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Photo-induced electron trapping in indirect bandgap $Al_xGa_{1-x}As$:Si at low temperature

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Abstract. A variation of photoconductivity excitation with wavelength is applied to Si-doped $Al_{0.56}Ga_{0.44}As$ (indirect bandgap material) for a wide range of temperature. The lower the temperature the lower the photocurrent below 70 K. In the range 13–30 K there is a decrease in the photoconductivity spectrum slightly above the bandgap transition energy, followed by another increase in the conductivity. We interpret these results in the light of existing models and confirm the trapping by the X-valley effective mass state, which is responsible for attenuation of persistent photoconductivity below 70 K. A DX^0 intermediate state which has non-negligible lifetime is proposed as responsible for the decrease in the photoconductivity with about 561 nm of wavelength of exciting light, in the investigated 13–30 K range.

1. Introduction

In the last twenty years many efforts have been concentrated towards the understanding of $Al_xGa_{1-x}As$ physical properties, since this alloy has wide application in opto-electronic devices. A deep level known as the DX centre is dominant for the electrical properties of this alloy and most of its physics has already been established [1]. A lot of good work was published on the physics of this deep level and its influence on the performance of opto-electronic devices, mainly on direct bandgap $Al_xGa_{1-x}As$. In 1988 Chadi and Chang [2] published their work which was accepted by most DX centre researchers. In their model, the DX centre is the substitutional donor atom itself which traps two electrons and moves towards an interstitial position, becoming negatively charged (DX^-). One of the most striking DX centre characteristics is the persistent photoconductivity (PPC), where the photoexcited conductivity at low temperature lasts for a long time, the lower the temperature, the longer the persistent photoconductivity. In 1989, Dmochowski *et al* [3] concluded that in indirect bandgap $Al_xGa_{1-x}As$, below 60 K, PPC has much lower magnitude since the shallow effective mass donor state is populated after photoexcitation of DX levels. After direct–indirect crossover the X-like state must participate in a carrier repopulation. The existence of an effective-mass-like level was demonstrated by Séguy and co-workers [4] using Raman scattering experiments. They detected bound phonons associated with electrons trapped at this level and did not find persistent photoconductivity effects. Based on the absence of PPC, a light induced experiment of thermally stimulated depolarization current (TSDC) [5] was proposed, and the existence

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of DX^--related electric dipoles in indirect bandgap $Al_xGa_{1-x}As$ was identified. Correlation among opposite charges such as DX^-–d^+, where d^+ means the substitutional donor after releasing an electron to the conduction band, have been proposed by some authors [6–8] in order to understand the electrical behaviour of samples containing DX centres. According to O'Reilly [6], in an electron capture process, the initial formation of DX^- centres involves replacing close pairs of positive charges by dipoles, because the capture is driven by electrostatic forces. Then for a large fraction of sites, d^+–DX^- are strongly correlated. These correlation effects may enhance the mobility and have been used to explain striking characteristics of $Al_xGa_{1-x}As$ based structures of complex geometries [9–11]. No great attention was given to indirect bandgap $Al_xGa_{1-x}As$ since the X valley (lowest energy in indirect bandgap material) has much lower mobility than the Γ valley, and then most of the work on DX centre in $Al_xGa_{1-x}As$ films were carried out in direct bandgap samples. Torres-Delgado and co-workers [12] measured the photoluminescence of indirect bandgap $Al_xGa_{1-x}As$ and concluded that for $x < 0.60$ the donor level may be related to the L valley rather than to the X valley, because the Si donor ionization energy is quite large. In this communication we report the variation of photoconductivity excitation with wavelength for indirect bandgap $Al_xGa_{1-x}As$ for a wide temperature range. A photoinduced electron trapping takes place for energy slightly higher than the bandgap transition below $\cong$34 K, and the spectra are DX centre dominated for temperatures higher than that, until about 120 K.

The sample used in this work was grown by molecular beam epitaxy on an undoped, semi-insulating GaAs substrate. In order to prevent a two-dimensional electron gas formation at the interface, since we are interested in the bulk properties, the sample has the following structure: 0.25 μm of undoped GaAs buffer layer, 0.25 μm of undoped $Al_xGa_{1-x}As$ that is compositionally graded from $x = 0$ to $x \cong 0.50$, 0.25 μm of undoped $Al_xGa_{1-x}As$ with $x \cong 0.50$ and 2.0 μm of indirect bandgap material ($x \cong 0.50$) active layer, that is 5×10^{17} cm^{-3} Si doped. Variation of photoconductivity excitation is obtained by scanning a monochromator at a fixed rate. The light source is a tungsten lamp. Although we have not monitored the photon flux, the light source spectrum intensity in the used wavelength range is rather uniform and we have precisely controlled the current of the light source at 10 A, with 10^{-2} A of precision, using a 68831 Radiometric power supply from Oriel Corporation. The scanning as well as the data acquisition from the electrometer are controlled by a microcomputer. All the spectra presented here are obtained with wavelength being scanned from higher to lower value.

2. Variation of photoconductivity excitation with wavelength and temperature

In figure 1 we show variation of photoconductivity excitation with wavelength for indirect bandgap $Al_xGa_{1-x}As$ for several temperatures. We start irradiating the sample with light with the beginning of scanning about 600 nm (or slightly different in some cases). Then the wavelength is decreased slowly which is controlled by a microcomputer. The scanning speed is about 9.8 nm min^{-1} from higher to lower wavelength. At 13 K there is a peak in the photoconductivity about 566 nm (2.19 eV) and the conductivity starts to decrease until about 561 nm (2.21 eV), where there is a local minimum and then there is another increase and another peak about 550 nm where a new decrease starts to take place. The first peak and the last decrease are expected since they correspond to the regular photoconductivity spectrum type of $Al_xGa_{1-x}As$, but the local minimum is rather unexpected. We can associate the peak at 566 nm with the bandgap transition, which is consistent with results for other temperatures since the peak wavelength position increases slightly with temperature as predicted by Adachi [13]. In figure 1(a) we apply 50 mV of voltage and measure the current. As the temperature is increased the current magnitude increases. It is not possible to measure the current with

50 mV at 17 K (figure 1(b)). Then we apply 100 mV. Since $Al_xGa_{1-x}As$ is very sensitive to the presence of light, there are oscillations in the measured current for this voltage, although the result is even worse for 50 mV (not shown). These oscillations are probably related to complex generation–recombination processes, since electrons coming from bandgap transitions have very short lifetime and are metastably kept in the conduction band by the presence of light. We record a very clean spectrum for 20 K using 100 mV (figure 1(c)), but as the temperature increases it becomes hard to control the oscillations in current as we can see in figure 1(d) at 25 K. Therefore we decrease once more the applied voltage and use 50 mV to carry out measurements at 34 K (figure 1(e)). The local minimum about 561 nm, observed at low temperature, vanishes about 34 K, leaving only a short plateau region around this wavelength.

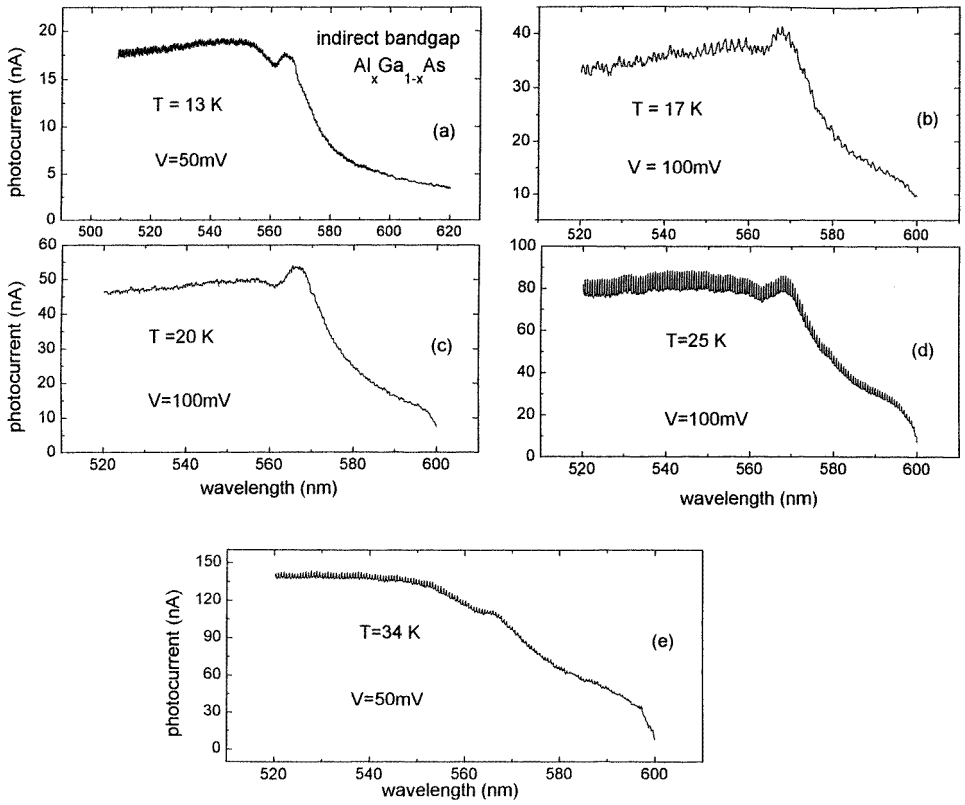

Figure 1. Variation of photoconductivity excitation with wavelength for indirect bandgap $Al_xGa_{1-x}As$ at several temperatures. Wavelength scanning speed = 9.8 nm min^{-1}. (a) 13 K, (b) 17 K, (c) 20 K, (d) 25 K, (e) 34 K.

For higher temperature there is no local minimum around 561 nm and the photoconductivity keeps increasing for energies higher than the bandgap transition as we can see in figure 2 for 40 K. This behaviour is very similar for all the spectra measured in the range 34–120 K, since the transport properties are dominated by DX centres in this range of temperature. It is not possible to evaluate the bandgap transition in this range since the beginning as well the end of the photoconductivity transition rate is not clearly observed in this range of temperature. In the beginning of the photoconductivity spectra, the monochromatic light has energy enough to excite electrons from DX centres and the excitation depends on time

of exposure and does not depend on the absolute value of energy, since it is much higher than the DX centre threshold photoionization energy [14]. As the bandgap transition is approached the photo-excitation rate is increased since we now have two combined processes: excitation from DX centres and bandgap transition. Therefore the photoconduction at the peak comes from electron–hole pairs generated from the bandgap transition and electrons coming from DX centres. As the wavelength becomes shorter the generation of electron–hole pairs is inhibited, but the photoconduction does not decrease, because electrons coming from DX centres, responsible for most of the transport, are under the PPC phenomenon. Above $\widetilde{=}$120 K, the transition excitation rate is much easier to observe as seen from the photoconductivity spectrum at 130 K, also shown in figure 2, since PPC is negligibly short. We estimate the Al composition as $x \widetilde{=} 56\%$ in this sample [13], which is far from the direct–indirect crossover and in the indirect bandgap region for $Al_xGa_{1-x}As$. In the inset of figure 2 we show the decay of excited photoconductivity using two different wavelengths and two temperatures. At 30 K the excitation with $\lambda = 566$ nm (which is the approximate peak position in figure 1), until saturation, leads to about 140 nA. After removing the light excitation, the current decays to about 120 nA where it remains constant practically forever. Illuminating with $\lambda = 561$ nm, the position of the local minimum in figure 1, the same behaviour is observed, but the value of current at saturation is lower and the persistent conductivity has a lower value, $\widetilde{=}$100 nA. This result indicates that there is a persistent component in the photoinduced current spectrum shown in figure 1 either for the peak or the local minimum and they have different amounts of free electrons. Illuminating the sample at 14 K a similar result is observed, but the current magnitude is lower and the distance between the persistent part of both decays is shorter than at 30 K.

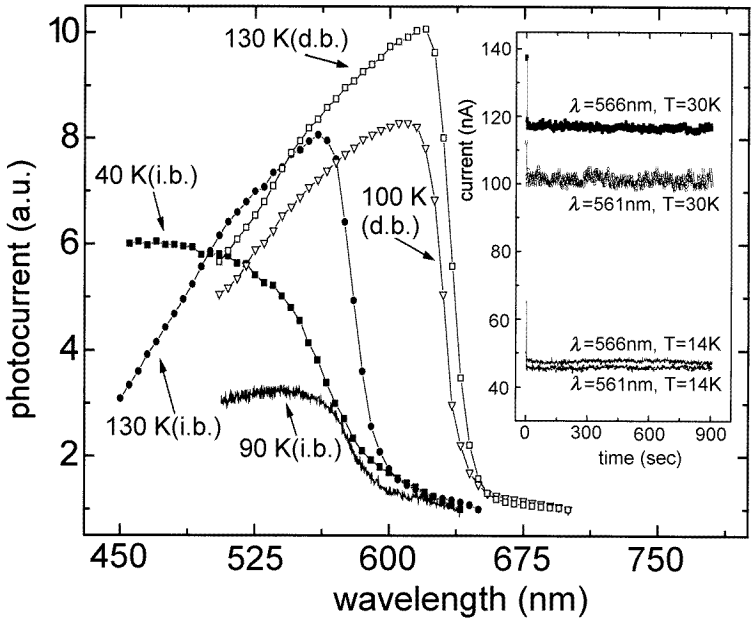

Figure 2. Variation of photoconductivity excitation with wavelength for indirect bandgap $Al_xGa_{1-x}As$ ($x \widetilde{=} 0.56$) at 40 K, 90 K and 130 K. Results for a direct bandgap (d.b.) $Al_xGa_{1-x}As$ sample ($x \widetilde{=} 0.32$) are also shown for comparison. Inset—decay of photoexcited current at low temperature. The temperature and excitation light of saturation are indicated.

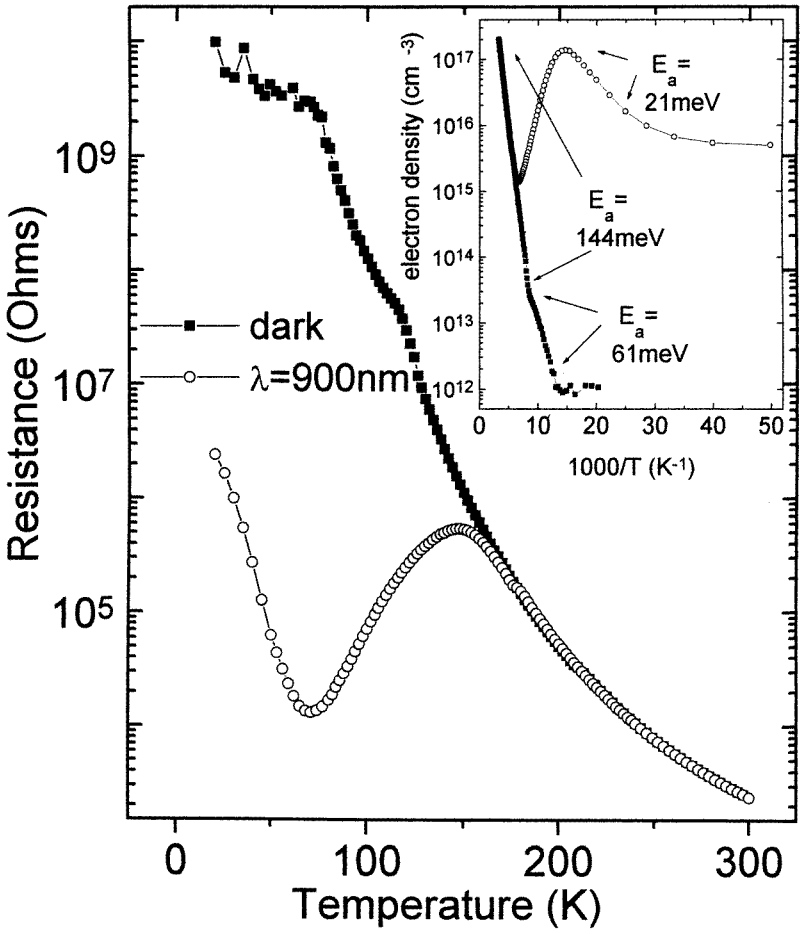

Figure 3. Resistance as function of temperature for indirect bandgap $Al_xGa_{1-x}As$ in the dark and under steady illumination with monochromatic light of $\lambda = 900$ nm. Inset—electron density as a function of $1000/T$ (Arrhenius plot) estimated from the resistance $\times$ temperature (RT) data of the inset, sample dimensions and electron scattering calculation.

Measured resistance as a function of temperature is shown in figure 3, where data are obtained by another photoconductivity procedure. Temperature is decreased to about 12 K in the dark and then it is allowed to increase very slowly ($\widetilde{=}$3 hours) when the sample resistance is measured. Temperature is decreased once more to 12 K and once more it is raised, this time under illumination with monochromatic light of $\lambda = 900$ nm ($\widetilde{=}$1.37 eV), with the help of a filter which cuts off wavelengths lower than 800 nm. Below $\widetilde{=}$70 K the resistance is higher than the value at 70 K, which represents a local minimum and has magnitude close to the value at room temperature. Since 1.37 eV is much lower than the bandgap transition energy, we believe that we have DX centre excitation since this illumination energy is much higher than the DX centre threshold photoionization energy [14], since DX centres were populated when the temperature was decreased in the dark. However the magnitude of photo-induced conductivity below $\widetilde{=}$70 K is lower, since there is trapping by another level. This result is in good agreement with figure 1 for the lower the temperature the lower the magnitude of

photoconductivity. As the temperature is raised this trapping becomes less important and the photoconductivity reaches its maximum about 70 K, where the resistance is minimum. The resistance increases again until about 150 K when it has maximum value, decreasing again and approaching the dark value of resistance about 200 K, which means that in the range 200–300 K light has very little influence on $Al_xGa_{1-x}As$ sample resistance. Free electron density may be estimated from these RT curves, as shown in the inset of figure 3. Resistivity is calculated from sample dimensions in order to estimate the free carrier concentration (n):

$$n = l[RAq\mu(n)]^{-1} \tag{1}$$

where R is the measured resistance, l is the distance between contacts (= 1.6 mm), A is the cross-sectional area, which is a parameter adjusted by electron concentration and mobility obtained by the Hall effect at 300 K, q is the electron charge and μ is the mobility. We have considered two electron scattering mechanisms [15] to obtain the mobility: ionized impurity scattering [16] and polar optical scattering [17] and the necessary parameters given by Adachi [13]. It is important to mention that ionized impurity concentration is kept constant since we are using the model of Chadi and Chang where the Si donor exists either in the d^+ state or DX^- state. A better estimation should take into account correlation among these charged impurities [6, 7]. Solving self-consistently all the equations (mobility is also a function of n, as indicated in equation (1)), we obtain electron density as a function of temperature as shown in the inset of figure 3. For resistance measured in the dark we have shown results with $1000/T$ ranging from 3.3. K^{-1} to 20 K^{-1}. We do not show results higher than that since RT results shown in the inset of figure 3 lead to a oscillatory behaviour in the low temperature range, probably due to limitations of the electrometer, and then it is hard to obtain any new information from the Arrhenius plot. Mizuta and Mori [18] have done a similar experiment in Si-doped indirect bandgap $A_xGa_{1-x}As$ and show the result only until about $1000/T = 8$ K^{-1}. It is interesting to note that the dark curve changes its slope very close to 8 K^{-1}, a result not observed by those researchers that claim that at low temperature the dark conductivity is too low to measure, as we agree, but our troubles were for lower temperature, as already discussed. For the range 3.3 to $\cong$8 K^{-1} our calculated activation energy (E_a) is 144 meV, very close to their result (136 meV [18]) and in the range $\cong$8–13 K^{-1} we estimated $E_a \cong 61$ meV. For the experiment done under steady illumination of $\lambda = 900$ nm we have an activation energy of 21 meV in the range 16–25 K^{-1}. Considering that our experimental procedure is rather different from Mizuta and Mori and the aluminium composition and Si doping are slightly different from their sample, both data are very consistent. They have used non-monochromatic light and illumination at 4 K with measurement in the dark whereas we have used $\lambda = 900$ nm monochromatic light and steady illumination while the temperature is raised. Therefore the minimum in the RT curve takes place at a lower temperature in our case.

3. Participation of the X-valley effective mass state, the DX^0 intermediate state and other levels in the recombination process

The variation of photoconductivity excitation with wavelength seen in figure 1 must be understood as follows: light at 600 nm has energy enough to excite electrons from DX centres, which is slowly excited since there is low photoionization rate (the amount of free carriers depends on exposure time). When the energy increases, bandgap transition is approached and the ionization rate becomes higher. The lower the temperature, the lower the photoconductivity signal which means that as electrons are excited from DX centres, they are retrapped at another level, which may be the X-valley effective mass state. The depth of the Si–X-like level might be of a similar order of magnitude as the $\cong$40 meV Te–X-like level [3]. The peak position in

figure 1 should not be associated with the bandgap transition energy since its location depends on scanning speed and the DX centre photoionization rate. One must be careful in associating the peak in photoconductivity spectral response with bandgap transition energy, since it is not a straightforward process. This maximum in photoconductivity occurs at a wavelength slightly longer than that corresponding to the absorption edge [19]. Even from the absorption spectral response curve the estimation of the energy gap is not a simple process, for transitions are subject to certain selection rules [20]. Moreover, one must account for the change in both energy and momentum since our sample has an indirect bandgap transition, which requires a two step process involving the participation of phonons that may be emitted or absorbed. Then around 566 nm there is a combination of electron–hole pairs generated from the bandgap transition and electrons from DX^- centres. As the exciting energy is increased, there is a decrease in the photocurrent, which may be associated with some kind of trapping. The X-valley effective mass state has an influence on the current magnitude at low temperature as shown in figures 1 and 3. The lower PPC magnitude found by Dmochowski and co-workers [3] for Te-doped $Al_xGa_{1-x}As$ is in good agreement with our results. Then the photoinduced trapping at $\widetilde{=}561$ nm must have another explanation.

For high temperature, the DX^- centre is photoionized, according to the reaction [2]:

$$DX^- \rightarrow d^+ + 2\,e^- . \tag{2}$$

This reaction means that besides releasing two electrons to the conduction band, the DX^- centre relaxes back to its original position, substitutional in a Ga site. For lower temperature, there is another reaction probable:

$$d^+ + DX^- \rightarrow d^0 + DX^+ + 1\,e^- \tag{3}$$

where DX^+ means the Si dopant after releasing two electrons to the conduction band but before relaxing to the substitutional position. This reaction means that one of the two electrons released by the DX^- state is captured by a X-valley effective mass state of another atom. According to our data this reaction is more probable as the temperature gets lower, since the excited photocurrent becomes lower. However the photoconductivity spectrum at low temperature presents a decrease in the current magnitude about 560 nm, which suggests another mechanism of trapping. We believe that at temperature as low as 13 K, the thermal energy is low and does not allow relaxation immediately. The DX^+ state is highly unstable and it is located $\widetilde{=}20$ meV above the conduction band minimum. Then it is able to recapture one electron from electron–hole pairs of the bandgap transition and becomes DX^0 according to the reaction

$$DX^+ + 1\,e^- \rightarrow DX^0 . \tag{4}$$

For illuminating energies higher than the DX^+ level, we have bandgap transition (although less efficient) and DX^- excitation by reactions (2) and (3). In both cases there is a release of electrons to the conduction band. As the temperature is raised the DX^+ lifetime becomes shorter and a decrease in the photoconductivity is no longer observed. A DX^0 state has been already proposed by some authors [21–23] and has been used again in a very recent paper [24]. In indirect bandgap $Al_xGa_{1-x}As$, in the range 13–30 K, the thermal energy is quite low and the DX^0 intermediate state has a lifetime long enough to be responsible for the local minimum in the photoconductivity curve. The experimental decay of photoexcited current shows that even at 14 or 30 K the current is excited to a higher value by using $\lambda = 566$ nm and the persistent part is also higher in this case; however the distance between the persistent current after excitation with 566 nm and 561 nm is larger at 30 K than at 14 K. This result means that after removing the light, bandgap transitions immediately take place, but states d^0 and DX^0, generated by reactions (2) and (3), are metastably kept by the low temperature. The reason

for the similarity in persistent part at 14 K is probably that, at this low temperature, impurity states of A_1 symmetry as proposed by Dmochowski *et al* [25] may be taking part in the capture process.

Another interesting point is that the energy of the photoconductivity local minimum is just above the bottom of the conduction band. It is well known that As vacancies (V_{As}) have an unoccupied level resonant with the conduction band [26, 27]. Van Vechten [28] predicts the participation of vacancies in the defects present in the sample, since there is a tendency for a V_{As} centre to be formed close to a donor. Thermally stimulated depolarization current measurements under the influence of monochromatic light [5] have revealed the possibility of V_{As} participation in the recombination process at low temperature. Therefore, the local minimum in the photoconductivity response may be related to trapping by As vacancies located close to DX^- centres.

4. Conclusion

We have confirmed the X-valley effective mass state as responsible for the low PPC signal below $\widetilde{=}$70 K, for Si-doped $Al_xGa_{1-x}As$, similarly to that observed previously for the Te donor [3]. The lower the temperature the lower the induced photocurrent, unlike what occurs for direct bandgap material. In the range 13–30 K we have found a decrease in the photoconductivity excitation with energy slightly higher than the bandgap transition, followed by another increase. We have interpreted this behaviour as capture by a DX^0 intermediate state, which in this case would be photoinduced. However the actual nature of such a local minimum in the photoconductivity needs further research.

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