

# Light and Thermal Excitation of Depolarization Current in Indirect Bandgap $\text{Al}_x\text{Ga}_{1-x}\text{As}$

Luis V. A. Scalvi and L. Oliveira

*Departamento de Física, Universidade Estadual Paulista,  
campus Bauru, Caixa Postal 473, Bauru, SP, 17033-360 Brazil*

M. Siu Li

*Departamento de Física e Ciência dos Materiais, Instituto de Física de São Carlos - USP  
Caixa Postal 369, São Carlos, SP, 13560-970, Brazil*

Received August 25, 1995

Monochromatic light excitation in conjunction with thermally stimulated depolarization current measurements are applied to indirect bandgap  $\text{Al}_x\text{Ga}_{1-x}\text{As}$ . The obtained average activation energy for dipole relaxation is in very close agreement with the DX center binding energy. Monochromatic light induces state transition in the defect and makes possible the identification of dipoles observed in the dark. Charge relaxation currents are destroyed by photoionization of  $\text{Al}_{0.5}\text{Ga}_{0.5}\text{As}$  using either 647 nm  $\text{Kr}^+$  or 488 nm  $\text{Ar}^+$  laser lines, which are above the DX center threshold photoionization energy. It suggests that correlation may exist among charged donor states  $\text{DX}^- - \text{d}^+$ . Sample resistance as a function of temperature is also measured in the dark and under illumination and shows the probable X valley effective mass state participation in the electron trapping. Ionization with energies of 0.8 eV and 1.24 eV leads to striking current peak shifts in the thermally stimulated depolarization bands. Since vacancies are present in this material, they may be responsible for the secondary band observed in the dark as well as participation in the light induced recombination process.

## I. Introduction

The striking properties of the well-known DX center in  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  has led many scientists to spend a good deal of work in its investigation in the past two decades. Although most of physics of this deep level has already been established, the peculiar situation by which the defect most likely presents a non-neutral ground state<sup>[1]</sup> has provided explanation for some new phenomena in  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  based structures<sup>[2,3]</sup>. The presence of electric dipoles in  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  has recently been demonstrated in direct bandgap sample<sup>[4]</sup> as well as indirect bandgap material<sup>[5]</sup> by using thermally stimulated depolarization current measurements (TSDC). The close agreement of dipole activation energy with the DX center binding energy and the use of asymmetric distribution of energies and dipole lengths has led us to associate them with DX centers (impurity donor placed at interstitial site and negatively charged).

O'Reilly<sup>[6]</sup> stated that mobility data in GaAs under hydrostatic pressure can only be explained if  $\text{DX}^- - \text{d}^+$  correlations are taken into account, where  $\text{d}^+$  means the substitutional impurity donor after releasing one electron. Preliminary measurements using TSDC in combination with monochromatic light excitation<sup>[7]</sup> indicates that this  $\text{DX}^- - \text{d}^+$  interaction can actually be the case, but there is also some possibility of carrier trapping by vacancies, which becomes very important in creating another dipole system, since vacancies do exist in  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  as assured by Van Vechten<sup>[8]</sup>. In this paper we present results of thermally stimulated depolarization current in conjunction with monochromatic light of several wavelengths in an indirect bandgap  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  sample. Monochromatic light excitation does not produce persistent photoconductivity (PPC) in this material. Then it is expected that no electronic current is photoinduced, due to absence of PPC, and moreover, TSDC bands obtained in the dark should

vanish because DX center-related dipoles are destroyed by this experimental procedure.

## II. Light induced dipole relaxation in indirect bandgap $\text{Al}_x\text{Ga}_{1-x}\text{As}$

The sample used in this work has been described elsewhere<sup>[4,7]</sup> and it is basically a molecular beam epitaxy (MBE) grown  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  structure on a semi-insulating GaAs substrate. The active layer has  $2\ \mu\text{m}$  of  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  where  $x \approx 0.50$ , doped with  $5 \times 10^{17}\ \text{cm}^{-3}$  Si.

In usual TSDC experiments, a voltage is applied to the sample at room temperature in the dark. It is used an electric field of  $15.7\ \text{V/cm}$ . Then the temperature is decreased to about  $10\ \text{K}$ , where the polarization voltage is removed and the temperature is allowed to increase at fixed rate. Using the same contacts where the voltage is applied, the current is now measured with an electrometer. Electric dipoles are polarized during cool down and relax to their original random distribution as the temperature is increased<sup>[4]</sup>. Since in usual TSDC procedure no light is shined on the sample, there is no photoionization of electrons to the conduction band when the temperature increases.

Fig. 1 shows recorded TSDC bands for  $\text{Al}_{0.5}\text{Ga}_{0.5}\text{As}$ . The inset in Fig. 1 is the usual TSDC procedure for this sample, where all the data is recorded keeping the sample in the dark. The measured current is clearly not electronic, considering its low magnitude and because no voltage is applied to the sample when the temperature is increased. No known process could be responsible for releasing a few electrons to the conduction band at low temperature and recapture at higher temperature in the dark. Our interpretation is that observed current is due to electric dipole reorientation<sup>[4,5]</sup>. In order to fit the data we use Havriliak-Negami relaxation time distribution approach<sup>[9]</sup>, which is shown in the inset in Fig. 1 by the continuous line. A distribution is needed since we have dipole-length distribution and Al atoms are randomly distributed around the defect<sup>[4]</sup>, which suggests distinct activation energies due to distinct neighborhood. There is two bands in the inset of Fig. 1, reproduced by stars in the main figure. The major one has average activation energy  $E_a \approx 0.132\ \text{eV}$

and the other current peak with lower intensity has  $E_a \approx 0.165\ \text{eV}$ . Although both peaks have activation energy value very close to the DX center binding energy for this Aluminum composition<sup>[10]</sup>, our results with direct bandgap material<sup>[4]</sup> and the preliminary investigation with monochromatic light excitation in dipole relaxation<sup>[7]</sup> indicate that the main peak is the DX center related one. We also expect a negatively charged DX center ground state, since no neutral state could be responsible for the obtained dipole relaxation current. Since there are many vacancies in the  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  sample<sup>[8]</sup>, we believe that they could trap electrons to become  $V_{As}^-$ . The  $V_{As}$  unoccupied level is resonant with the conduction band, while the occupied level relaxes to a bound state in the gap<sup>[11]</sup>.  $V_{As}^-$  presents a Jahn Teller distortion in its position<sup>[12]</sup> and could be responsible for the dipole system that generates the lower TSDC band shown in Fig. 1. This figure also shows the effect of monochromatic light of two distinct wavelengths in the TSDC band, that we call modified TSDC. This procedure can be shortly explained as being the same of usual TSDC with the only modification that when the temperature is at its minimum value ( $\approx 10\ \text{K}$ ), monochromatic light irradiates the sample for some time. The sample is returned to darkness and the electric field is removed. Then, the temperature is increased at fixed rate. We use  $\lambda = 488\ \text{nm}$  from an  $\text{Ar}^+$  laser line, which is above the bandgap transition or  $\lambda = 647\ \text{nm}$  from a  $\text{Kr}^+$  laser line, which is below the bandgap energy but well above the DX center threshold photoionization energy (about  $0.8\ \text{eV}$  independent of aluminum mole fraction<sup>[13]</sup>). The irradiation time is 5 minutes. Either laser line destroys the TSDC bands shown in Fig. 1. This is the expected behavior for a dipole system DX center-related. The current peak at  $62\ \text{K}$  is also destroyed by each laser lines. The same effect is obtained using a tungsten lamp and a monochromator.

In Fig. 1 it is also presented the effect of illuminating the sample with  $\lambda = 1.55\ \mu\text{m}$ , the DX center threshold photoionization energy, for different exposure times. This is obtained from a monochromator with a tungsten lamp source and a filter which cuts off wavelengths below  $\approx 800\ \text{nm}$ . This light does not destroy the TSDC bands but induces a shift of the main current peak from

50 K (regular TSDC) to another temperature, which is the lower the longer the exposure time.

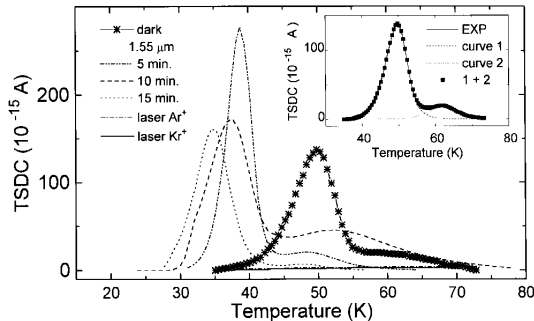


Figure 1. Thermally stimulated depolarization current in the darkness for  $Al_{0.5}Ga_{0.5}As$  and influence of monochromatic light in TSDC bands for  $Ar^+$  laser excitation (488 nm),  $Kr^+$  laser excitation line (647 nm) and TSDC bands after irradiating 1.55  $\mu m$  excitation line for 5 to 15 minutes. Inset - TSDC obtained in the dark and data fitting using Havriliak-Negami approach.

We chose an indirect bandgap sample because when monochromatic light is used to excite electrons from DX centers to the X valley conduction band, below  $\approx 60$  K, the sample does not show PPC, because electrons are immediately trapped metastably by the X valley effective mass state<sup>[14-16]</sup>. Fig. 2 shows the resistance as a function of temperature for this sample in the dark and under illumination with  $\lambda = 488$  nm  $Ar^+$  laser line and with  $\lambda = 1.55 \mu m$ . In the dark, the resistance increases as the temperature is lowered down, which is an expected behavior in  $Al_xGa_{1-x}As$  of any Al composition, due to electron freezeout at DX centers, which is more effective above 150 K to about 60 K, when the resistance becomes practically constant. The temperature lowering is slow enough ( $\approx 5$  hours) to prevent any sort of metastability. Keeping the laser line illuminating the sample during the cooling, electrons remain in the conduction band, mainly due to photoexcitation of DX centers. The increase in resistance, starting about 60 K is a very good indication of the participation of the X valley effective mass state in the recombination process. Results of transient decay of persistent photoconductivity presented earlier<sup>[17]</sup> show the existence of this type of trapping. The illumination with  $\lambda = 1.55 \mu m$  has an intermediate photoexcitation, as expected, since 0.8 eV is too low to completely photoexcite the DX center, particularly in this composition where the photoionization cross-section increases over 6 orders of magnitude when the excitation light energy goes from 0.8 eV to

2.0 eV [13]. Therefore, below  $\approx 60$  K, indirect bandgap  $Al_xGa_{1-x}As$  is suitable for testing the origin of TSDC bands, since no electronic current is photogenerated in this sample. It assures that even after monochromatic light excitation, the measured thermally stimulated current comes from relaxing dipoles. The X-valley effective mass state after trapping electrons metastably, does not induce a new TSDC band because it is a neutral state ( $d^0$ ) placed at a substitutional site.

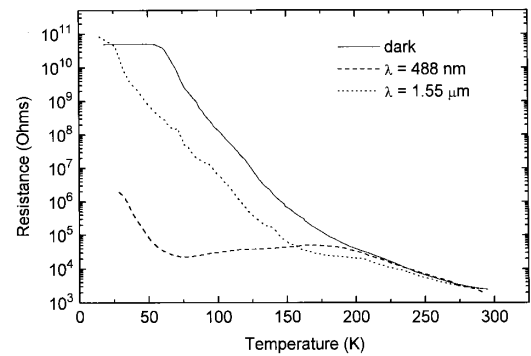


Figure 2. Resistance as a function of temperature in the dark and under constant illumination of  $Ar^+$  laser excitation line (488 nm) and 1.55  $\mu m$  excitation line.

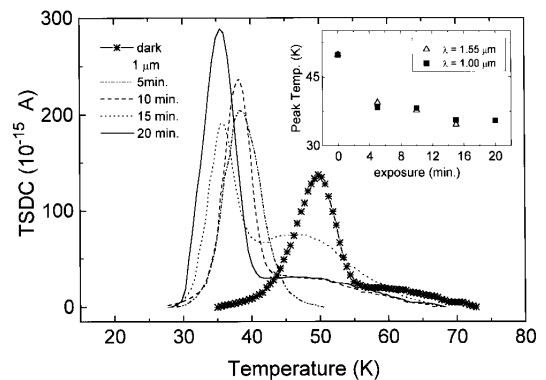


Figure 3. Influence of irradiating  $\lambda = 1.0 \mu m$  excitation line in TSDC bands for  $Al_{0.5}Ga_{0.5}As$ , for 5 to 20 minutes. Inset-main peak temperature for several exposure times for 1.55  $\mu m$  and 1.0  $\mu m$  excitation lines.

Fig. 3 shows the effect of illumination with another wavelength,  $\lambda = 1.0 \mu m$ , which is above the DX center threshold energy, but not yet in the saturation region<sup>[13,18]</sup>. The results are similar to those obtained for illumination with  $\lambda = 1.55 \mu m$ , where the main current peak is shifted to a lower temperature. The results obtained from numerical simulation of TSDC bands shown in Figs. 1 and 3 are summarized in table I. The illumination with 5 minutes of monochromatic

light of  $1.55 \mu\text{m}$  induces a shift of the main peak from 50 K, obtained in the dark to 39 K, corresponding to an average activation energy for dipole relaxation of 0.121 eV. There remains some dipole relaxation current with average activation energy of 0.132 eV, but this dipole system has lower concentration. For  $\lambda = 1.55 \mu\text{m}$  the main peak becomes narrower and higher, meaning that the dipole distribution is not as wide as in the darkness, but concentrated around a specific dipole system. As shown in Fig. 1 and table I, the longer the illumination time, the lower the peak temperature and the lower the average activation energy of the main peak. No matter how long the exposure time with  $1.55 \mu\text{m}$  illumination, there is always a remaining peak in position very close to the TSDC band obtained in the dark, which means that the original dipole system has not been completely destroyed even after 15 minutes of illumination. It is also interesting to notice that  $\tau_0$ , the average constant relaxation time has the same order of magnitude ( $\approx 10^{-12}$  sec) for the TSDC band obtained in the dark and for all the secondary current peaks after illumination. This is clearly described in table I. Main bands obtained after  $1.55 \mu\text{m}$  light irradiation for different exposure times also have  $\tau_0$  with the same order of magnitude ( $\approx 5 \times 10^{-13}$  sec), which is more than two orders of magnitude lower than the band obtained in the dark. Also in table I are the results of exposure to  $\lambda = 1.0 \mu\text{m}$  at 10 K. The bands are in very good agreement with results obtained for  $\lambda = 1.55 \mu\text{m}$ , since the main peak for illumination with  $1.0 \mu\text{m}$  is shifted to a lower temperature and the average activation energy is even lower than those obtained from data fitting of the main peaks shown in Fig. 1. It is expected since 1.24 eV is fairly higher than the DX center threshold photoionization energy, but not in the saturation region. Then the  $1.0 \mu\text{m}$  excitation line drives the bands to a lower dipole relaxation energy, but does not destroy the band, unlike laser lines excitation.  $\tau_0$  has a value ( $\approx 5 \times 10^{-14}$  sec) between the value calculated for the main band and the values for the secondary bands of Fig. 1. In the case of  $1.0 \mu\text{m}$  we still have secondary peaks with activation energies and average constant relaxation time coincident with the main peak obtained in the dark. It means that the dipole system with  $E_a \approx 0.132$  eV is not completely destroyed by illumination with this wavelength, a similar result to what happens

when the monochromatic light has  $\lambda = 1.55 \mu\text{m}$ . There is a tendency for driving the main peak position to lower temperature as the exposure time gets longer, either for  $1.0 \mu\text{m}$  curves or  $1.55 \mu\text{m}$  curves. This is seen in the inset of Fig. 3. It seems that the temperature is going to steady at some limiting value, since the peak position seems to have exponential-like behavior.

Our interpretation for the recorded shifted bands is as follows: in indirect bandgap  $\text{Al}_x\text{Ga}_{1-x}\text{As}$ , there are three states responsible for trapping the electrons, the X-valley effective mass state, the DX center and the As vacancy, the occupation obeys some statistical distribution. The first is a neutral state ( $d^0$ ) which can not generate a TSDC band. DX and  $V_{As}^-$  are negatively charged and could be responsible for the two peaks obtained in the dark and seen in Fig. 1. This is quite consistent with the much lower dipole current magnitude recorded in this sample, if compared to direct bandgap  $\text{Al}_x\text{Ga}_{1-x}\text{As}$ <sup>[4,5]</sup>, which does not present a double TSDC band. The DX<sup>-</sup> ground state can be photoexcited, releasing electrons to the conduction band. When excited with 0.8 eV, the DX is taken to an activated state which can release electrons to the X valley due to parabolic potential [7]. The same behavior is expected for illumination with 1.24 eV, the excited state being located higher than in the case of Fig. 1, but still not crossing the parabolic X valley potential. Some electrons can populate the conduction band and be metastably trapped by the X valley effective mass state. Both wavelengths, although not enough to deplete the DX<sup>-</sup> related dipole system, destroy the peak obtained at 62 K. Lang *et al.*<sup>[11]</sup> suggest a Franck-Condon shift of about 0.8 eV for the vacancy transition between occupied and unoccupied states. We believe that the peak at 62 K is destroyed by the  $V_{As}^-$  transition to an unoccupied vacancy level. In both cases, of illumination with  $1.55 \mu\text{m}$  and  $1.0 \mu\text{m}$ , there is generation of a much more defined dipole system, concentrated around a specific system, after some exposure time. The excited DX level is able to give electrons to vacancies located close to  $d^+$  center, since there is a tendency for a  $V_{As}^-$  to be formed close to a Si donor<sup>[8]</sup>. Then a more localized  $d^+ - V_{As}^-$  dipole system is generated. This is observed in Fig. 1 with 5 minutes of illumination. This assumption is reinforced by the fact that light is applied in

Table I - Parameters for TSDC data fitting in  $\text{Al}_{0.5}\text{Ga}_{0.5}\text{As}$  using Havriliak-Negami asymmetric relaxation time distribution approach.

Temperature main peak (K)	$E_a$ (eV)	$\tau$ ( $10^{-14}$ sec)	Temperature sec. peak (K)	$E_a$ (eV)	$\tau$ ( $10^{-14}$ sec)	OBS.
49.8	0.132	92	62.1	0.165	97	dark
39.4	0.121	0.5	48.6	0.132	97	$\lambda=1.55\mu\text{m}$ - 5 minutes
37.7	0.114	0.75	52.7	0.143	97	$\lambda=1.55\mu\text{m}$ - 10 minutes
34.6	0.110	0.59	47.7	0.129	97	$\lambda=1.55\mu\text{m}$ - 15 minutes
38.3	0.104	30.0	—	—	—	$\lambda=1.0\mu\text{m}$ - 5 minutes
38.2	0.109	6.0	48.5	0.133	97	$\lambda=1.0\mu\text{m}$ - 10 minutes
35.6	0.102	5.0	46.3	0.132	97	$\lambda=1.0\mu\text{m}$ - 15 minutes
35.5	0.102	4.5	48.7	0.133	97	$\lambda=1.0\mu\text{m}$ - 20 minutes

conjunction with the electric field, which could drive the electrons from DX centers to vacancies. There is another kind of  $V_{As}$  related dipole, since the occupied  $V_{As}$  is subject to a Jahn-Teller distortion.

For excitation with 0.8 eV, as the illumination time gets longer than 5 minutes, electrons populate more vacancies leading to the broader TSDC band observed, since the dipole length distribution and aluminum neighborhood widens the distribution. For excitation with 1.24 eV, after 15 minutes of illumination the secondary peak becomes fairly high, reaching almost half of the main peak height. When the exposure time is 20 minutes, the secondary peak becomes very low and the main peak is the highest recorded for this excitation line, the area under the curve remaining practically constant. Once again the more localized dipole  $d^+ - V_{As}^-$  system is generated and the number of similar pairs seems to increase with the exposure time. There is also the possibility that the excited DX center state has negative charge and could form pairs with vacancies<sup>[7]</sup>.

## Conclusion

Electric dipoles present in  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  have relaxing characteristics strongly influenced by the wavelength of irradiation light. The way wavelength affects TSDC bands is in close agreement with measurements of resistance as a function of temperature, under influence of  $\lambda = 488$  nm  $\text{Ar}^+$  laser line and  $\lambda = 1.55\mu\text{m}$  monochromatic light. In the saturation region of DX center photoionization<sup>[13]</sup>, light destroys the dipole current, which does not happen when the excitation en-

ergy is about the DX center photoionization threshold energy, neither higher than this energy, since it is still below the saturation region. The presence of the X-valley effective mass state is also verified in the curve of resistance under illumination. Results presented here, reinforced by results obtained for direct bandgap  $\text{Al}_x\text{Ga}_{1-x}\text{As}$ <sup>[4,5]</sup> strongly suggests that a DX ground state is related to TSDC bands obtained in the dark. This dipole system could be  $\text{DX}^- - d^+$  pairs as predicted by O'Reilly<sup>[6]</sup>. The presence of vacancies in these samples is also verified, since a secondary peak is always present in the TSDC spectrum, either in the dark or after illumination with 0.8 eV or even with 1.24 eV monochromatic light.

## Acknowledgments

This sample was grown at Oregon State University, U.S.A.. Luis V.A. Scalvi would like to thank profs. John Arthur and John Wager for the hospitality and technical support during his stay in Corvallis. The authors acknowledge Brazilian financial agencies: CNPq, CAPES, FAPESP and FINEP.

## References

1. D. J. Chadi and K. J. Chang, Phys. Rev. **B39**, 10063 (1989).
2. E. Buks, M. Heiblum and H. Shtrikman, Phys. Rev. **B49**, 14790 (1994).
3. C. Ghezzi, A. Parisini and V. Dallacasa, Phys. Rev. **B50**, 2166 (1994).
4. L. V. A. Scalvi, L. Oliveira, E. Minami and M. Siu Li, Appl. Phys. Lett. **63**, 2658 (1993).

5. L. V. A. Scalvi, L. Oliveira and M. Siu Li, *Mat. Res. Soc. Symp. Proc.* **325**, 285 (1994).
6. E. P. O'Reilly, *Appl. Phys. Lett.* **55**, 1409 (1989).
7. L. V. A. Scalvi, L. Oliveira and M. Siu Li, *Phys. Rev. B* **51**, 13864 (1995).
8. J. A. Van Vechten, *J. Phys. Condens. Matter* **1**, 5171 (1989).
9. P. Bräunlich, *Thermally Stimulated Relaxation in Solids*, (Springer Verlag, New York 1979).
10. N. Chand, T. Henderson, J. Klem, W. T. Masselink, R. Fischer, Y.C. Chang and H. Morkoç, *Phys. Rev.* **B30**, 4481 (1984).
11. D. V. Lang, R. A. Logan and M. Jaros, *Phys. Rev.* **B19**, 1015 (1979).
12. M. Jaros and S. Brand, *Phys. Rev.* **B14**, 4494 (1976).
13. P. M. Mooney, G. A. Northrop, T. N. Morgan and H. G. Grimmeiss, *Phys. Rev.* **B37**, 8298 (1988).
14. J. E. Dmochowski, L. Dobaczewski, J. M. Langer and W. Jantsch, *Phys. Rev.* **B40**, 9671 (1989).
15. L. Dobaczewski and P. Kaczor, *Phys. Rev.* **B44**, 8621 (1991).
16. P. Séguy, M. Zigone and G. Martinez, *Phys. Rev. Lett.* **68**, 518 (1992).
17. L. V. A. Scalvi, L. Oliveira, E. Minami, M. H. Degani and M. Siu Li, *Braz. J. Phys.* **24**, 379 (1994).
18. R. Legros, P. M. Mooney and S. L. Wright, *Phys. Rev.* **B35**, 7505 (1987).