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Photoquenching of hopping conduction in low-temperature-grown molecular-beam-epitaxial GaAs

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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. The properties of LT-MBE GaAs, which are largely controlled by an AsGa 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 AsGa centers and those excited to the conduction band from these same deep centers. In this letter we report the growth anneal conditions. For example, we have shown that AsGa 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. 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\(_\text{dark}\)), the photocurrent (I\(_\text{ph}\)), and the thermally stimulated current (I\(_\text{tr}\)). In the measurements the samples were always fast cooled from 380 to 82 K in the dark. The I\(_\text{ph}\)'s were measured immediately after IR light illumination at 82 K. The IR light (hν<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\(_\text{dark}\)'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\(_\text{dark}\) at 82 K indicated for each LT-MBE GaAs sample. First of all, it is seen that the net I\(_\text{ph}\) 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 \(T_A=300^\circ\text{C}\). Furthermore, it is seen, for the first time, that the quenched I\(_\text{ph}\) becomes much lower than the original I\(_\text{dark}\) for the sample with \(T_A=600^\circ\text{C}\), 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\(_\text{dark}\)'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) the slope, which is related to the AsGa concentrations decreases with increasing \(T_A\), suggesting that [AsGa] 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 \(T_A=600^\circ\text{C}\); 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 \(T_d\), \(T_d\), and \(T_d\), can be observed. However, for the LT-MBE GaAs samples, no meaningful TSC peaks can be observed, except for the sample with \(T_A=300^\circ\text{C}\), which would show a strong TSC peak, \(T_d\), at 136 K if \(I_{\text{nc}}-I_{\text{dark}}\) were plotted. What we observe for the samples with \(T_A\)'s of 400 and 600 °C are \(I_{\text{nc}}\)'s which are less than the respective \(I_{\text{dark}}\)'s at low temperatures, but which are restored to the original \(I_{\text{dark}}\) at a temperature around 140 K. In other words, the IR illumination actually quenches \(I_{\text{dark}}\), a very unusual phenomenon.

As an example, the thermal restorations of the photoquenched \(I_{\text{dark}}\)'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 illumination time, the hopping conduction recovers at about 140 K, with a thermal activation energy of about 0.3 eV.
time (between 1 s and 10 min), the thermal restoration temperatures for the photoquenched $I_{\text{dark}}$'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. More definitive studies of the recovery kinetics are now underway.

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. and this energy was assigned to $V_{\text{Ga}}$ 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 $V_{\text{As}}$ motion but possibly including $V_{\text{Ga}}$ motion). Indeed, $V_{\text{Ga}}$ motion may mediate the movement of $A_{\text{Ga}}$ to form $A_{\text{Ga}}$ precipitates, which are known to exist in LT-MBE GaAs annealed above 500°C. It is interesting that the sample annealed at only 300°C shows a strong TSC peak (a "normal" TSC peak), $T_{\text{f}}$, which we have earlier associated with $A_{\text{Ga}}$ conditions in SI LEC GaAs. Thus, $T_{\text{f}}$ could be related to $V_{\text{Ga}}$ as observed, it would be expected to decrease in intensity as $T_{\text{f}}$ is raised. However, $T_{\text{f}}$ could also be related to $A_{\text{Ga}}$, which similarly decreases in concentration at higher $T_{\text{f}}$. Evidently, the dominant form of $A_{\text{Ga}}$ in the 300°C-annealed sample is nearly unquenchable, as was found earlier in transmission experiments. Whether or not a different form of $A_{\text{Ga}}$ 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 ($\sigma_{n,n} \sim \exp(-\varepsilon_{3}/kT)$) and long-range ($\sigma_{l,n} \sim \exp(-T_{0}/T_{l,n})$) components. However, both $\varepsilon_{3}$ and $T_{0}$ are related to $N_{D}$, the deep-donor ($A_{\text{Ga}}$) concentration ($\varepsilon_{3} \sim N_{D}^{1/3}$ and $T_{0}^{1/4} \sim N_{D}^{-2}$). Furthermore, $\sigma_{n,n}$ contains another term, $\exp(-\gamma R/a)$, where $\gamma = 1.8$, $R \sim N_{D}^{-1/3}$ is the distance between donors, and $a$ is the extent of the donor wave function. Thus, in both cases, $\sigma$ (and therefore $I_{\text{dark}}$) will decrease as $N_{D}$ (or [As$_{Ga}$]) decreases. As we anneal, the $A_{\text{Ga}}$ defects literally disappear, but as we photoquench, they simply change to their metastable states. Note that the slopes (related to $\varepsilon_{3}$ or $T_{0}^{1/4}$) in Fig. 3 also decrease, as they should. (Observe that $I_{\text{sc}}$ 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 AsGa 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 AsGa 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 AsGa 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. However, they did not deduce a recovery activation energy.

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