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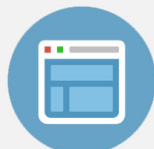
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Dipole relaxation current in n -type $\text{Al}_x\text{Ga}_{1-x}\text{As}$

L. V. A. Scalvi

Departamento de Física, Universidade Estadual Paulista Júlio de Mesquita Filho-Bauru, Caixa Postal 473, Bauru-SP 17033, Brazil

L. de Oliveira, E. Minami, and M. Siu-Li

Departamento de Física e Ciência dos Materiais, Instituto de Física e Química de São Carlos, Universidade de São Paulo, Caixa Postal 369, São Carlos-SP 13560, Brazil

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We report for the first time the thermally stimulated depolarization current (TSDC) spectrum for a direct band-gap AlGaAs sample, where the presence of DX centers is clearly observed by photoconductivity measurements. A TSDC band is obtained, revealing the presence of dipoles, which could be attributed to DX^-d^+ pairs as indeed predicted by O'Reilly [Appl. Phys. Lett. **55**, 1409 (1989)]. The data are fitted by relaxation time distribution approach yielding an average activation energy of 0.108 eV. This is the most striking feature of our data, since this energy has approximately the same value of the DX center binding energy.

Self-consistent total energy calculations were performed by Chadi and Chang¹ to show that the DX center is just another state of the substitutional donor itself which traps two electrons and moves along a $\langle 111 \rangle$ direction towards an interstitial site. Although the hypothesis of migration was first proposed by Morgan,² the striking feature of Chadi and Chang's model is the trapping of two electrons by the same center becoming negatively charged. At low temperatures the concentration of neutral impurities is negligible since the DX center state of the substitutional impurity is much more stable.^{1,3} Charge balance assures that the creation of a certain number of DX^- centers generates the same amount of d^+ centers. DX and d represent threefold (interstitial) and fourfold (substitutional) coordinated states of the same atom, respectively. In such a picture DX has a negative- U effective interaction with strong electron-phonon coupling. Although Chadi and Chang's model has been accepted by most DX center researchers, Maude *et al.*^{4,5} have used mobility data to support a positive- U model. These data show increasing mobility as the free carrier density decreases with pressure in highly doped GaAs. In a positive- U model trapout occurs as impurity donors become neutral, so the number of ionized scattering centers decreases with decreasing free carrier concentration. In the negative- U Chadi and Chang's model, the number of ionized impurity is essentially unchanged since the donor exists either in a d^+ state or DX^- state. In both models, scattering centers are isolated ionized impurities since the donors are uniformly distributed throughout the sample.

O'Reilly⁶ has argued that there is another important point which should be taken care of: d^+ and DX^- centers will be strongly correlated and a dipole-like picture should be used to describe scattering by DX^-d^+ pairs. In order for the DX^-d^+ to be considered as a dipole, the d^+ center should be located close enough to the DX^- center such that it may be a perturbation in the DX potential. If impurity donors are randomly distributed in the GaAs sample, the probability that d^+ and DX^- are first neighbors is negligibly low. However, the extra electrostatic energy gained by placing a d^+ center close to a DX^- center is of

significant magnitude at a large fraction of sites.⁶ Then it is energetically very favorable for a DX^- center to be created close to a d^+ center. This behavior is quite different from conventional samples, where charged defects are introduced during growth, usually at high temperatures.

DX centers are present in n -type $\text{Al}_x\text{Ga}_{1-x}\text{As}$ with x higher than approximately 0.22 with no need of applying pressure. Our measurements are carried out in a Si-doped ($\approx 1 \times 10^{18} \text{ cm}^{-3}$) molecular beam epitaxy (MBE) grown $\text{Al}_x\text{Ga}_{1-x}\text{As}$ sample 2 μm thick. Care was taken to avoid the formation of a two-dimensional electron gas (2DEG) using the following sample structure: a 0.25 μm undoped GaAs buffer layer was grown on the semi-insulating substrate followed by a 0.25 μm of undoped $\text{Al}_x\text{Ga}_{1-x}\text{As}$ that is compositionally graded from $x=0$ to $x \approx 0.32$, a 0.25 μm layer of undoped $\text{Al}_{0.32}\text{Ga}_{0.68}\text{As}$ and the 2 μm active layer. The photoconductivity spectrum at 10 K is shown in Fig. 1. The wavelength is scanned from higher to lower value. The wavelength scanning speed is 50 nm/min. At about 640 nm there is a sharp increase in the conductance which ends at about 620 nm. Using the equations given by Adachi

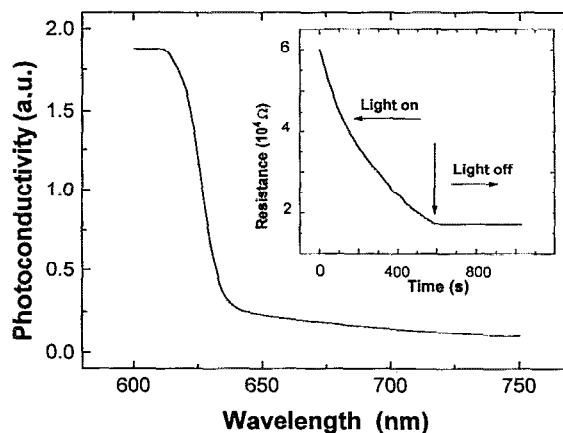


FIG. 1. Photoconductivity spectrum for the $\text{Al}_x\text{Ga}_{1-x}\text{As}$ sample at about 10 K. Inset—illumination of this sample proceeds until 600 s with a 560 nm monochromatic light, then light is turned off and the conductance remains steady ($T \approx 10$ K).

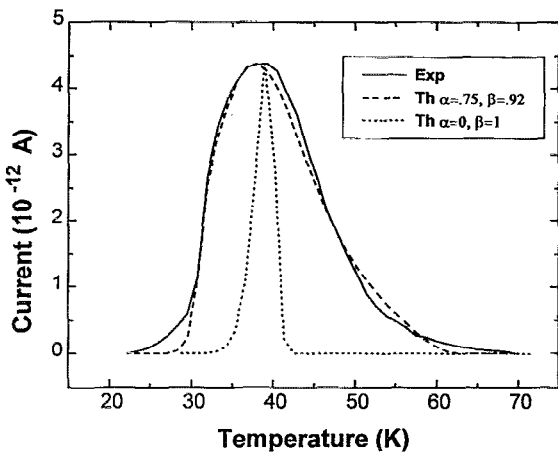


FIG. 2. Thermally stimulated dipole current for the $\text{Al}_x\text{Ga}_{1-x}\text{As}$ sample. Solid line—experimental data. Dashed line—Havriliak–Negami fit (see Ref. 10). Dotted line—single relaxation time approach.

and an average wavelength (630 nm) the band gap is estimated, considering the temperature corrections for the Γ valley.⁷

$$E_g^\Gamma(10 \text{ K}) = E_g^\Gamma(300 \text{ K}) + (-3.95 - 1.15x)10^{-4}\Delta T, \quad (1)$$

where

$$E_g^\Gamma(300 \text{ K}) = 1.424 + 1.247x, \quad (2)$$

resolving Eqs. (1) and (2) for 630 nm, it is found that $x \approx 0.32$. In this composition, it is expected that DX center dominates the transport properties, since DX energy level is located below the conduction band. There are two features in Fig. 1 which confirm this behavior: When the monochromatic light energy is around the forbidden gap transition, there is a slow increase in the photoconductivity which is a DX center characteristic. Above this transition the electrons do not return to the nonconductive state, trapped at DX centers. Besides, the inset in Fig. 1 shows that after illuminating the sample with a monochromatic light of 560 nm the resistance decreases slowly to a steady value, close to its value at room temperature ($\approx 1 \text{ k}\Omega$). In this figure we have measured the current with 50 mV of applied voltage. Then the illumination is removed and the resistance remains unchanged for an unmeasurable time. This is a well-known property related to DX centers and called persistent photoconductivity (PPC). So we can deduce from Fig. 1 that the $\text{Al}_x\text{Ga}_{1-x}\text{As}$ sample has a direct forbidden band-gap transition ($x \approx 0.32$) and shows PPC, a property related to DX center in bulk AlGaAs and furthermore, it shows no 2DEG conduction since the resistance does not reach a value lower than at room temperature.

Figure 2 shows the experimental TSDC spectrum (solid line). In this measurement the sample is biased at room temperature (2.5 V). Then the temperature is lowered down to liquid He temperature in the dark. The applied bias is removed and the temperature is allowed to increase at fixed rate (0.081 K/s), always in the dark, when the depolarization current is measured with an electrometer and recorded. In other words, no illumination is

done on the sample during the whole measurement. As the temperature goes down in the dark an electron freezeout at the DX centers occurs. Considering that there is no light on the sample, no electron is photoexcited to the conduction band and, moreover, the electron thermal energy is not enough to overcome the emission barrier. The only way to explain the current peak seen in Fig. 2 is by a dipole reorientation current. The dipoles, originally randomly distributed throughout the sample, are biased with the applied voltage at room temperature. When the applied bias is removed at liquid-helium temperature, the oriented dipoles are frozen at a metastable state. Then, the temperature is allowed to increase at a fixed rate and the dipoles relax to their equilibrium positions generating a depolarization current shown in Fig. 2, with a peak at 39 K. A strong evidence that it is indeed, a dipole reorientation current is the order of magnitude of the experimental curve. Comparing the values calculated from Fig. 1 (inset) using 50 mV of applied voltage, and values from Fig. 2 it is clearly seen that the electronic photoinduced current is about five orders of magnitude higher than the TSDC spectrum current. There is no known process which could be responsible for releasing a few electrons and trapping them back at 39 K in the dark, under such experimental conditions, since there is not enough thermal energy to overcome the DX center thermal emission barrier.

The TSDC curve is usually fitted by a single curve, showing Debye behavior^{8,9} with a single relaxation time and activation energy. However, our data are better fitted by an asymmetric relaxation time distribution, the type of distribution first proposed by Havriliak and Negami.¹⁰

By using their equations, where the dimensionless parameters α and β evaluate the shape of distribution, the best fit to the TSDC experimental curve is obtained with a continuous distribution of activation energies. The average activation energy is about 0.108 eV and the constant relaxation time is $9 \times 10^{-13} \text{ s}$, with a peak at 37 K. It is shown in Fig. 2 by a dashed line. The other curve (dotted line) in Fig. 2 is another attempt to fit the experimental data with similar parameters, but by using a single relaxation time approach. With the same parameters as the average ones used for the distribution we get a curve with the same shape as the dotted line, but with a peak at 41 K. In order to obtain the dotted line of Fig. 2 we use a single constant relaxation time of $2.3 \times 10^{-13} \text{ s}$ with the same activation energy ($E_a = 0.108 \text{ eV}$) and get a peak at 39 K. As it can be seen the results are poor compared to the one obtained with the Havriliak–Negami relaxation time distribution approach. Fitting of experimental data with single relaxation time gives quite unreasonable parameters and must be discharged.

The TSDC spectrum reported here is strong evidence that the electron freezeout, which occurs when temperature is lowered, leads to charged localized states, since neutral impurities would not be responsible for an ionic dipole current. Such a current takes place when the biased dipoles are relaxing to more stable states, returning to their random distribution throughout the sample. It is very important to notice the dipole current magnitude because it as-

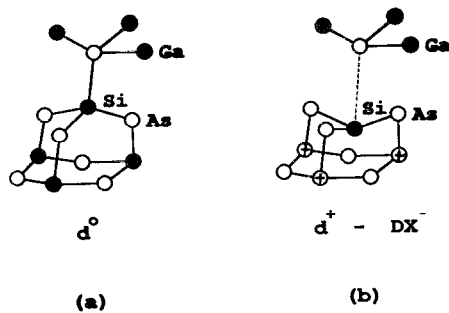


FIG. 3. (a) Si placed at a normal substitutional site in GaAs. (b) DX^- center [according to Chadi and Chang's model (see Ref. 1)] and d^+ center placed at first neighbor sites. The \oplus symbols represent the three equivalent positions for dipole relaxation.

sures that there is no electronic current, which means the strongly localized DX^- center does not release electrons to the conduction band. The question which arises from these data is whether or not d^+DX^- is close enough to be considered a dipole. In Chadi and Chang's model, shown in Fig. 3, there are three equivalent first neighbor positions for the d^+ state, which could be responsible for dipole reorientation. Although it is energetically very favorable to a d^+ center to be formed close to a DX^- center, the randomly distributed impurities are placed at their sites during growth and the probability that two impurities are first neighbors is negligibly small. However, the reported data shows that if the measured current comes indeed from the d^+DX^- pair there is a non-negligible fraction of d^+ centers located as close to the DX^- center as to be considered a perturbation in the DX^- screening potential.

Another strong point to support his hypothesis is the fact that no single relaxation time approach can be used to fit satisfactorily the experimental data. Good fit is only obtained with activation energy distribution. This is quite consistent with at least two major points.

- (1) The random distribution of d^+ center around the DX^- impurity (generation of a dipole length distribution).
- (2) The random distribution of Al atoms around both charge states should also contribute to the local field potential. The DX^- center has different energy levels depending on how many Al atoms are first neighbors.^{11,12}

Another striking feature of our results is that the average activation energy obtained is $E_a \cong 0.108$ eV which is the same value of the dominant donor level binding energy,¹³ which coincides with the DX^- center binding energy in this Al composition. It suggests another possibility for the dipole relaxation: The extra electron trapped at the DX^- center could be donated to a d^+ center and the interstitial donor atom migrates back to its normal substitutional site. This migration is responsible for the TSDC signal as recorded.

If the extra electrostatic energy gained by placing a d^+ center close to a DX^- center is of significant magnitude,⁶ there is a tendency for the electrons to be trapped at a Si donor located close to another Si donor. Moreover, the way the TSDC measurement itself is carried out adds an external factor to this picture: The temperature is decreased with an applied voltage to the sample. As the temperature gets lower, more electrons are trapped at DX^- centers. So the most stable situation corresponds to the creation of oriented dipole states. Even with such a favorable picture it is still hard to believe that all the measured dipole currents of Fig. 2 come from first neighbors relaxation alone, but a similar picture can be drawn for atoms located at farther apart sites. The idea will remain essentially as the one shown in Fig. 3.

Our conclusion is that a dipole relaxation current occurs in a Si-doped $Al_xGa_{1-x}As$ sample where the presence of DX^- centers is clearly evident. We interpret such a dipole current using the Chadi and Chang's DX^- model, and mainly the dipole scattering effects present in this alloy as predicted by O'Reilly. The most interesting data obtained from our calculation is that the activation energy for dipole relaxation is about 0.108 eV, approximately the same value as the binding energy of the dominant impurity state in $Al_xGa_{1-x}As$ (DX^- center). Further work is needed to clearly determine whether this dipole is the d^+DX^- pair or some other kind of dipole is present in this sample.

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