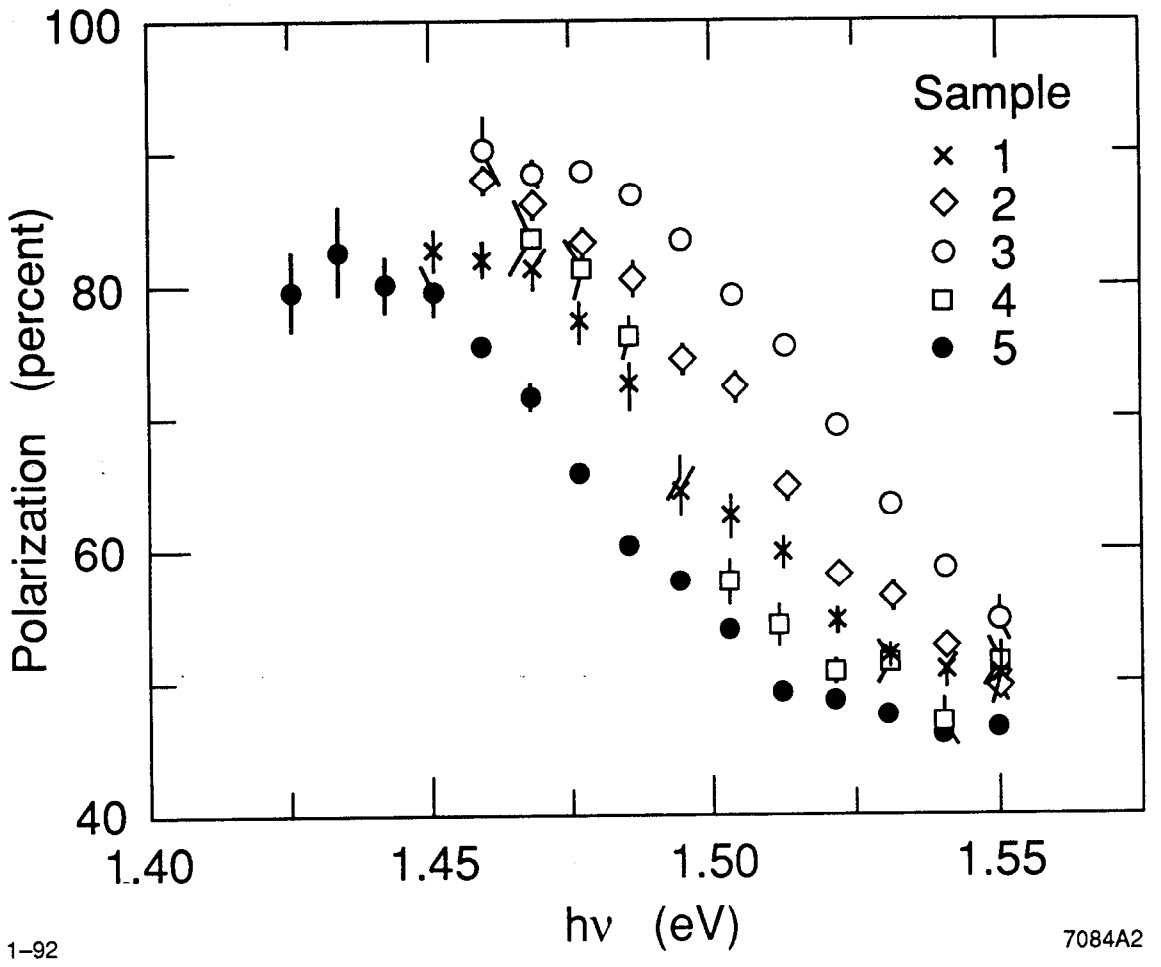
100
80
60
40
Polarization (percent)
1.40
1.45
1.50
1.55
hν (eV)
Sample
1
2
3
4
5
1–92
7084A2

Fig. 2

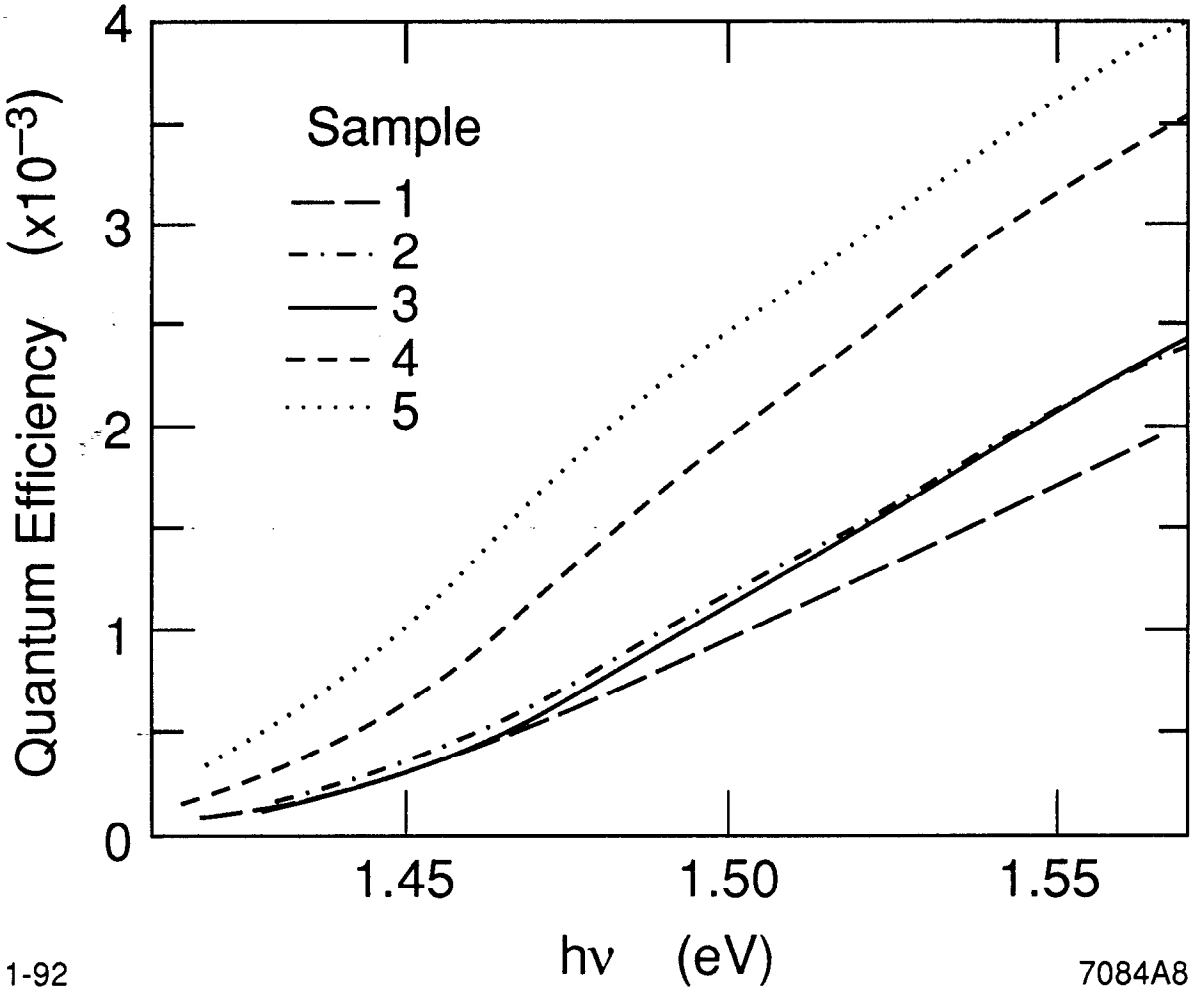
Quantum Efficiency (x10^{-3})
4
3
2
1
0
Sample
1
2
3
4
5
1.45
1.50
1.55
hν (eV)
1-92
7084A8

Fig. 3