Unoccupied states in the band gap of δ-doped Si in GaAs probed by Auger resonance spectroscopy

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Auger *KLL* and 1*s* photoelectron spectra of silicon have been studied from samples of Si delta doped layers in GaAs with very thin capping layers. By tuning the exciting photon energy through the Si K edge resonant Auger spectra were obtained. A comparison of the *x*-ray absorption spectra (XAS) in the vicinity of the Si K edges with the energy dependence of the resonant Auger spectra indicates that in each material the resonant process involves a narrow state in the GaAs band gap localized at the Si sites. The energy dispersion of the resonant peak sheds light on the nature of these localized states and XAS data around the Si K edge indicates that they are located in the GaAs band gap. The results yield information on the positions and widths of the localized states.

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I. INTRODUCTION

There is considerable interest in the electronic properties of delta (δ) doped layers in semiconductors because of their importance in the development of electronic devices.[1] However since such layers are usually buried at least 100 angstroms under the surface it is difficult to probe their electronic structure using the techniques of x-ray photoelectron spectroscopy and Auger electron spectroscopy because of the low escape depths of electrons.[2] Consequently previous studies of such systems by electron spectroscopy have been limited to concentrated δ layers of >1 monolayer (ML)[3–6] the electronic structures of which are likely to be very different to those of the dilute systems important in devices.

In this study we report the results of electron spectroscopy measurements on δ layers of 0.3, 0.05, and 0.01 ML of Si in GaAs grown with extremely thin, $\sim$10 ML, capping layers of GaAs. This range of concentrations is around that at which the donor activity is known to saturate, $\sim 10^{13}$ cm^{-2}. We have exploited the fact that since the Si δ layers are buried the condition of the surface of the specimen will not influence the results for the electron spectra of Si and consequently it was possible to achieve good signal to noise from these dilute systems by scanning for very long periods.

Photoemission of the Si 1*s* level indicates that while there is only one Si site in the 0.01 ML δ layer Si occupies two different sites in the more concentrated δ layers. We have observed an Auger resonance accompanying the Si (*KLL*) Auger spectra of these systems when these transitions are excited close to threshold. This resonance is associated with an unoccupied state in the electronic structure of the Si δ layers, which is also observed in x-ray absorption. This unoccupied state is likely to have an important influence on the electron transport properties of Si δ layers and we have determined its position and width for the most dilute specimen.

II. EXPERIMENT

The samples were grown using a modified VG V80H molecular beam epitaxy, (MBE) machine. The As_4 (6.4×10^{14} molecules s^{-1} cm^{-2}) and Ga (1 μm/h) fluxes were calibrated using reflection high energy electron diffraction intensity oscillations. The Si cell temperature was maintained at 1250 °C, equivalent to a flux of 6×10^{11} Si s^{-1}, cm^{-2}, confirmed by secondary ion mass spectroscopy. Substrate temperatures were monitored using an IRCON pyrometer, which was in turn calibrated by using the oxide desorption temperature of 600 °C and the $(2\times4)->c(4\times4)$ change in surface reconstruction which occurs at 520 °C under an As_4 flux of 6.4×10^{14} molecules s^{-1} cm^{-2}.

Epiready n^+ GaAs wafers were mounted onto Mo blocks using In solder before loading into the MBE machine via a load lock. After outgassing at 300 °C for 1 h the substrate was transferred to the growth chamber where the oxide was removed by heating to 600 °C under an As_4 flux. After the growth of a 1000 Å GaAs buffer layer at 580 °C the substrate was cooled to 400 °C. After growth of a further 100 Å of GaAs, the Ga was turned off and Si deposited. The As_4 flux was maintained during the Si deposition, which took between 10 s for the 0.01 ML specimen and 300 s for the 0.3 ML specimen. Samples with 0.3, 0.05, and 0.01 ML Si layers were used in this study. The Si layer was subsequently overgrown with 10 ML of GaAs. The substrate was then cooled to room temperature and capped with As. It has been shown that in general diffusion of Si out of the δ layer at these growth temperatures is negligible[7] though this has not been confirmed for the specimens used in this work.

The experiments were carried out on beamline 4.2 on the Synchrotron Radiation Source at the Daresbury Laboratory (Cheshire, United Kingdom). The beamline is equipped with a double crystal monochromator using InSb crystals, which give a resolution of 1–2 eV at photon energies in the region of 1800–2000 eV. A separate I_0 chamber containing a copper mesh recorded the beam intensity by measuring the drain

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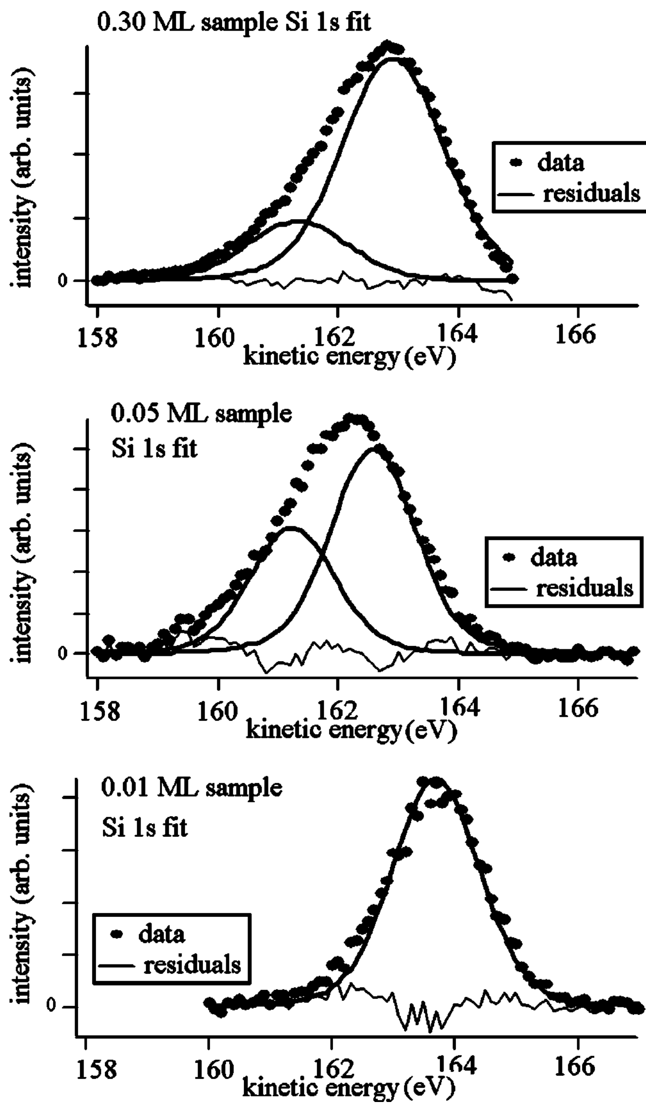

FIG. 1. Photoelectron spectra of the silicon 1*s* level shown with fitted components.

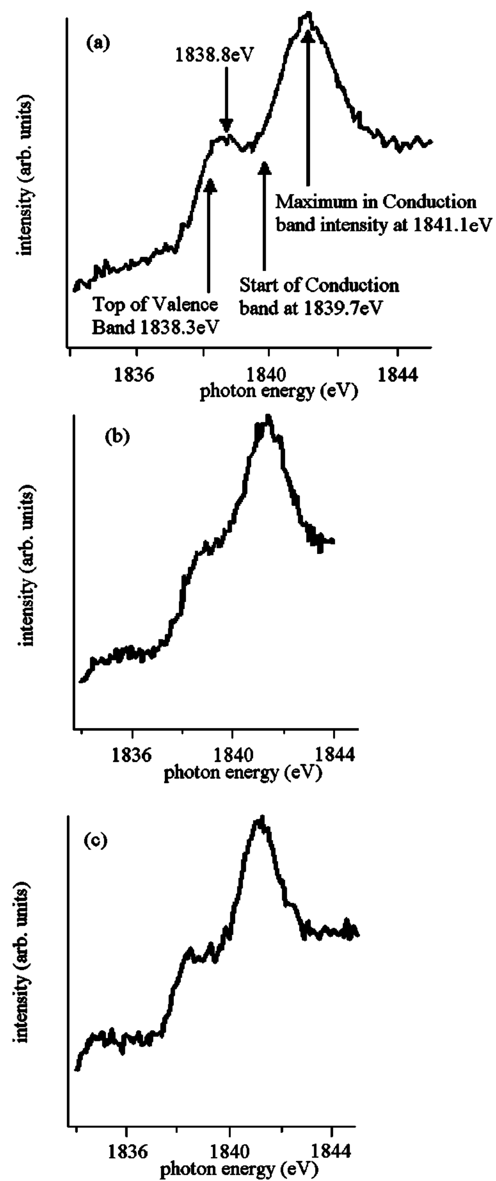

FIG. 2. X-ray absorption spectrum taken over the K absorption edge of Si from (a) the 0.01 ML delta layer sample, (b) the 0.05 ML delta layer sample, and (c) the 0.30 ML delta layer sample.

current on the mesh. All the spectra presented here have been normalized to the I_0 reading. The end-station chamber was equipped with a 150 mm radius Vacuum Science Workshop (VSW) hemispherical analyzer that was mounted at 90° to the incident light in the plane of the beam and collected spectra with a take off angle of 45° with respect to the sample surface. The contribution to the energy resolution of electron spectra arising from the analyzer when operated in the "fixed analyzer transmission" mode with a pass energy of 90 eV was 1.2 eV. x-ray absorption spectra were measured by means of a partial yield detector mounted at 45° to the incident radiation and out of the plane of the synchrotron.

III. RESULTS

The photoelectron spectra of the Si 1*s* level measured from the three specimens using 2000 eV exciting radiation are shown in Fig. 1. The spectra obtained from the 0.05 and 0.3 ML layers were obtained with a total Gaussian instrumental resolution derived from the monochromator and analyzer of full width half maximum (FWHM) of 1.47 eV whereas that measured from the weaker 0.01 ML specimen was obtained with a lower resolution of FWHM 1.79 eV. In spite of the higher resolution the spectra obtained from the more concentrated systems are significantly broader than that observed from the 0.01 ML layer. The lifetime broadening of the Si 1*s* level is known to be 0.38 eV.[8] A convolution of a Lorentzian line of FWHM 0.38 eV with a Gaussian of FWHM of 1.79 eV gives a good fit to the Si 1*s* photoelectron spectrum observed from the 0.01 ML specimen (Fig. 1). It is not possible to fit the Si 1*s* spectra obtained from the more concentrated specimens to a single component with the expected lifetime broadening. However by allowing the fitting routine to use two components an acceptable fit can be obtained. Figure 1 shows the results obtained by fitting these

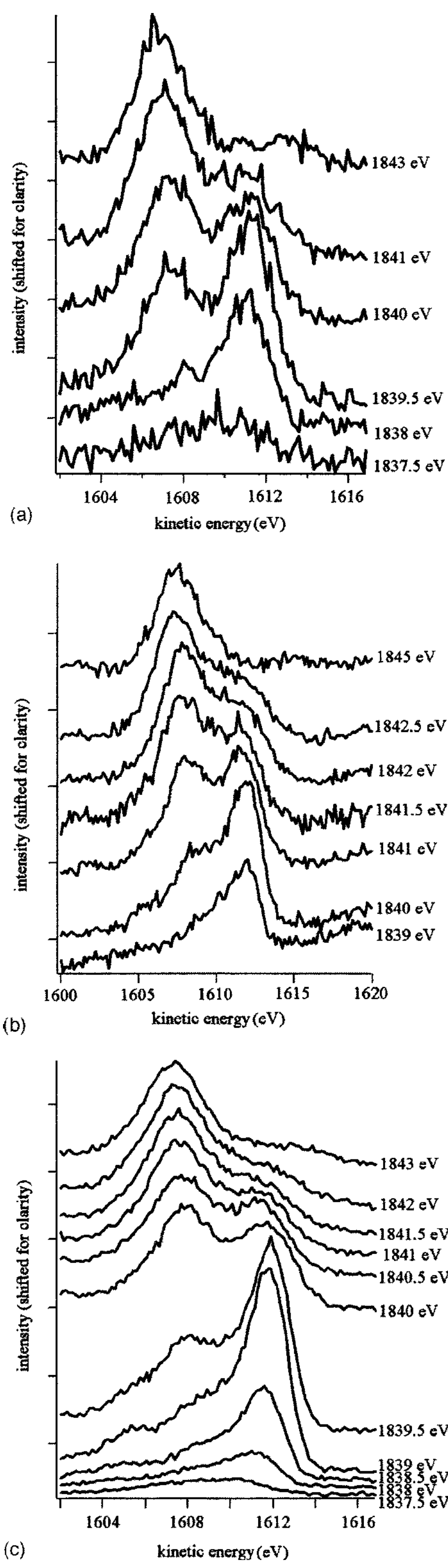

FIG. 3. (a). Si KLL Auger spectra from the 0.01 ML delta layer sample as hν is tuned through the Si K threshold. The energy of the exciting radiation is shown next to each spectrum. An extra peak can be seen at a kinetic energy of 1611 eV that only exists between 1838$<$h$\nu$$<$1841. (b) The Si KLL Auger spectra from the 0.05 ML sample as the photon energy is tuned through the K edge. (c) The Si KLL Auger spectra from the 0.30 ML sample as the photon energy is tuned through the K edge.

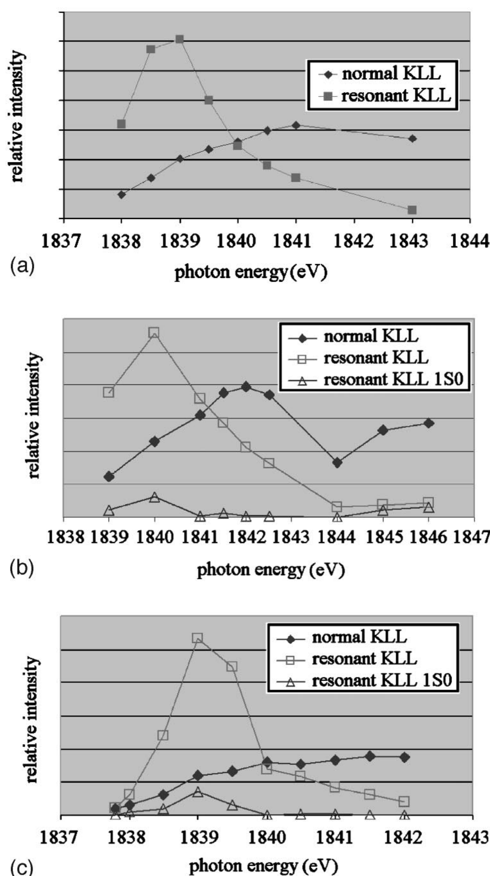

FIG. 4. (a). Intensity variations of the Si Auger KLL components with photon energy in the 0.01 ML Si delta layer sample. (b) Intensity variations of the Si Auger KLL components with photon energy in the 0.05 ML Si delta layer sample. (c) Intensity variations of the Si Auger KLL components with photon energy in the 0.3 ML Si delta layer sample.

spectra to two components with the appropriate lifetime and instrumental broadenings.

The normalized x-ray absorption spectra (XAS) of the Si thresholds of these materials are shown in Fig. 2. Each spectrum shows two distinct features in the absorption spectrum; a pre-edge to the absorption which occurs at an excitation energy of about 1838.8 eV and a second peak at about 1841 eV. The energy of the second peak corresponds to the binding energy of the Si 1s level and is the threshold at which the Si 1s electrons receive enough energy to be ejected into the continuum. Previous XAS data of pure silicon show only this second peak at ~1841 eV.[9] All three samples in this study show the pre-edge feature but for the more concentrated delta layers [Figs. 2(b) and 2(c)] the pre-edge peak is broader than for the 0.01 ML sample [Fig. 2(a)].

When excited by photons far above threshold the Si $KL_{2,3}L_{2,3}$ Auger spectra of all three materials showed the characteristic multiplet structure usually observed for these transitions. This consists of a strong component arising from the $^1D_2$ term of the $2p^4$ final state which was observed at a kinetic energy of ~1606 eV and a weaker component which arises from the $^1S_0$ term and occurs 6 eV lower in energy and has ~8% of the intensity of the $^1D_2$ term. However when

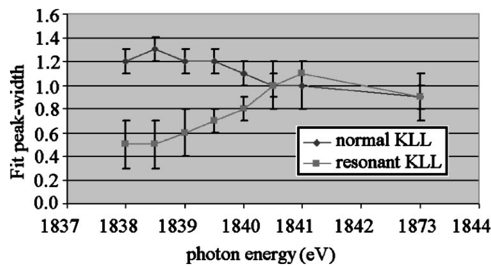

FIG. 5. Peak-width variations of the Si Auger KLL components with photon energy in the 0.01 ML Si delta layer sample.

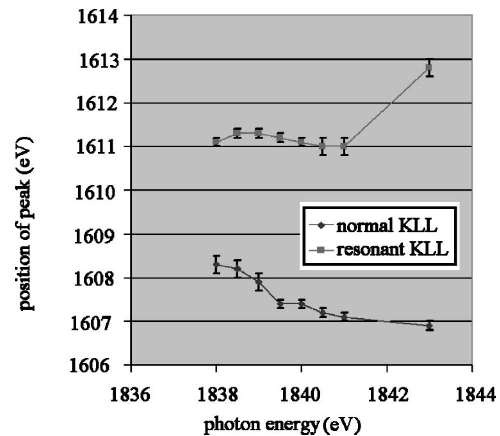

FIG. 6. Fit results for the positions of the normal and resonant Auger peaks in the 0.01 ML sample. Note that the resonant Auger peak shows no dispersion with photon energy except when excited by the highest energy photons.

excited by photons close to threshold the Si $KL_{2,3}L_{2,3}$ Auger spectra do not show this characteristic multiplet structure and vary markedly with photon energy as shown in Figs. 3(a)–3(c) for the three samples. Each spectrum consists of two main peaks and of these the one to lower kinetic energy evolves into the $^1D_2$ term observed at a constant kinetic energy of ~1606 eV when excited by photons far above threshold. We associate the peak to higher kinetic energy with a resonant Auger process as it is only observed when excited by radiation close to the K threshold and it exhibits a narrower width than the $^1D_2$ term. We also note that the maximum intensity of this feature observed for each specimen occurs when the spectrum is excited by photons corresponding to the maximum of the first peak in the x-ray absorption spectrum [Figs. 2(a)–2(c)].

In order to characterize the behavior of the resonant Auger process in more detail the Si $KL_{2,3}L_{2,3}$ Auger spectra were fitted to two Lorentzian line shapes of variable widths and intensity convoluted with the Gaussian contribution of FWHM 1.2 eV arising from the analyzer. The results obtained for the variation with photon energy in the intensities of the peaks are shown in Figs. 4(a)–4(c) for the three specimens. The results obtained for the variation with photon energy of the peak width and kinetic energies of the resonant feature observed from the 0.01 ML specimen are shown in Figs. 5 and 6, respectively. The peak positions and widths observed for the Si $KL_{2,3}L_{2,3}$ Auger spectra of the 0.05 ML and the 0.30 ML samples show similar behavior to those observed for the 0.01 ML specimen.

IV. DISCUSSION

The evidence from the 1*s* photoelectron spectra that Si occupies a single site for the 0.01 ML δ layer but that two sites are present in the more concentrated layers is consistent with previous work in which at low concentrations Si is thought to occupy Ga sites (Si_{Ga}) but in more concentrated δ layers both Si_{Ga} sites and Si_{Ga}–Si_{As} paired sites are present.[10,11] In the following discussion we restrict the detailed analysis to the spectra obtained for the 0.01 ML specimen, which is more relevant to technological applications. This avoids the need to consider the presence of two sites in the more concentrated and less technologically important specimens.

It is useful to compare the x-ray absorption spectra of Si (Fig. 2) in these materials with the electronic structure of GaAs (Ref. 12) We can produce a consistent explanation of the x-ray absorption and Auger spectra if we assume that the pre-edge peak in the x-ray absorption spectrum, which has a maximum at 1838.8 eV in the spectrum observed from the 0.01 ML specimen, arises from a localized Si state at the Si δ layer and does not involve the electronic structure of the GaAs. The observation that the maximum intensity of the resonant Auger peak observed from all three specimens occurs at a photon energy corresponding to the peak energy of this pre-edge feature is strong evidence that this feature arises from the excitation of an electron from the Si 1*s* core level into a localized state associated with the δ layer. The resonant Auger process requires a long-lived localized excited state and these are the expected characteristics of a state localized at a δ layer in the band gap of a semiconductor. In previous studies on Silicon and SiO_2 (Refs. 9 and 13–16) the XAS data shows no pre-edge to the main Si K edge absorption. However Yubero *et al.* did observe a similar feature in spectra obtained from complex SiO_x specimens, which were composed of a spread of different local Si environments and from which resonant Auger signals were observed.[17]

We can locate the position of the resonant state in the GaAs band gap for the 0.01 ML specimen by considering the results of bremsstrahlung isochromat spectroscopy (BIS) of GaAs (Ref. 12) which show that the maximum intensity of the conduction band of GaAs occurs at ~1.4 eV above the bottom of the conduction band. It is reasonable to associate the maximum intensity of the Si x-ray absorption edge with the maximum of the GaAs conduction band density of states as indicated in Fig. 2(a). When this is done the BIS data imply that for the 0.01 ML specimen a Si 1*s* core electron will be excited to the bottom of the GaAs conduction band by x rays of energy ~1839.7 eV. Such an electron would be expected to delocalize from the excitation site rapidly leaving the core-ionized Si to decay by a normal *KLL* process. This is in good agreement with the photon energy dependence of the Si Auger spectra shown in Fig. 3 since the threshold for excitation of the normal $KL_{2,3}L_{2,3}$ $^1D_2$ component lies around 1839.5 eV and by 1840.0 eV this component is more intense than the resonant Auger line. Having

established that the bottom of the GaAs conduction band corresponds to a photon energy of 1839.7 eV [shown in Fig. 2(a)], the known band gap of GaAs, 1.4 eV, allows us to associate the top of the valence band with a photon energy of 1838.3 eV. The maximum intensity of the Si localized state is seen at 1838.8 eV, which places it at 0.5 ± 0.2 eV above the top of the GaAs valence band in the band gap.

The results obtained from the fits to the resonant Auger spectra obtained from the 0.01 ML specimen provide further insight into the nature of the localized state. The pre-edge in the x-ray absorption spectrum [Fig 2(a)] occurs at ~1838.8 eV and this is the photon energy which gives rise to the maximum intensity of the resonant Auger line [Fig 4(a)]. The resonant peak is intense when excited by radiation in the range 1838 to 1840 eV suggesting that the resonant state has a width of <2 eV. Since the resolution of the monochromator is between 1 and 2 eV at these energies this implies that the width of the resonant state is quite narrow, possibly less than 1 eV.

A comparison of the photon energy at which the resonant Auger peak has a maximum intensity [Fig 4(a)] with the binding energy of the Si 1*s* level relative to the valence band maximum of GaAs confirms that the resonant state is localized just above the maximum of the GaAs valence band.

We expect that the localized state will be broadened by the effects of interactions between the Si sites in the more concentrated specimens and this view is supported by the increased width of the pre-edge feature in the x-ray absorption of the 0.05 and 0.30 ML specimens [Figs. 2(b) and 2(c)]. However the presence of two Si sites in these specimens makes it difficult to draw any firm conclusions from a more detailed analysis.

The resonant Auger process is analogous to the resonant Raman effect.[18–20] It has been observed in noble gasses,[21–28] metals,[29–39] semiconductors,[9,13–15] and many other systems.[16,17,40–56] Resonant Auger peaks are usually narrower than the normal Auger line[14,23,31] and this characteristic is exhibited by the results of our experiments (Fig. 5). We suggest that in our experiments the resonant Auger process is caused by the excitation, at threshold, of a Si 1*s* core electron into a state in the GaAs band gap that is localized on the Si site. This spectator electron will occupy the localized state during the Auger decay of the 1*s* core hole causing a shift on the Auger kinetic energy. The resonant Auger electron will have a higher kinetic energy than the normal Auger electron due to the Coulomb repulsion between the spectator electron and the Auger electron. We observe this behavior in our data (Fig. 6). The maximum difference between the normal and resonant Auger electrons observed from the 0.01 ML specimen is ~3 to 4 eV. This is significantly smaller than the value of the appropriate atomic Coulomb integrals, Fo (2*p*3*p*) and Fo (2*s*3*p*), that atomic structure calculations give as ~12 eV.[57] The difference is probably due to the variations in the polarization of the local environment in the different initial states and by differences in the final state relaxation energy for these processes, all of which are very difficult to estimate.

In the earliest study of Si systems Baba *et al.*[15,16] observed a strong resonant Auger effect in SiO_2 but no effect in pure Si or SiC. The resonant Auger effect in SiO_2 correlated with the observation of a narrow band above the Fermi energy in the x-ray absorption spectrum of the Si *K* edge and the resonant Auger peak showed a linear increase in kinetic energy as the energy of the exciting radiation increased above threshold. This "positive dispersion" of the resonant Auger line is generally observed and is expected theoretically. It has been observed in several other systems.[21,22,33–35,45,46] In SiO_2 it is attributed to the energy released when the electron excited from the Si 1 *s* level into the band above the Fermi energy falls back to the bottom of the band, which is several electron volts wide, during the resonant Auger emission. This energy is transferred to the resonant Auger electron and the process is known as participator decay. The failure to observe a resonant Auger process in Si and SiC was attributed by Baba *et al.*[15] to the greater width and less localized character of the conduction band in these systems.

Yubero *et al.*[17] have observed a resonant Auger process in thin SiOx films and in these systems the resonant Auger peak shows a negative dispersion with increasing photon energy. This is puzzling behavior which is not explained theoretically and which Yubero *et al.*[17] suggest might be due to the post collision interaction (PCI) mechanism or to differences in the extra-atomic relaxation around single and double ionized states in these systems. However the spectra are difficult to interpret due to the composite nature of the specimens, which are formed with a variety of local Si environments.

The kinetic energies of the two contributions to the Si Auger spectrum of the 0.01 ML specimen obtained from the fit are shown in Fig. 6. As in all three specimens excitation with photons at the threshold gives rise to a normal component with a kinetic energy around 1606–1608 eV and this kinetic energy decreases smoothly with increasing photon energy. It is possible that this effect could be caused by the PCI mechanism[22,23] since at the threshold the initial photoelectron will be moving slowly and can transfer energy to the Auger electron. However it is not clear that the PCI mechanism can explain the data shown in Fig. 6 and a calculation of the effect of the PCI mechanism would be difficult for a Si atom in a delta layer in GaAs. Within the accuracy of the fitting procedure the resonant Auger line shows a constant kinetic energy over the 2 eV photon excitation range for which it is intense [Fig. 4(a)]. This is in contrast to most other resonant Auger processes, which show a positive dispersion with increasing photon energy.[15,16,21,22,33–35,45,46] A likely explanation for this behavior is that in the δ layer the unoccupied state is very narrow and shows no dispersion in energy. With these characteristics it can only be excited by a narrow range of photon energies, a range that may be narrower than the 1–2 eV resolution of the double crystal monochromator. At higher photon energies it is possible that the localized state can be occupied by electrons, which are first excited into the GaAs conduction band and which then transfer their energy to the resonant Auger electron when dropping into the localized state. While this would provide a mechanism to increase the energy of the resonant Auger electron, as exhibited in the spectrum excited by the highest energy photons [Figs. 3(a) and 6], there will be little mixing between the localized state in the Si δ layer and the GaAs conduction band and the transition will be weak, as observed.

V. CONCLUSION

We have studied the electronic structure of Si delta doped layers in GaAs of concentrations 0.01, 0.05, and 0.30 ML. Si occupies a single site in the 0.01 ML specimen and two sites in the more concentrated δ layers. We have observed an unoccupied state in the GaAs band gap in all three materials. The observation of a Si resonant Auger spectrum establishes that the unoccupied state is located on the Si layers and the lack of dispersion of the resonant Auger spectrum indicates that it is narrow, possibly less than 1 eV in width.

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