

## Brief Reports

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### Relaxation of persistent photoconductivity in $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$

J. Y. Lin, A. Dissanayake, G. Brown, and H. X. Jiang\*

*Department of Physics, Kansas State University, Cardwell Hall, Manhattan, Kansas 66506-2601*

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Stretched-exponential decay of persistent photoconductivity (PPC) has been observed in  $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$  in the temperature region  $T < 90$  K. The decay time constant and exponent have been measured at different temperatures. Experimental results show that the thermal activated capture of electrons at the  $DX$  centers becomes negligible at  $T < 40$  K. However, a transition from a thermally activated to a nearly-temperature-independent tunneling capture at a temperature on the order of 40 K has been observed for the first time for  $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ , which is consistent with the multiphonon-emission model for  $DX$  centers of large lattice relaxation.

There has been a considerable amount of experimental and theoretical effort directed towards the understanding of persistent photoconductivity (PPC) in  $\text{Al}_x\text{Ga}_{1-x}\text{As}$ . It is widely accepted that photoexcitation of electrons from deep-level traps,  $DX$  centers, which undergo a large lattice relaxation is the origin of PPC observed in these materials.<sup>1,2</sup> In this model, PPC has resulted because recapture of electrons by  $DX$  centers is prevented by a thermal barrier at low temperatures. The nature of the  $DX$  center is under intensive investigation. Recently, Chadi and Chang indicate that  $DX$  is a highly localized and negatively charged defect center ( $D^-$ ) which behaves as a negative- $U$  center.<sup>3,4</sup> However, recent magnetic studies of PPC in  $n$ -type  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  by Khachatryan *et al.*<sup>5</sup> show evidence that the  $DX$  center is a paramagnetic donor with one unpaired electron. There is evidence that the number of  $DX$  centers equals the number of doped donors,<sup>6</sup> and that  $DX$  centers behave as deep and shallow bistable states.<sup>7-10</sup> It has been suggested by several groups that  $DX$  centers are impurities linked to the  $L$  conduction band,<sup>11-14</sup> which undergo a shallow-deep instability due to intervalley mixing.<sup>14</sup>

The capture kinetics of electrons on  $DX$  centers has been studied by several groups,<sup>15-17</sup> however, they have not yet been well established. Systematic study of the electron-capture process at low temperatures ( $T < 50$  K) has never been achieved previously because of very long PPC decay time and the lack of knowledge of decay form. Obviously, the decay kinetics is very important in determining the origin for the slow relaxation of the photoexcited charge carriers. Recently, the random local potential fluctuations induced by compositional fluctuations in  $\text{Zn}_{0.3}\text{Cd}_{0.7}\text{Se}$  mixed crystals were found to be responsi-

ble for PPC, and a PPC-related phase transition was observed for the first time in which the stored charge carriers experience a transition from localized to delocalized states at a transition temperature.<sup>18,19</sup> The decay of PPC observed in this material follows a stretched-exponential function. Because of nonexponential decay of PPC observed previously in  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  materials, one usually cannot determine accurately the capture time constants  $\tau_C$  and the capture thermal barrier. Instead, one usually determines  $\tau_C$  from the  $\frac{1}{2}$ - or  $\frac{1}{3}$ -signal point.<sup>20,21</sup> It was suggested that the decay of PPC observed in  $\text{Al}_{0.35}\text{Ga}_{0.65}\text{As}$  can be described by variations of stretched-exponential forms.<sup>17</sup> The decay of PPC observed in Si-doped GaAs under pressure and in various other materials is found to follow a stretched-exponential function at low temperatures and a power-law form at higher temperatures.<sup>22</sup>

In this paper, the PPC relaxation process is investigated for  $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$  material throughout the temperature range of  $T \geq 10$  K. We found that in the temperature region  $T < 90$  K, the PPC relaxation follows stretched-exponential decay behavior. The characteristic decay time constant and the decay exponent have been determined systematically as functions of temperatures. In the temperature region  $40 < T < 90$  K, the decay rate of charge carriers shows an activated behavior. However, a transition from a thermally activated to a tunneling capture at a temperature of about 40 K and a nearly constant PPC relaxation rate at  $T \rightarrow 0$  K have been observed for the first time, which is consistent with the multiphonon-emission model for  $DX$  centers of large lattice relaxation.<sup>20,23</sup>

The samples used in this study were  $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$  epi-

taxy layers of  $2 \mu\text{m}$ , doped with  $3.3 \times 10^{17} \text{ cm}^{-3}$  Si, grown on a semi-insulating GaAs(100) substrate, and were supplied by Spire Corporation. Ohmic contacts 1 mm in diameter and about 7 mm apart were formed by indium alloying ( $400^\circ\text{C}$  20 min) on the layer surface. The sample was attached to a copper sample holder, which is inside a closed-cycle He refrigerator, with care taken to ensure good thermal contact yet electrical isolation. A temperature controller enabled us to stabilize the temperatures to about 0.1 K. A neon lamp was used along with appropriate filter so that a few lines within the region of  $6000 \text{ \AA} < \lambda_{\text{exc}} < 7050 \text{ \AA}$  dominated the output of the excitation source. Typical intensity used in the measurements was about  $10^{14}$  photons/ $\text{cm}^2\text{s}$ . PPC can be thermally quenched at about 150 K in this sample, and the intensity we used induces no photoconductivity (PC) at all temperatures. The data obtained at different conditions were taken in such a way that the system was always heated up to room temperature to convert the illuminated sample to its initial state and then cooled down again in darkness to the temperature of measurements. The equilibration time at each temperature is about 40 min.

In Fig. 1, we show PPC decay curves for three representative temperatures. The results are obtained by illuminating the sample by exactly the same amount of photon dose at different temperatures. All three curves are normalized to unity at  $t=0$ , the moment illumination is terminated, and the dark equilibrium current has been subtracted out. We see that the relaxation time of PPC becomes very long below 60 K and the PPC level is still about 98% of its initial value after 3000 s of decay at 60 K. However, the decay behavior in the temperature region ( $T < 90$  K) is very well described by stretched-exponential functions:

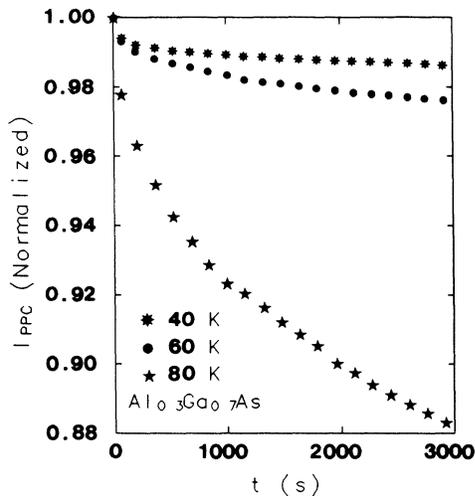


FIG. 1. PPC decay curves obtained at three representative temperatures. Each curve is normalized to unity at  $t=0$ , the moment of illumination is terminated, and dark current has been subtracted out. Illumination intensity used is about  $10^{14}$  photons/ $\text{cm}^2\text{s}$  and exposure time is 100 s.  $I(0)=8.1$  mA at  $T=80$  K,  $I(0)=7.8$  mA at  $T=60$  K, and  $I(0)=7.4$  mA at  $T=40$  K.

$$I_{\text{PPC}}(t) = I_{\text{PPC}}(0) \exp[-(t/\tau)^\beta], \quad 0 < \beta < 1, \quad (1)$$

where  $\tau$  is the characteristic decay time constant and  $\beta$  is the decay exponent. However, as the temperature increases to above 90 K, the decay deviates from stretched-exponential behavior. The stretched-exponential decay frequently describes the relaxation of a wide class of disordered systems toward equilibrium. Observation of stretched-exponential decay of PPC in  $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$  material suggests that random potential fluctuations induced by impurity distribution may play an important role in the process of carrier relaxation.<sup>22</sup> In Fig. 2, the experimental decay curves shown in Fig. 1 are replotted as  $\ln[\ln I_{\text{PPC}}(0) - \ln I_{\text{PPC}}(t)]$  versus  $\ln t$ ; linear behavior is evident.  $\tau$  and  $\beta$  as functions of temperature have been determined. Figures 1 and 2 show that the decay rate increases as temperature increases. Figure 3 presents a plot of  $\ln \tau$  versus  $1/T$ , which shows that  $\tau$  changes by 8 orders of magnitude as temperature varies from 80 to 10 K. To our knowledge, this is a first experimental attempt of systematically measuring the PPC decay time constant as a function of temperature in the entire region of interest.  $\tau$  as large as  $4.7 \times 10^{13}$  s at 10 K is observed for the first time here. From Fig. 3, the decay time constant monotonically increases with decreasing temperature. The important physics in Fig. 3 is twofold. First, the decay time constant  $\tau$  in the temperature region of  $T > 40$  K has an activated temperature dependence of the form

$$\tau = A \exp(E_C/kT), \quad (2)$$

which is well known.<sup>24</sup> Here,  $E_C$  denotes activation energy of the capture of electrons at the  $DX$  centers measured from the quasi-Fermi-level in the lowest conduction band.<sup>15</sup> In writing Eq. (2), we assume that the variation

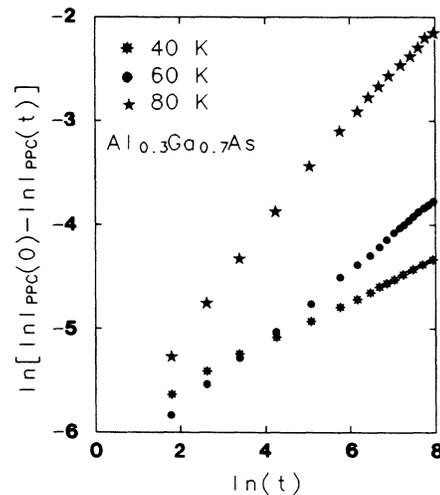


FIG. 2. Replot of Fig. 1 as  $\ln[\ln I_{\text{PPC}}(0) - \ln I_{\text{PPC}}(t)]$  vs  $\ln t$ . Linear behavior indicates that the PPC relaxation is well described by stretched-exponential function,  $I_{\text{PPC}}(t) = I_{\text{PPC}}(0) \exp[-(t/\tau)^\beta]$ , where  $\tau$  is the characteristic decay time constant and  $\beta$  is the decay exponent.

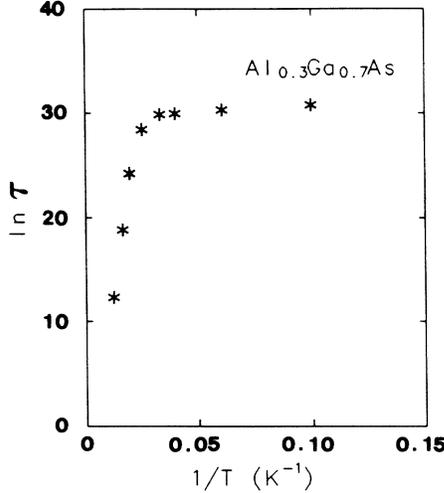


FIG. 3. Plot of  $\ln \tau$  vs  $1/T$ . Excitation intensity used is about  $10^{14}$  photons/cm<sup>2</sup>s and exposure time is 100 s.

of quasi-Fermi-level during the decay process at a constant temperature is negligible because the lifetime of the charge carriers in the conduction band is very long at low temperatures ( $T < 90$  K). However,  $E_C$  depends on the initial quasi-Fermi-level induced by photoexcitation, since  $\tau$  depends on the initial PPC buildup level at a constant temperature.<sup>25</sup>  $E_C$  calculated from the region of  $40 < T < 90$  K of the plot is about 160 meV. Second, Fig. 3 clearly shows two distinctive regions at the high-temperature ( $T > 40$  K) and the low-temperature ( $T < 40$  K) regions. Most importantly, a nonzero decay rate (finite lifetime) at temperature  $T=0$  can be obtained unambiguously by extrapolating experimental results to  $T=0$ . In the low-temperature region, the decay time constant depends weakly on temperature.

In the large lattice-relaxation model, PPC decay at higher temperatures is dominated by a thermal activation capture of electrons in the conduction band at the *DX* centers which are separated from the conduction band by a thermal barrier in the configurational space. At low temperatures, the capture has been predicted to be dominated by tunneling via multiphonon emission.<sup>20,23</sup> In this model description, the capture rate becomes constant as  $T \rightarrow 0$ . The appearance of the  $\ln \tau$  versus  $1/T$  plot in Fig. 3 shows precisely the behavior, which demonstrates that the thermal activated capture of electrons at the *DX* centers is negligible below about 40 K and the PPC decay is caused by the tunneling processes at  $T < 40$  K.

From multiphonon recombination theory,<sup>23</sup> the capture cross section of the electrons at low temperatures can be written as

$$\sigma_n = \sigma_{n0}(N+1)^p e^{-2NS}, \quad (3)$$

where  $p\hbar\omega$  is the net binding energy  $E_0$ ,  $\hbar\omega$  is phonon energy, and  $N = [\exp(\hbar\omega/kT) - 1]^{-1}$  is the phonon population.  $\sigma_{n0}$  is the cross section at temperature  $T=0$ .  $S$  is the electron-phonon coupling constant. From Eq. (3), we can write the electron relaxation time constant at low

temperatures as

$$\tau = \tau_0(N+1)^{-p} e^{2NS}, \quad (4)$$

where

$$\tau_0 = \frac{1}{\sigma_{n0} n \langle V \rangle}. \quad (5)$$

Here,  $n$  is the free-electron concentration and  $\langle V \rangle$  is their average velocity. An important point is that  $\langle V \rangle$  is not zero even at  $T=0$  K because of the Fermi distribution.

Equation (4) has been compared to the experimental results of Fig. 3 at low temperatures ( $T < 40$  K) to estimate tunneling parameters. We found that phonon energy  $\hbar\omega$  is about 3.7 meV, the electron-phonon coupling constant  $S \approx 9$  and  $\tau_0$  is about  $5 \times 10^{13}$  s. The net binding energy  $E_0$  has been taken to be  $\sim 100$  meV.<sup>1,2</sup> The important result we obtained here is that the multiphonon tunneling between the conduction band and the *DX* centers is assisted by *acoustic phonons* at low temperatures, since the average phonon energy we obtained is about 3.7 meV. The coupling between electrons and phonons seems to be very strong here.

From Eq. (5), we can also obtain the capture cross section  $\sigma_{n0}$  of the free electrons. At  $T=0$  K,

$$\begin{aligned} \langle V \rangle &\approx \langle V^2 \rangle^{1/2} \\ &= \left\langle \frac{2\varepsilon}{m^*} \right\rangle^{1/2} \\ &= \left[ \frac{3}{5} \varepsilon_f \frac{2}{m^*} \right]^{1/2} \\ &= \left[ \frac{6}{5m^*} \varepsilon_f \right]^{1/2} \end{aligned} \quad (6)$$

with  $\varepsilon_F = (\hbar^2/2m^*)(6\pi^2n/g)^{2/3}$  being the Fermi energy. Thus, we have  $\sigma_n$  at  $T=0$  K as

$$\sigma_{n0} = \frac{1}{\tau_0 n \langle V \rangle} = \frac{1}{n \tau_0} \sqrt{\frac{5}{3}} \frac{m^*}{\hbar(3\pi^2n)^{1/3}}, \quad (7)$$

where the  $g$  factor has been taken as 2. We take the electron effective mass  $m^*$  for  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  as  $m^* = (0.067 + 0.083x)m_0$ .<sup>26</sup> From Eq. (7) we obtain

$$\sigma_{n0} = 4 \times 10^{-16} \frac{1}{[n(\text{cm}^{-3})]^{4/3}} \text{cm}^2$$

with the units of  $n$  being in  $\text{cm}^{-3}$ . The free-charge-carrier concentration is on the order of  $10^{12} \text{cm}^{-3}$ , which gives  $\sigma_0 \approx 4 \times 10^{-32} \text{cm}^2$ . The cause of the very long lifetime of PPC at low temperatures is this small capture cross section.

The decay exponent  $\beta$  as a function of temperature is plotted in Fig. 4.  $\beta$  is almost a constant value ( $\approx 0.20$ ) in the temperature region  $T < 40$  K but varies in the thermally activated temperature region, increasing from about 0.21 to 0.47 as temperature increases from 40 to 80 K. The physical significance of  $\beta$  is not clear at this stage. However, the temperature dependence of  $\beta$  gives

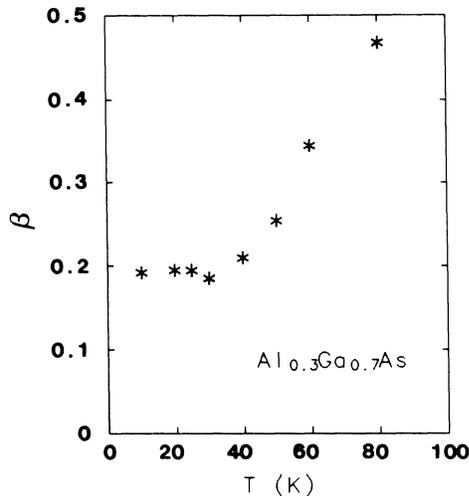


FIG. 4. Plot of decay exponent  $\beta$  vs temperature  $T$ .

more supporting evidence that two different decay mechanisms dominate in the low- and high-temperature regions.

Although the decay of charge carriers in PPC mode follows stretched-exponential form for both II-VI and III-V mixed semiconductors, the decay mechanisms are

completely different. For  $\text{Al}_x\text{Ga}_{1-x}\text{As}$ , the decay of the conduction electrons is caused by the process of thermal activation over the capture barrier  $E_C$  in the high-temperature region and is due to tunneling via multiphonon emission in configurational space in the low-temperature region. For II-VI mixed semiconductors,<sup>18,19,22</sup> the decay of the electrons is caused by wavefunction overlap between electrons and holes in real space, where electrons are localized (delocalized) at low (high) temperatures, while holes are almost always localized because of their heavier masses.

In conclusion, PPC relaxation processes in  $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$  material have been investigated in the temperature range  $T \geq 10$  K. We find that the PCC relaxation follows stretched-exponential behavior in the entire temperature region  $T < 90$  K. A constant PPC decay rate has been observed at  $T \rightarrow 0$  K, consistent with the results expected from the large lattice-relaxation model. A transition from a thermally activated decay to a multiphonon assisted tunneling at a temperature of about 40 K has been observed. The origin for the decay behavior remains to be investigated.

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\*To whom all correspondence should be addressed.

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