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Repository Citation

Fang, Z., & Look, D. C. (1992). Photoquenching of Hopping Conduction in Low-Temperature-Grown Molecular-Beam-Epitaxial GaAs. *Applied Physics Letters*, 61 (12), 1438-1440.
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Photoquenching of hopping conduction in low-temperature-grown molecular-beam-epitaxial GaAs

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(Received 11 May 1992; accepted for publication 7 July 1992)

We have observed IR photoquenching of the hopping conduction in GaAs samples grown by molecular beam epitaxy at the low temperature of 250 °C and annealed at temperatures from 300 to 600 °C. A key element in the success of this study is removal of the layers from their substrates. The hopping conduction recovers at about 140 K, with a thermal activation energy of about 0.3 eV.

Recently the growth of GaAs by molecular beam epitaxy (MBE) at low substrate temperatures (200–400 °C), often referred to as LT-MBE GaAs, has attracted considerable interest, because of its technological importance and unique compositional, electrical, and optical properties.¹ The properties of LT-MBE GaAs, which are largely controlled by an As_{Ga}-related defect and other point defects in the material, are closely dependent on the growth and post-growth anneal conditions. For example, we have shown that the unusual conductivity and Hall-effect properties result from two types of carriers, those hopping between the As_{Ga} centers and those excited to the conduction band from these same deep centers.² In this letter we report the photoquenching of the hopping conduction and its thermal recovery as a function of the post-growth anneal temperature (T_A). These data prove that As_{Ga} in its metastable state cannot participate in the hopping phenomenon.

The LT-MBE GaAs samples used in the study were grown at 250 °C in a modified Varian 360 system under normal As-stabilized conditions. Post-growth anneals were performed on 6×6 mm² samples at temperatures from 300 to 600 °C for 10 min in an inert gas with a GaAs wafer on top. To avoid any interference from the semi-insulating (SI) GaAs substrate, the layers were separated from their respective substrates and mounted on glass pieces by a technique described elsewhere.³ Indium contacts were applied on the four corners of each layer with a soldering iron, but were not annealed. Either pair of the diagonal contacts was used to measure the dark current (I_{dark}), the photocurrent (I_{ph}), and the thermally stimulated current (I_{tsc}). In the measurements the samples were always fast cooled from 380 to 82 K in the dark. The I_{ph} 's were measured immediately after IR light illumination at 82 K. The IR light ($h\nu \leq 1.12$ eV) was provided by a tungsten lamp (8 W) through a Si-wafer filter. After 5 min illumination at 82 K, the thermal scan for the TSC spectrum ($82 < T < 300$ K with a heating rate of 0.3 K/s), was always started 30 s after the termination of the illumination. The I_{dark} 's were recorded during slow warming (0.3 K/s) from 82 to 380 K without infrared (IR) light illumination at 82 K. All the currents were measured under a bias of 20 V using an electrometer (Keithley 617). For comparison, a liquid-encapsulated Czochralski (LEC) SI GaAs sample grown from an As-rich melt, with a thickness of 700 μm , was also used in the study.

The photocurrent responses after IR light illumination at 82 K are shown in Fig. 1, with the I_{dark} at 82 K indicated for each LT-MBE GaAs sample. First of all, it is seen that the net I_{ph} at the beginning of the IR light illumination for each of the LT-MBE GaAs samples is much lower than that of the LEC SI GaAs sample and is further reduced by increasing the annealing temperature T_A . Secondly, IR light obviously causes quenching of the photocurrent in most of the samples (at least initially), but not in the LT-MBE GaAs sample with $T_A = 300$ °C. Furthermore, it is seen, for the first time, that the quenched I_{ph} becomes much lower than the original I_{dark} for the sample with $T_A = 600$ °C, and the same phenomenon is also found for samples with $T_A = 500$ and 550 °C (not shown). Both the dark currents and the TSC spectra are pictured in an Arrhenius plot, shown in Fig. 2. From the figure, we see that: (1) at $T < 200$ K, the I_{dark} 's of the LT-MBE GaAs samples, grown at 250 °C and annealed at T_A 's from 300 to 600 °C, are dominated by hopping conduction, as evidenced by the shallow slope;² (2) the slope, which is related to the As_{Ga} concentrations decreases with increasing T_A , suggesting that [As_{Ga}] is decreasing; (3) however, at $T > 300$ K, a band conduction with an activation energy of about 0.64 eV can be observed for the sample with $T_A = 600$ °C; and (4) in contrast to the LT-MBE samples, the LEC sample simply shows band conduction over the whole range, with an activation energy of 0.76 eV, i.e., controlled by the usual EL2 center. Now consider the TSC spectra in Fig. 2. For the LEC SI GaAs sample, a standard IR-quenched TSC spectrum with a spectral feature II, containing large T_0^* , T_2^* , and T_1 ,⁴ can be observed. However, for the LT-MBE GaAs samples, no meaningful TSC peaks can be observed, except for the sample with $T_A = 300$ °C, which would show a strong TSC peak, T_5 , at 136 K if $I_{\text{tsc}} - I_{\text{dark}}$ were plotted. What we observe for the samples with T_A 's of 400 and 600 °C are I_{tsc} 's which are less than the respective I_{dark} 's at low temperatures, but which are restored to the original I_{dark} 's at a temperature around 140 K. In other words, the IR illumination actually quenches I_{dark} , a very unusual phenomenon.

As an example, the thermal restorations of the photoquenched I_{dark} 's, as a function of the illumination time, are shown in Fig. 3 for the sample annealed at 550 °C. From Figs. 2 and 3, it can be seen that no matter how high the T_A (between 400 and 600 °C) and how long the illu-

FIG. 1. IR photocurrent responses at 82 K for the LT-MBE GaAs and LEC SI GaAs samples, showing IR quenching of I_{ph} , except for the LT-MBE GaAs sample annealed at 300 °C.

mination time (between 1 s and 10 min), the thermal restoration temperatures for the photoquenched I_{dark} 's are always about 140 K. Through changing the heating rate from 0.05 to 0.4 K/s, a thermal activation energy of 0.3 eV for the thermal restoration process can be deduced, which is in agreement with the values of 0.25–0.30 eV obtained by IR absorption measurements for the thermal regeneration rate of EL2 from the metastable state to the normal state in undoped SI GaAs.⁵ More definitive studies of the recovery kinetics are now underway.

FIG. 2. Dark current and TSC spectra, showing IR quenching of hopping conduction and its thermal recovery for the LT-MBE GaAs samples annealed at $400 < T_A < 600$ °C and the transition of the conduction mechanism from hopping conduction to band conduction with increasing T_A . A bulk LEC SI GaAs sample was used as a reference.

FIG. 3. IR quenching of hopping conduction as a function of the illumination time on an LT-MBE GaAs sample annealed at 550 °C, showing a decrease of the activation energy for the hopping conduction due to the decrease of $[As_{Ga}]$.

It may be noted that an activation energy of 1.7 ± 0.3 eV was measured in the transmission experiments on LT-MBE GaAs by Bliss *et al.*⁶ and this energy was assigned to V_{Ga} diffusion. A very similar activation energy (~ 1.6 eV) is observed for the 250–300 °C annealing stage of 1 MeV-electron damage in GaAs (presumably V_{As} motion but possibly including V_{Ga} motion).⁷ Indeed, V_{Ga} motion may mediate the movement of As_{Ga} to form As precipitates, which are known to exist in LT-MBE GaAs annealed above 500 °C.⁸ It is interesting that the sample annealed at only 300 °C shows a strong TSC peak (a “normal” TSC peak), T_5 , which we have earlier associated with As-rich conditions in SI LEC GaAs.⁹ Thus, T_5 could be related to V_{Ga} and as observed, it would be expected to decrease in intensity as T_A is raised. However, T_5 could also be related to As_{Ga} , which similarly decreases in concentration at higher T_A . Evidently, the dominant form of As_{Ga} in the 300 °C-annealed sample is nearly unquenchable, as was found earlier in transmission experiments.¹⁰ Whether or not a different form of As_{Ga} exists in the higher-temperature-annealed samples, or there simply is a different environment which influences the quenchability, is not clear at this time.

The main point of the present work is to explain the quenching of the hopping conduction. At the recovery temperature of about 140 K, the hopping will probably have both nearest-neighbor ($\sigma_{nn} \sim \exp(-\epsilon_3/kT)$) and long-range $\sigma_{lr} \sim \exp(-T_0^{1/4}/T^{1/4})$ components. However, both ϵ_3 and T_0 are related to N_D , the deep-donor (As_{Ga}) concentration ($\epsilon_3 \sim N_D^{1/3}$ and $T_0^{1/4} \sim N_D^{1/4}$). Furthermore, σ_{nn} contains another term, $\exp(-\gamma R/a)$, where $\gamma \simeq 1.8$, $R \sim N_D^{-1/3}$ is the distance between donors, and a is the extent of the donor wave function. Thus, in both cases, σ (and therefore I_{dark}) will decrease as N_D (or $[As_{Ga}]$) decreases. As we anneal, the As_{Ga} defects literally disappear, but as we photoquench, they simply change to their metastable states. Note that the slopes (related to ϵ_3 or $T_0^{1/4}$) in Fig. 3 also decrease, as they should. (Observe that I_{tsc} is on a log scale, which is why the slope changes are much smaller for the longer illumination times.) Then, as the

measurement temperature is raised to 140 K, the As_{Ga} centers return to their ground states, and can once more participate in the hopping process. A rather important and controversial point is whether or not the metastable As_{Ga} centers are electrically active. From the point of view of hopping conduction, we can categorically state that they are not. That is, their bound electrons (assuming that the metastable centers are neutral) cannot hop to an empty (unoccupied) neighbor. It is then not obvious that metastable As_{Ga} centers can compensate an acceptor in normal GaAs if thermal energy alone is involved.

In summary, we have observed the IR photoquenching of the hopping conduction in LT-MBE GaAs. The conduction recovers at about 140 K, with a thermal activation energy of about 0.3 eV.

Note: When this manuscript was nearing completion, a paper by Kuriyama, Yokoyama, and Taniguchi on the quenching of hopping conduction in neutron-irradiated GaAs appeared in the literature.¹¹ However, they did not deduce a recovery activation energy.

D.C.L. was supported by USAF Contract F33615-86-C-1765 and Z.Q.F. by ONR Contract N00014-90-J-11847.

The LT-MBE GaAs samples were grown at the Solid State Electronics Directorate, Wright Laboratory, Wright Patterson AFB, OH by C. E. Stutz and E. N. Taylor. Thanks are also due to D. C. Walters and J. T. Prichard for the sample annealing and the substrate removal, respectively.

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