Title

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DISCLAIMER

The Effects of Dopants and Defects on Light-induced Metastable States in a-Si:H

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Using photothermal deflection spectroscopy we measure the gap-state optical absorption of light-induced metastable defects in undoped, singly doped, and compensated a-Si:H. We observe an enhancement in the gap-state absorption after illumination which is shown to be due to the creation of *new* silicon dangling bond defects and *not* to a shift in the Fermi level. The results provide evidence that the doping level influences the light-induced defect formation mechanism, and imply that *simply* breaking Si-Si bonds may not be the primary mechanism.

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I. Introduction

Reversible light-induced changes in amorphous semiconductors have attracted much attention. For both the chalcogenide glasses and amorphous hydrogenated silicon (a-Si:H), prolonged illumination creates metastable defect states which disappear upon annealing. In the case of the chalcogenide glasses, the defects are attributed to reversible structural change [1]. For a-Si:H, the mechanism is still not yet understood despite numerous experimental studies [2] . This effect was first reported by Staebler and Wronski [3] [4] who observed a decrease in the dark and photo- conductivities after several hours of illumination, with a subsequent recovery after annealing at elevated temperatures. Electron spin resonance (ESR) studies [5] reported that after illumination there is an increase in the ESR signal associated with Si dangling bond defects which lie in the gap. Photoluminescence measurements [6] observed a shift in peak energy from 1.4 to 0.9 eV. Other light-induced changes include a drop in the electron lifetime [7], a reduction in the diffusion length of holes[8], an increase in the density of gap states as measured by field effect[9], and a deterioration of solar cell performance [10].

The various changes in the properties of a-Si:H after illumination are generally consistent with an increase in the number of Si dangling bonds. However, it has been suggested that Fermi level shifts could account for this apparent increase [11] [12]. Thus, the dangling bond density could remain constant and the changes after illumination would be due to a shift in the Fermi level into a region of higher defect-state density. Another interesting question is the nature of the interaction between the pre-existing defects that are present in the annealed state of the material and those induced by illumination.

In order to examine these issues further, we used the sensitive optical technique of Photothermal Deflection Spectroscopy (PDS) [13], to measure the gap-state absorption and monitor the changes in the spectra due to illumination. This measurement [14] provides information about the identity of the defect (Si

dangling bond), the defect density, and its energy level in the gap. It is necessary to use PDS for these measurements since the relatively low density of gap-states exhibits only weak absorption. Furthermore, the optical measurement is not strongly dependent on the position of the Fermi level, unlike, for example, deep level transient spectroscopy (DLTS). In the case of DLTS, the samples must be doped n-type and the changes in the Fermi level can yield different spectra even though the density of states may not change. Furthermore, absorption can occur from both the valence band to unoccupied defect levels, as well as from occupied levels to the conduction band. This allows absorption to probe states which would be otherwise undetected. Another advantage is that surface band bending is not a problem as it would be for photoconductivity or field effect measurements.

The results show an enhancement in the gap-state absorption after illumination which disappears upon annealing. We present evidence that this enhancement is due to an increase in the Si dangling bond defect density after illumination [15]. A surprising result is that the number of light-induced defects increases with increased doping concentration, but is constant for undoped films over a wide range of pre-existing defect densities. Another unexpected result is that fully compensated material exhibits the smallest increase in the number of light-induced defects. We discuss the implications of these results in section *IV.*

II. Experimental considerations

The a-Si:H films were undoped, singly doped, and compensated materials, and were deposited by glow discharge under a wide range of deposition conditions [16]. For the undoped samples the rf deposition power varied from 1 watt to 40 watts with a constant deposition temperature of 230 °C. Two samples were examined that had substrate temperatures of 100 °C , and 330 °C. Two others, had oxygen intentionally introduced during deposition [17]. The singly-doped samples had boron or phosphorus concentrations that ranged from 10^{-6} to 10^{-2}

in the gas phase, and the compensated sample had 10^{-3} phosphorus with the boron concentration ranging from 0 to 4 x 10^{-3}. The illumination-anneal cycle consisted of exposing the a-Si:H films to ~ 1.0 W/cm^2 of unfiltered light from a quartz tungsten halogen lamp for typically 1.5 hours. Annealing was achieved by heating the samples to ≥ 150 °C for 1.5 hours in the dark under vacuum. The absorption measurement itself had no detectable effect on either the annealed or illuminated state.

III. Results

Figure 1 shows a typical absorption spectrum where the main features are the Urbach edge and the gap-state absorption tail for both the annealed and illuminated states. It is clear that exposure to light enhances the gap-state absorption, and annealing restores the absorption to its original annealed-state value. The enhancement is consistently reproducible when the illumination-anneal cycle is repeated several times. Further, if only a partial anneal is performed, at lower temperatures or for shorter times, the absorption tail lies between the two curves. Error bars are not shown on the curves since they are on the order of only a few percent. Previous work has demonstrated that the magnitude of gap-state absorption in a-Si:H provides a direct measure of Si dangling bond defect density, N_s [14]. Thus we can quantitatively determine the *change* in defect density between the annealed and illuminated states, ΔN_s. For the undoped material this change in defect density is approximately 1 x 10^{16} cm^{-3}. ESR measurements of the defect density of a particular film showed a change of 2 ± 1 x 10^{16} cm^{-3}, which is consistent with ESR values obtained by Dersch *et al.* [5], and which agreed with the change deduced from PDS for the same film. The agreement demonstrates that the optical cross section of the light-induced defect is ~ 1 x 10^{-16} cm^2. By comparing the experimental absorption spectra with calculated spectra generated from density-of-states models, the light-induced defect energy level is estimated to be approximately 1.0 - 1.3

eV below the conduction band. Since the energy of the defect and its cross section are identical to those measured for Si dangling bond defects, we conclude that the change in sub-gap absorption is due to Si dangling bond defects.

Figure 2 shows the dependence of light-induced defect density on dopant concentration (in the gas phase). As can be clearly seen, the light-induced defects scale with dopant concentration over a wide range of doping levels. The ratio ΔN_s / N_s is found to be independent of doping level to within a factor of 2. Thus, the effect is largest for high doping, unlike the case of conductivity changes [9]. The fully compensated material exhibited the least enhancement, $\sim 10^{15}$ cm^{-3} defects, which is an order of magnitude smaller than that of the undoped material.

The dependence of ΔN_s on N_s in the annealed state for all the undoped and singly doped films is shown in figure 3. For undoped a-Si:H, the increase in defect density, ΔN_s, is constant within a factor of ~4, independent of deposition parameters, whereas for the doped material, ΔN_s increases with N_s. The two oxygenated samples show an approximately equal increase which is comparable to the undoped a-Si:H. Note that the ratio ΔN_s / N_s for the lowest annealed-state N_s undoped and oxygenated films is about a factor of 3-4 greater than that of the doped material.

In the case of the compensated material, by varying the degree of compensation, it is possible to isolate effects due to Fermi level motion and also due to dopant incorporation. In addition, previous evidence indicates that fully compensated material has a relatively low density of dopant-induced Si dangling bond defects [14] [18] . So we can also examine the change in defect density after illumination for these low defect films. The results are shown in figure 4, and indicate that fully compensated material has the smallest increase in defects after illumination. Any departure from full compensation leads to a larger increase in light-induced defect density. This is also evident in figure 5 where compensated films are compared to the doped films.

A set of undoped and phosphorus-doped samples were used to study the dependence of the effect on wavelength. The samples were illuminated with various lines in the visible from Argon and Krypton ion lasers. After correction for penetration depth the results indicated that there was no wavelength dependence.

We have also investigated the dependence of ΔN_s due to illumination on the thickness of the film for several undoped samples. The results are shown in figure 6 for films ≤ 4 μm. The estimated surface increase in defects resulting from illumination is on the order of 10^{11} defects/cm^2.

IV. Discussion

The results presented above have several implications regarding the nature of the light-induced defects, its energy level, and the possible origin of this defect.

A: Nature of metastable state

The results conclusively show that the increase in defect density is due to the creation of *new* dangling bond defects by illumination and that the effect is not due to simply a shift in the Fermi level towards midgap. First, a lowering of the Fermi level, without any change in the dangling bond defect density, results in less sub-gap absorption rather than the observed increase. This can be seen from the density of states presented in figure 7. The low energy optical transitions, i.e. below about 1.5 eV, are from localized to localized, or from localized to band tail state transitions. In the case of undoped or n-type material, the final states are the conduction band states [19] . Consequently, if the Fermi energy shifts towards the middle of the gap, there will be fewer initial states available for transitions at the lower photon energies, and as a result, the absorption at those photon energies will decrease. Second, for large dopant concentrations ($\gtrsim$0.1 % in the gas phase), the Fermi level moves into the band tails so that even after illumination it is still in a region of smaller Si dangling bond state density.

As a result, the effect (as detected by sub-gap absorption) should decrease with increased doping for photon energies above ~ 0.5 eV, which is the opposite of what is observed.

The results from the compensated films also bear out this conclusion. If Fermi level shifts were the only mechanism, one would expect that as the concentration of boron is increased from zero towards full compensation, ΔN_s would also increase. This is to be expected since the Fermi level at full compensation lies near mid-gap and consequently near the maximum of the dangling-bond state. This is clearly not the case. In addition, for the boron rich material, since fewer dangling bond states are occupied, ΔN_s should drop off. This too is inconsistent with our results. Fermi level motion alone cannot account for the observed changes, thus we conclude that the observed increase in ΔN_s is unambiguously due to the creation of new Si dangling bond defects.

A second important conclusion is that the Stabler-Wronski effect is primarily a bulk effect, in agreement with other studies [4] [20]. The light-induced defects essentially scale with film thickness for the films $\leq 4\ \mu m$. Any residual surface component in the undoped films may be related to the oxide layer or band bending. It should be noted that electrical measurements are inherently more sensitive to surface effects such as band bending, which would tend to emphasize the surface properties over those of the bulk.

Finally, preliminary results indicate that the creation of metastable defects by illumination is an intrinsic property of the material. This will be discussed below in section *C*.

B: Light-induced changes in the density of states and comparison with related measurements

A significant advantage of measuring gap-state absorption is that it yields the energy level of the light-induced defect state. From the measured spectra, we determine that this state lies around 1.0 - 1.3 eV below the conduction band

for the undoped material. This can be seen from figure 1 where the largest difference between the spectra lies in this energy range. Previous modeling of the absorption [21] has shown that for undoped material the annealed-state defect peak also lies approximately 1.0 - 1.3 eV from the conduction band. In phosphorus doped material this peak is higher in energy, at about 0.8 - 0.9 eV below the conduction band, because of correlation effects [22]. These annealed-state defect peaks are shown in the approximate density of states (fig. 7) as derived from DLTS [23], dispersive transport [24], and PDS [19]. The PDS measurements also determined that the defect peak near mid-gap is due to Si dangling bonds. Consequently, we conclude that the observed enhancement in the sub-gap absorption is due to an increase in the existing dangling bond defect peaks.

Several other measurements agree with the results from PDS. Previous studies of ESR [5], luminescence [6] [25], spin-dependent photoconductivity [26], and transient photoconductivity [27], implied an increase in the dangling bond density with illumination. The absorption measurements shows explicitly that this is the case.

In contrast, some conductivity measurements [9] indicated a quenching of the Staebler-Wronski effect at the higher doping levels. However, since these measurements were electrical in nature, they are dependent on the position of the Fermi level. Thus for the higher doping levels, even though there may be a change in the defect density, the Fermi level may not shift significantly, and there would be no change in the conductivity.

Recently, Lang *et al.* [11] have used DLTS to study n-type a-Si:H. They conclude that after illumination there is no change in the density of states in the upper half of the gap or in the midgap density of states peak attributed to the dangling bond defect. Instead they interpret their observations, and the increase in dangling bond spin density (as measured by ESR), as due to motion of the Fermi level without an increase in the number of dangling bonds.

From their DLTS data, Lang *et al.* deduce that the predominant effect of illumination is a large increase of about 10^{18} cm^{-3} in donor-like states at 0.5eV above the *valence* band (i.e. about 1.5 eV below the conduction band since for electrical measurements the gap is about 2.0 eV [11]). However, the PDS data is inconsistent with this change. If we calculate changes in the absorption due to a change in the valence band density of states, we find an increase in the absorption near 1.4 - 1.6 eV but not at lower energies. In addition, the magnitude of the absorption increase would be substantially larger than that observed. Instead, calculations with the appropriate increase (1×10^{16} cm^{-3}) in the dangling bond peak produces spectra very similar to those observed. Furthermore, a large change in the density of states 1.5 eV below the conduction band would alter the slope of the Urbach edge since the photon energies that are absorbed by states that deep into the gap would lie higher in energy than the gap-state absorption tail (see fig. 7). However, the optical absorption spectra exhibit no change in the Urbach edge, which in the case of a-Si:H is dominated by the valence-band edge [28]. This result was further confirmed by a series of photoinduced absorption studies which probe the valence band exponential tail [29]. There was no change in the decay rate of the photo-excited carriers through the band-tail states between the illuminated and annealed states. Hence, the optical data are most consistent with the conclusion that the only major change of the density of states for doped and undoped material is an increase in the existing defect peak, and that illumination does not significantly alter the valence band tail [30]. This conclusion is supported by recent drift mobility experiments which found no evidence for light-induced hole traps, indicating that the valence band is not modified by illumination [27].

Lang *et al.* acknowledge that a small increase of 3×10^{16} cm^{-3} in acceptor-like states at 1.0 eV which they cannot resolve, could account for the shift in the Fermi level. Indeed, this increase in states at mid-gap would closely agree with the change in defect density ΔN_s, predicted from the dependence shown in figure

2 for their stated dopant concentration of 3×10^{-5}.

The reason for this discrepancy with DLTS is not clear at the present time. The most likely possibility is that there are difficulties with the interpretation of DLTS measurements in the lower half of the gap. Below mid-gap, the states must be filled with holes using optical filling pulses rather than voltage filling pulses. However, because the optical excitation generates both electrons and holes, it is necessary to apply various assumptions about filling and depleting the traps. For instance, the capture cross section for valence band holes is taken to be equal to that for electrons, the temperature dependence of electron-hole generation and recombination is neglected, and the temperature dependence of the dispersive hole transport out of the depletion region is ignored. Hence, although there is generally excellent agreement between DLTS and optical measurements for the upper half of the gap, the valence band data may disagree.

C: Possible mechanisms

The results described here have several significant implications regarding the mechanisms responsible for the light-induced dangling bonds.

i) Impurity related mechanism. The most notable feature of our results is that ΔN_s increases as the doping level increases. This implies a connection between dopant atoms, or doping-induced effects, and the light-induced defects. Since we show that for singly doped material, the ratio ΔN_s / N_s is roughly constant, independent of film thickness, dopant type, and dopant concentration, this suggests that the light-induced defects are related to defects associated with doping, or impurity-defect complexes where the dopant atoms act as impurities. First we present evidence that the light-induced defects are intrinsic to a-Si:H and are not associated with impurities. Later, in section *iii*, we discuss the connection with dopant atoms and with dopant-induced defects.

Because the light-induced defects correlate with the dopant atom concentration in the a-Si:H matrix, one might expect any non-silicon atom to play a role

in light-induced defect formation. In the case of undoped material, residual impurities, such as oxygen, nitrogen or carbon, could influence the creation of the metastable defect. However, preliminary results on samples where the impurity concentration was characterized by SIMS indicate that this is not the case [17]. Films with oxygen concentrations ranging from 1 x 10^{18} to 5 x 10^{21} cm^{-3} exhibited the same increase of ~ 1 x 10^{16} defects cm^{-3} after illumination. This confirmed earlier work which concluded that for a-Si:H films with relatively small concentrations of oxygen introduced in the gas phase ($\lesssim$ 1000 ppm) there was no dependence of light-induced defect density on oxygen concentration [31]. Films with nitrogen showed similar behavior, and preliminary indications imply this to be the case for carbon. Furthermore the increase in ΔN_s with dopant concentration indicates that in doped material these impurities are not responsible unless there is simultaneously a systematic and marked increase in the uptake of impurities with dopants. Consequently, since for the undoped material with low concentrations of impurities ($\lesssim$ 5 at. %), ΔN_s is independent of impurity content, the effect appears to be intrinsic.

In the case of films with substantially higher concentrations of oxygen ($>$ 5 at. %) there is a significantly larger light-induced effect present. However, it may be more appropriate to consider these oxygen rich films as alloys since the optical gap, the Urbach edge, and the defect density exhibit significant differences from films containing less oxygen [17].

ii) Weak bonds. The striking result that in the case of undoped material ΔN_s is constant over a wide range of annealed-state defect densities has other implications. It has been suggested that the light-induced defects result either from breaking weak Si-Si bonds or weak Si-H bonds. We have shown that while N_s varies by three orders of magnitude, ΔN_s remains fairly constant. One would expect the high N_s material to have more strain as compared to low defect material, thus potentially weakening additional Si-Si bonds. However, the films with high N_s do not show any corresponding increase in ΔN_s, even though the disorder is greater

(fig. 3). Thus, the creation of light-induced defects cannot simply be due to breaking weak Si-Si bonds. If indeed the breaking of these weak bonds is the mechanism involved, one would then have to postulate saturation effects where the defect creation process is self-limiting--i.e. newly created light-induced dangling bonds inhibit defect formation in weak bonds that would otherwise become metastable. As to illumination breaking weak Si-H bonds, this is also unlikely. While the hydrogen concentration is roughly constant for the undoped material, it does not increase dramatically with doping level and consequently ΔN_s should not increase either.

iii) Doping level dependence. The fact that ΔN_s increases with the doping level can be examined in a "chemical" context or a more macroscopic context. Either the light-induced defects are caused by the dopant atoms, or by some change in the properties of the film associated with the presence of dopants. In the latter case, the changes in the annealed state film properties due to increased doping are shifts in the Fermi level, or increased lattice strain. Although the data cannot conclusively determine which mechanism is responsible, it provides additional information and sets some constraints on the possibilities.

In the chemical picture, some of the dopant atoms which are electrically active and in four-fold bonding configurations, relax after illumination to the "chemically preferred" three-fold configuration. NMR studies [32] have shown that approximately 80% of the phosphorus atoms enter the material in a three-fold site. Another study of heavily boron-doped material indicated that as much as 90% of the boron atoms are in three-fold sites [33]. Annealing then causes the atoms to become activated again, i.e. in four-fold configuration, in a manner similar to crystalline material.

Furthermore, if the dopant atoms cause the light-induced defects directly, one might expect that the defect density would increase for the compensated material, independent of the degree of compensation. Figure 4 clearly shows

that the light-induced defect density decreases as the films become compensated. However, there is evidence from photoluminescence [18] and from NMR measurements [32] that boron and phosphorus form complexes during deposition. This is supported by the observation that there is a significant enhancement in the incorporation of the dopant introduced in the lower concentration [18]. (The reduction of the light-induced changes at full compensation is even more drastic when ΔN_s is compared to the amount of boron actually incorporated into the films.) Thus, these B-P complexes could be stable against light-induced reconstruction since the local bonding requirements are satisfied.

The second possibility is that the light-induced defects are caused by some macroscopic change in the film properties, such as Fermi level position or the degree of disorder, due to dopant incorporation. If the dopant incorporation increases the strain within the film and gives rise to more strained bonds, one might expect the light-induced defect density to increase. (For the compensated films, there are fewer defects and consequently fewer strained bonds.) However, as stated above in section *ii*, this mechanism does not allow for the behavior of the undoped films.

A more likely mechanism accounting for the variation in ΔN_s with dopant concentration is Fermi level position. As the Fermi level moves towards the band tails, the creation of dangling bond defects becomes more energetically favorable, enhancing the probability of defect formation. For example, if the Fermi level is near the band edge, as much as ~ 0.9 eV more energy is available for breaking weak Si-Si bonds over the case when the Fermi level is near mid-gap. The origin of defect formation could be from weak Si-Si bonds and/or hydrogen bond switching. As the dopant concentration is increased, the Fermi level rises, enhancing the defect creation probability. For the compensated samples, defect creation is suppressed since the Fermi level is near the center of the gap. The compensated defect creation rate is lower than the rate for undoped films since the Fermi level is closer to mid-gap than in the slightly n-type undoped samples.

Furthermore, there are additional defects in the compensated films which might inhibit defect formation.

The Fermi level mechanism of light-induced defect formation also suggests a connection with the doping-induced defect formation. According to the " 8-N " doping rule [34] in order to obtain four-fold dopant atoms the film must create three-fold Si atoms with a concomitant dangling bond. This process becomes increasingly favorable when the Fermi level is near the band edges. Because illumination of the film raises the quasi-Fermi level, one would expect the light-induced defects to exhibit a similar dependence on doping, which is qualitatively consistent with our results. The data suggests that there is an intimate connection between doping induced and light-induced defects. The same mechanism may operate in both cases.

The preceding discussion presents essentially two different perspectives for viewing the origin of the light-induced dangling bond defects, a chemical (i.e. atomic level) one, and a more macroscopic one based on the film properties. Clearly the detailed nature of the Stabler-Wronski effect requires further examination before it is fully elucidated.

Summary

In summary we have observed an enhancement in the gap-state absorption after illumination, which disappears upon annealing. We attribute this enhancement to the creation of *new* Si dangling bond defects. It appears that the increase in defects is an intrinsic property of the material. In the case of singly doped material, the density of new light-induced dangling bond defects increases with the dopant concentration whereas for compensated films this increase is drastically reduced as full compensation is reached. The results imply that the light-induced defects are not related in any simple way to weak Si-Si bonds or weak Si-H bond as has been suggested.

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Figure Captions

Figure 1: The effect of illumination on gap-state absorption of undoped a-Si:H.

Figure 2: The dependence of the light-induced defect density, ΔN_s, on dopant concentration.

Figure 3: The dependence of the light-induced defect density, ΔN_s, on the annealed-state defect density, N_s, for singly doped and undoped material.

Figure 4: The dependence of the light-induced defect density, ΔN_s, on the ratio of boron to phosphorus introduced in the gas phase.

Figure 5: The dependence of the light-induced defect density, ΔN_s, on the annealed-state defect density, N_s, for all films, showing the deviation for the fully compensated material.

Figure 6: The dependence of ΔN_s cm^{-2} on film thickness.

Figure 7: Density of states for undoped and n-type a-Si:H showing the changes resulting from illumination.

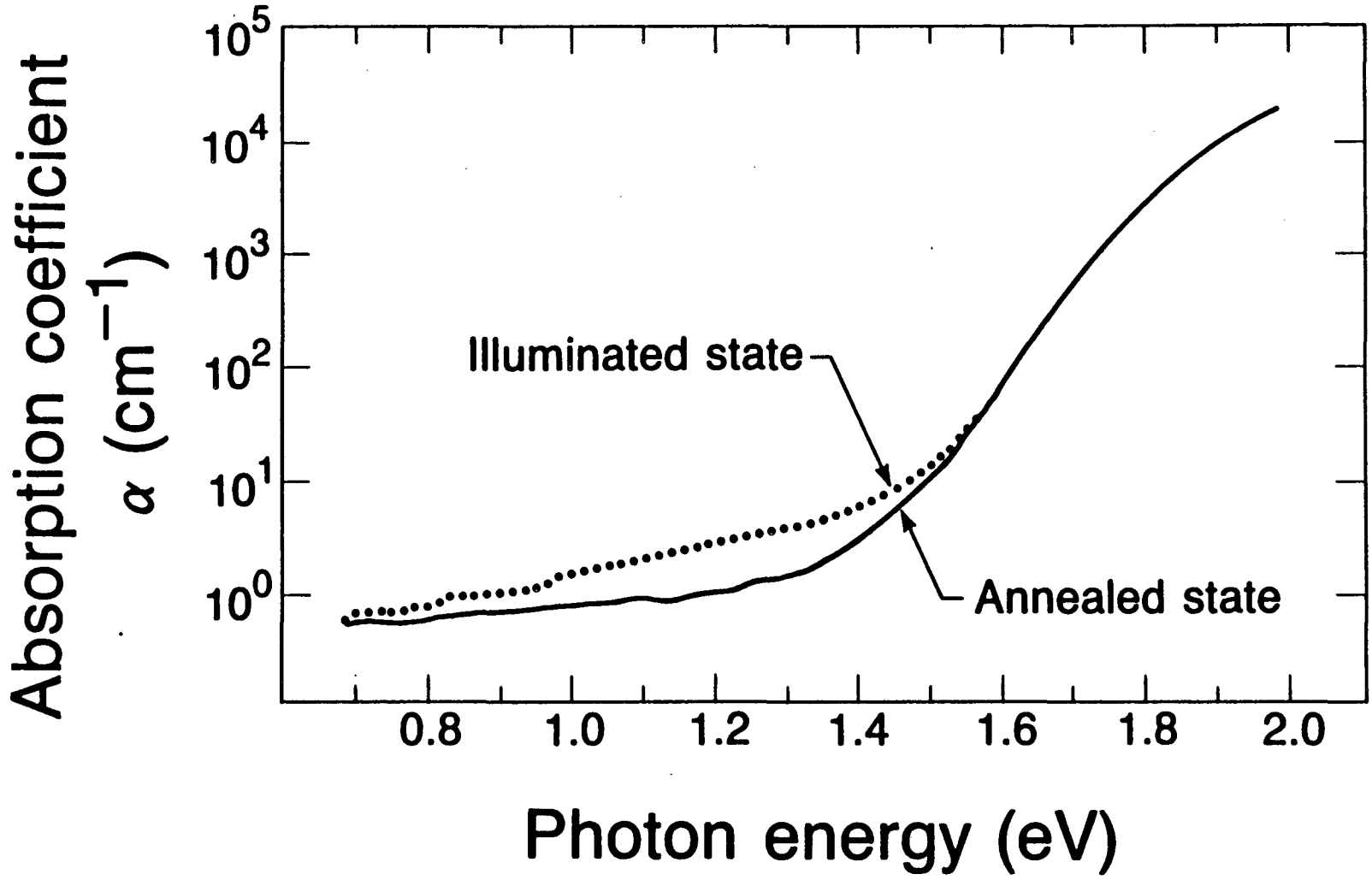


Fig. (1)

XBL 847-10700

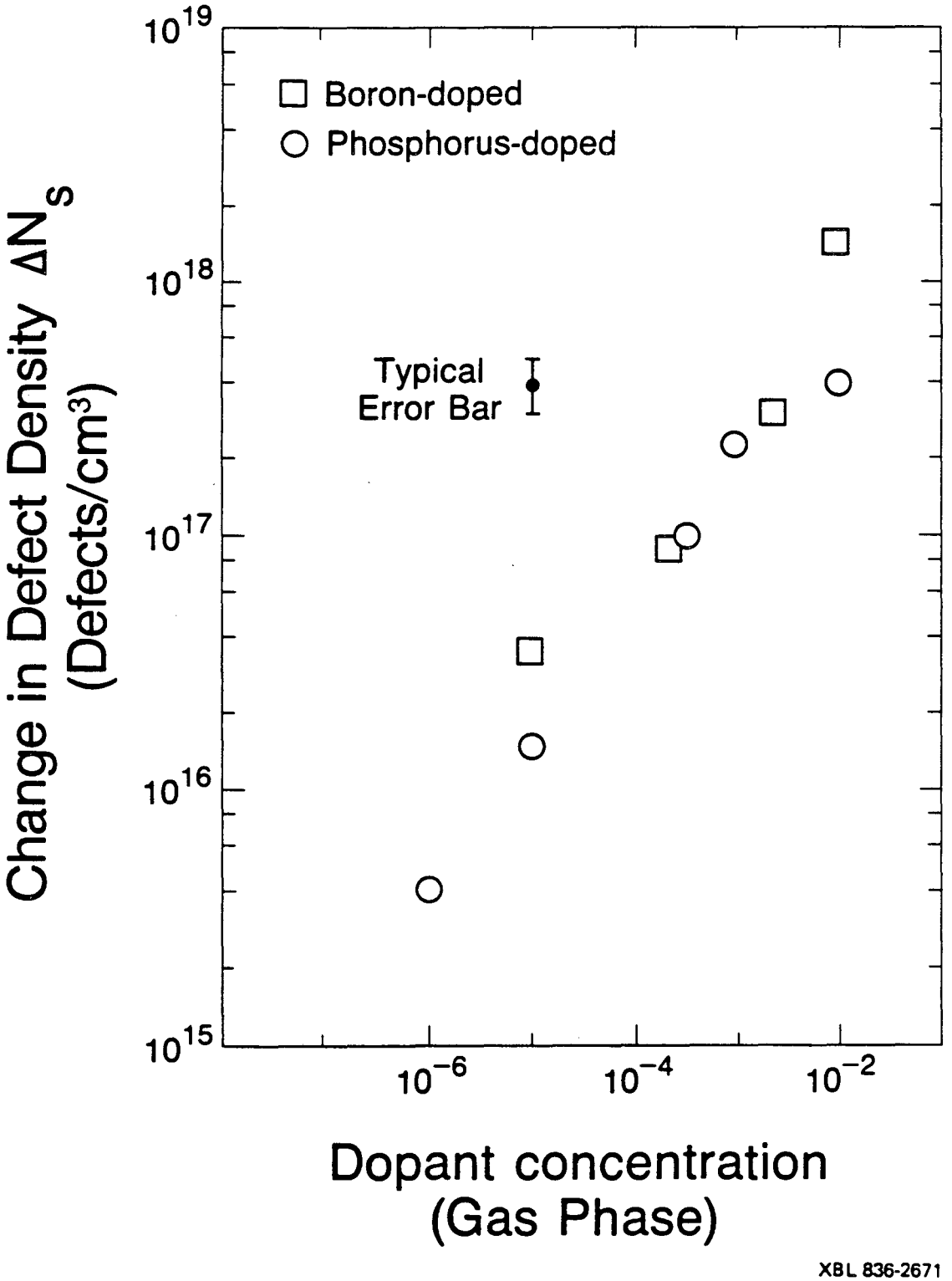
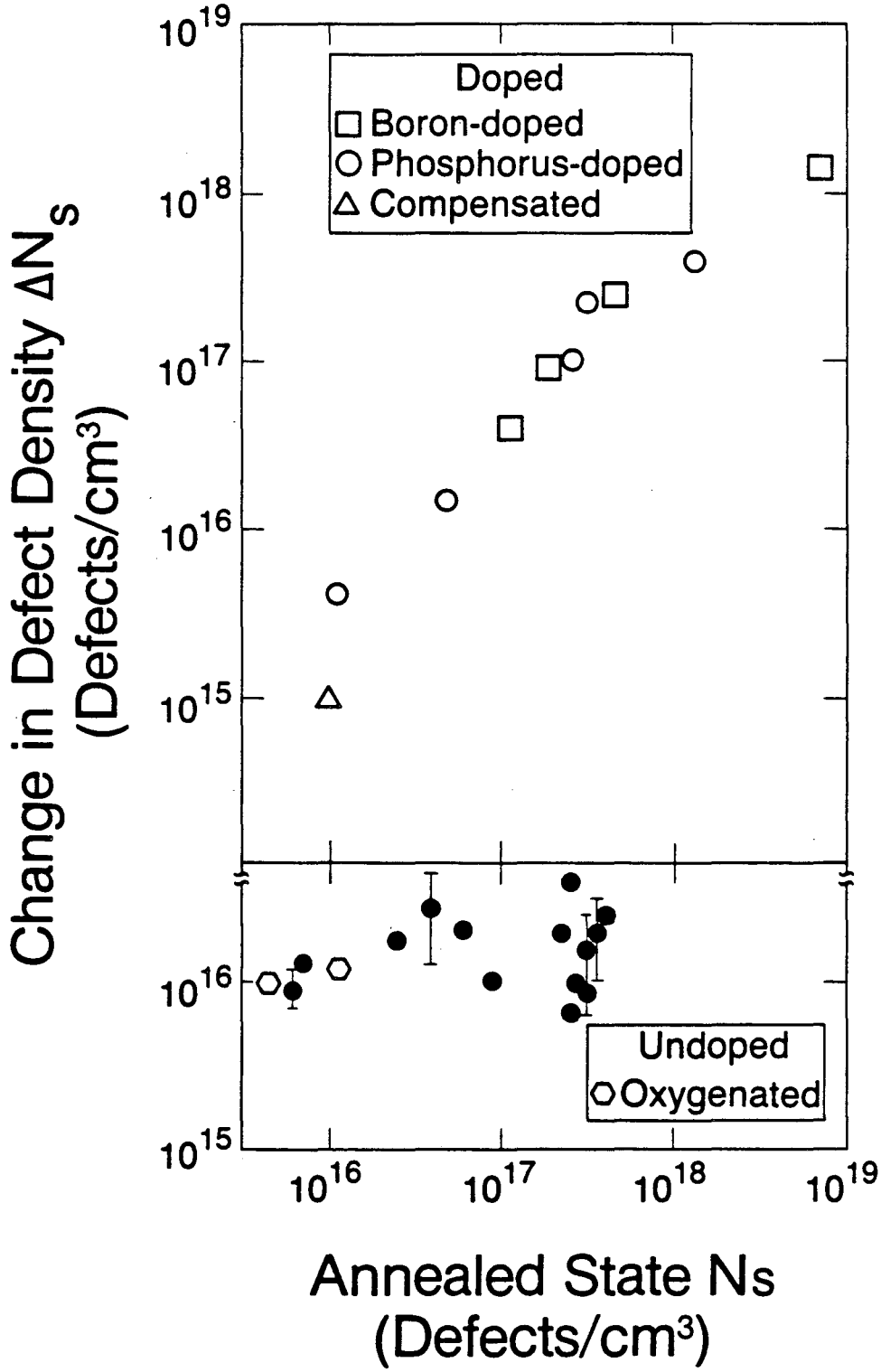

Boron-doped
Phosphorus-doped
Typical
Error Bar
Change in Defect Density ΔN_s
(Defects/cm^3)
10^{19}
10^{18}
10^{17}
10^{16}
10^{15}
10^{-6}
10^{-4}
10^{-2}
Dopant concentration
(Gas Phase)
XBL 836-2671

Fig. (2)



XBL 847-10703

Fig. (3)

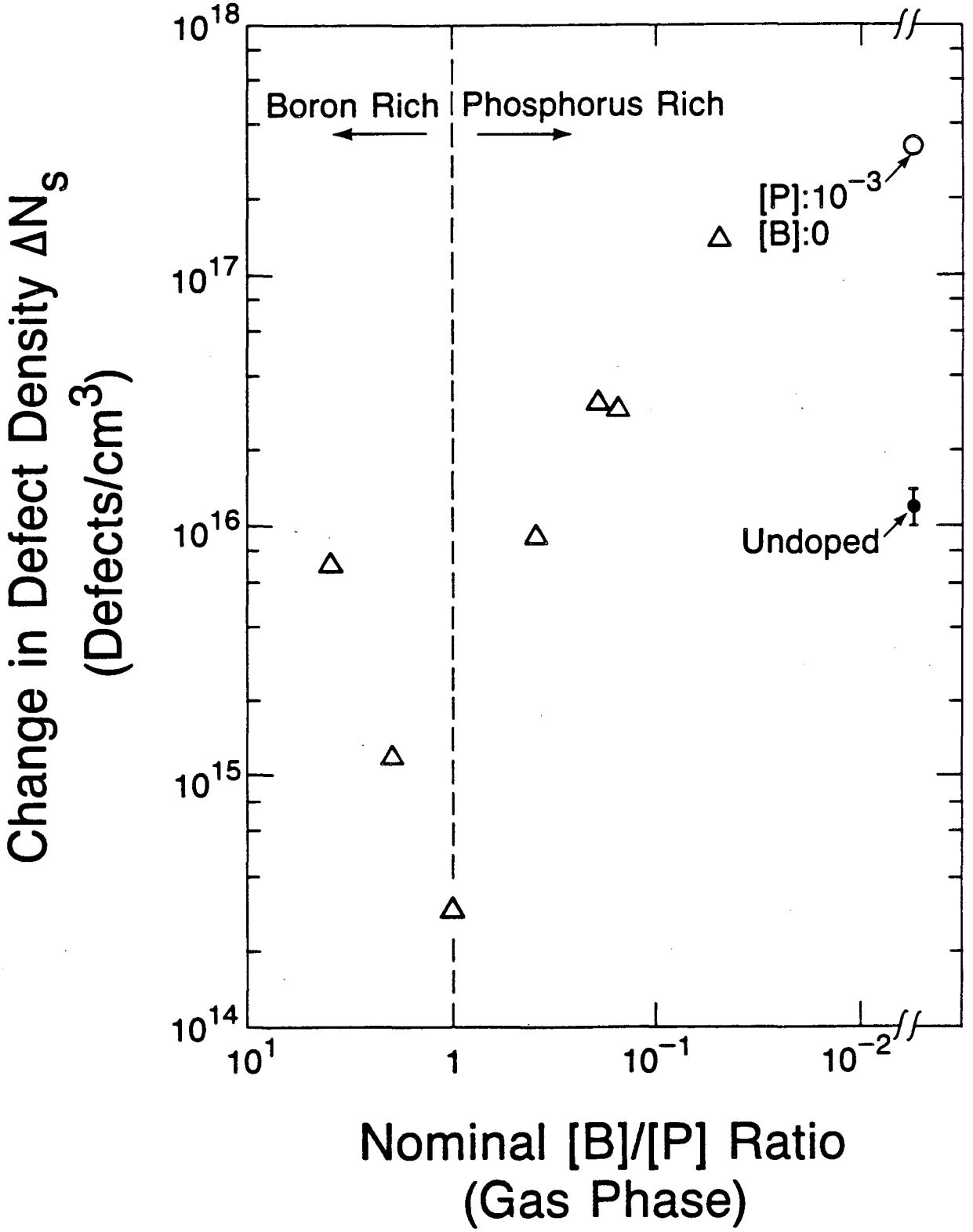
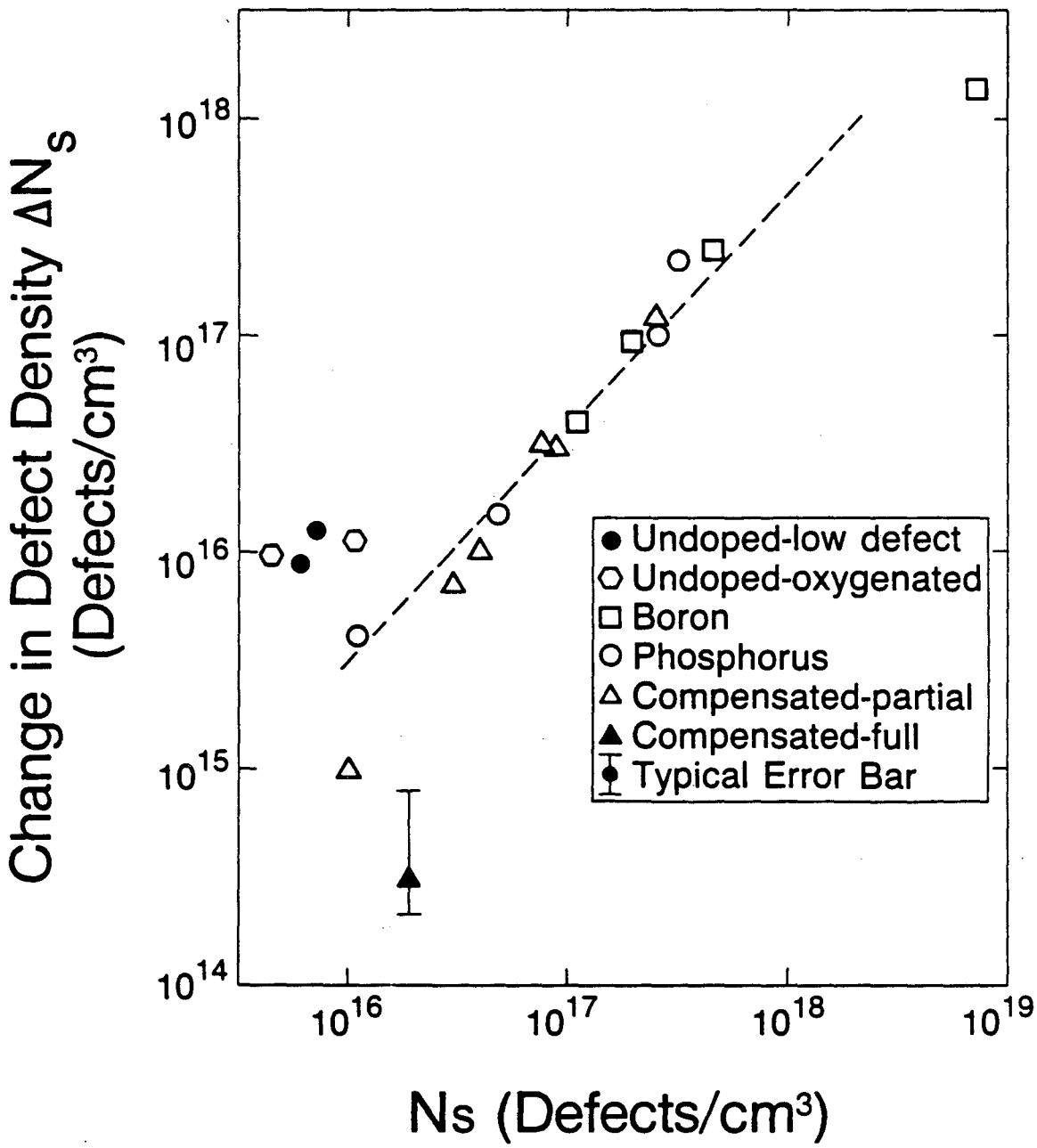

Change in Defect Density ΔN_s
(Defects/cm^3)
10^{18}
10^{17}
10^{16}
10^{15}
10^{14}
Boron Rich
Phosphorus Rich
[P]:10^{-3}
[B]:0
Undoped
10^{1}
1
10^{-1}
10^{-2}
Nominal [B]/[P] Ratio
(Gas Phase)
XBL838-820

Fig. (4)



XBL 838-528

Fig. (5)

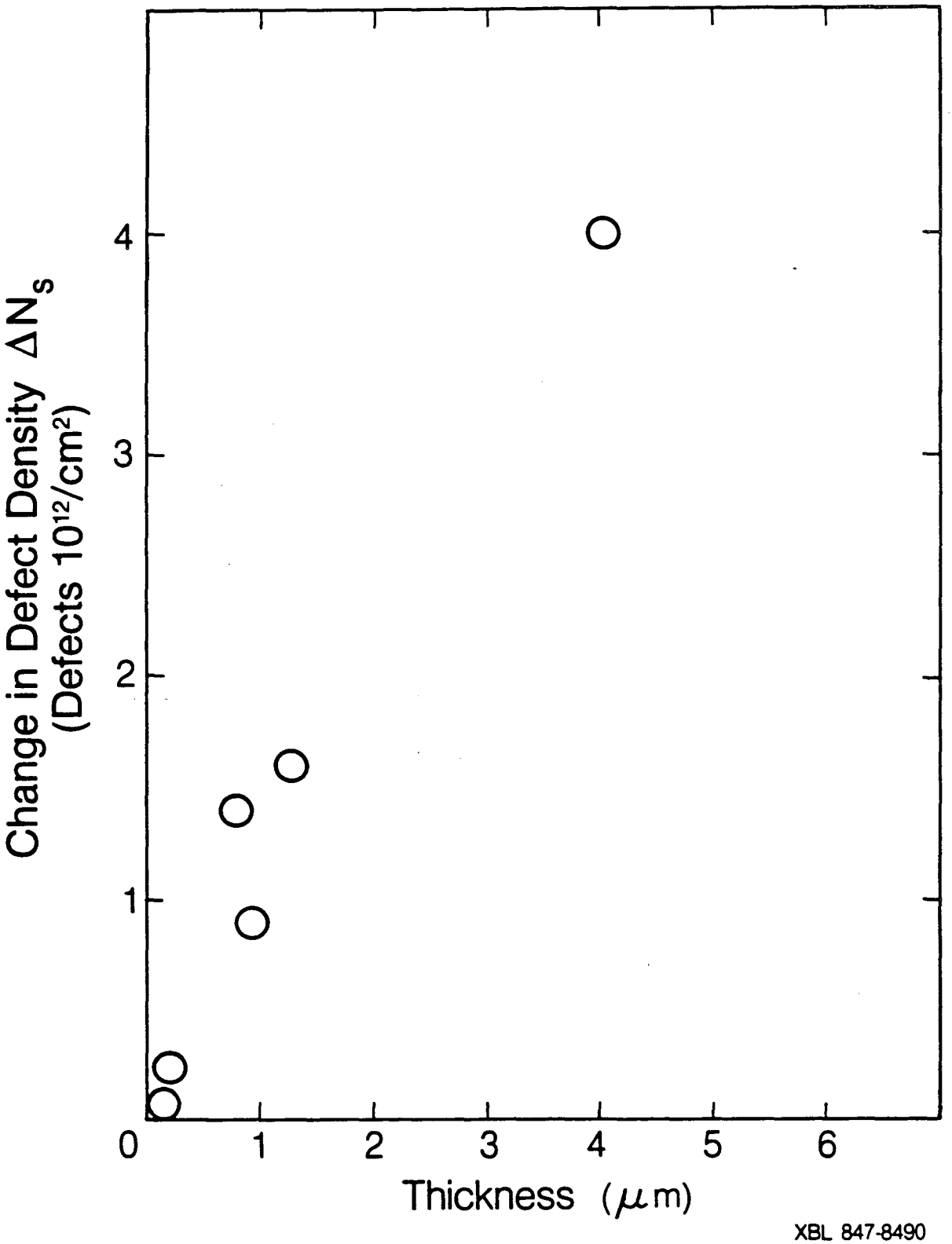

Change in Defect Density ΔN_s
(Defects 10^{12}/cm^2)
0
1
2
3
4
0
1
2
3
4
5
6
Thickness (μm)
XBL 847-8490

Fig. (6)

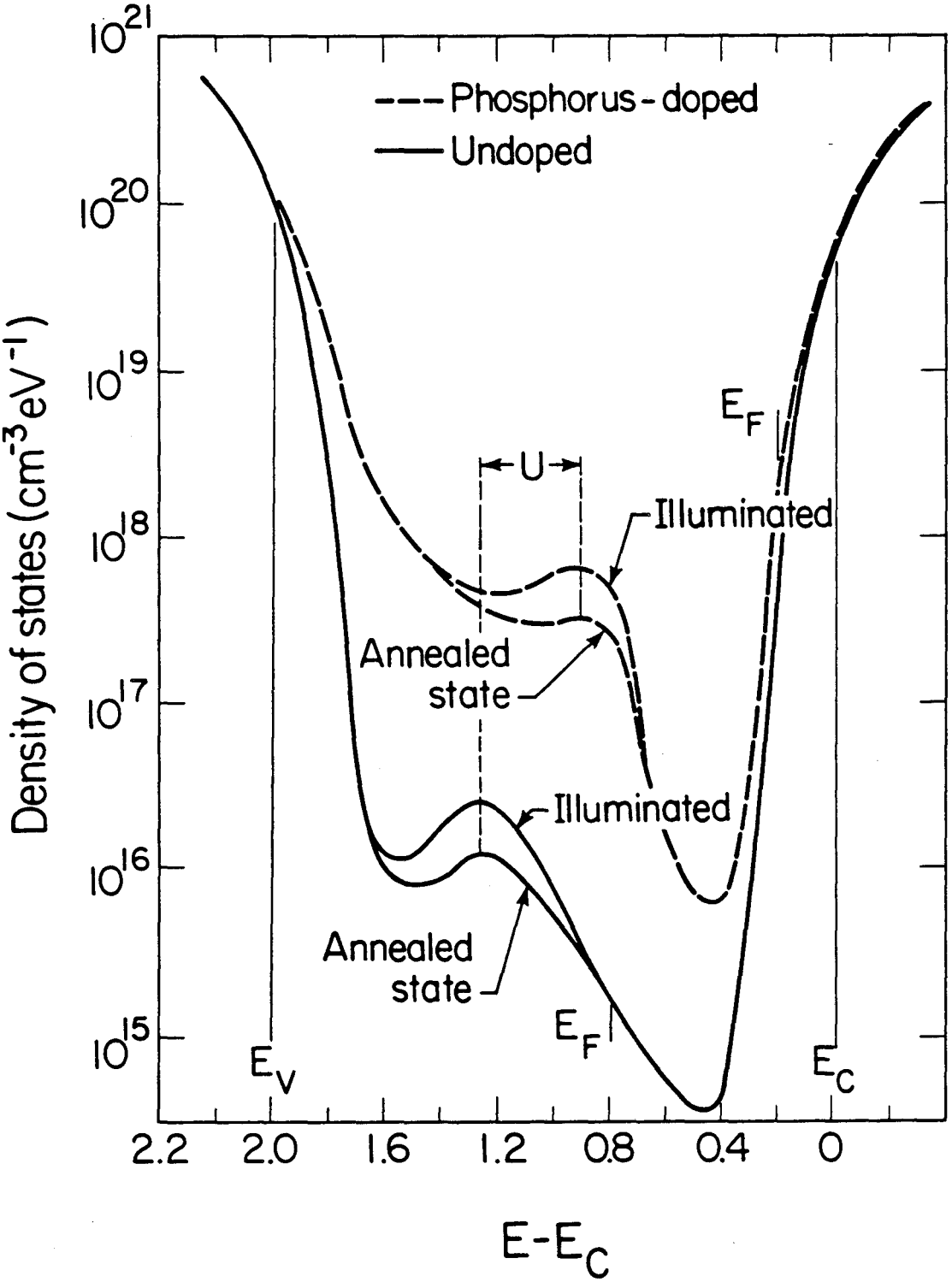

Phosphorus-doped
Undoped
Illuminated
Annealed state
Illuminated
Annealed state
U
E_F
E_F
E_V
E_C
Density of states ($cm^{-3}eV^{-1}$)
10^{21}
10^{20}
10^{19}
10^{18}
10^{17}
10^{16}
10^{15}
2.2
2.0
1.6
1.2
0.8
0.4
0
$E-E_C$
XBL 836-2672

Fig. (7)

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