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Quenching phenomenon of hopping conduction in neutron-transmutation-doped semi-insulating GaAs

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The quenching phenomenon in tunneling-assisted hopping conduction below 125 K has been observed in neutron-transmutation-doped semi-insulating GaAs irradiated with a fast-neutron fluence of 3.7×10^{18} cm^{-2}. No quenchable component is observed in as-irradiated GaAs, while the *EL*2 or *EL*2-like defects are generated by annealing above 250 °C and activated as the quenchable component in the hopping conduction. The *EL*2 (or *EL*2-like) defects are likely to be formed by the interaction of the As antisite and a mobile defect such as the As interstitial and divacancy (As vacancy+Ga vacancy). According to the tunneling-assisted hopping model, the concentration of the quenchable component generated is larger than that of the native *EL*2 defect decomposed in irradiated GaAs.

Recently, Manasreh and Fischer[1] have reported that the high-dose neutron irradiation decomposes completely the atomic structure of the midgap electron trap (*EL*2 defect) in semi-insulating GaAs, while the *EL*2-like defect is produced after annealing for 6 min at 600 °C. It is well known that the *EL*2 defect is transferred from normal to metastable states by photoillumination in the range from 1.0 to 1.3 eV below a temperature ranging from 110 to 140 K (Refs. 2-4). There are two models for the atomic components of *EL*2: an arsenic antisite ($\mathrm{As_{Ga}}$)–arsenic interstitial (As_i) model proposed by von Bardeleben *et al.*,[5] and the $\mathrm{As_{Ga}}$-Ga vacancy (V_{Ga})–As vacancy (V_{As}) model proposed by Wager and Van Vechten.[6] Other workers[7] besides Wager and Van Vechten have independently proposed that the *EL*2 defect consists of a divacancy ($V_{\mathrm{As}}+V_{\mathrm{Ga}}$) with $\mathrm{As_{Ga}}$. To survey the correlation between the *EL*2 defect and the isolated $\mathrm{As_{Ga}}$ defect, infrared absorption[1,8] and electron-spin resonance[9,10] (ESR) experiments have been carried out using neutron-irradiated samples. These experiments indicated that the neutron-induced isolated $\mathrm{As_{Ga}}$ does not exhibit metastability. These results cast doubt on the identification of *EL*2 with the isolated $\mathrm{As_{Ga}}$ defect. We have reported[11] the irregularity of the tunneling-assisted hopping conduction around 120 K under photoexcitation in neutron-transmutation-doped (NTD) GaAs annealed at 250 and 400 °C. Our observation suggests that the quenchable defects exist in the neutron-irradiated GaAs.

In this Rapid Communication, we report the photoquenching phenomenon of the tunneling-assisted hopping conduction induced in NTD GaAs. We also discuss the origin of the quenchable component through the annealing behavior.

The crystals used in this study were undoped semi-insulating GaAs ($\rho\simeq2\times10^{7}$ Ω cm) grown by the liquid-encapsulated Czochralski technique. The as-grown crystal contains the *EL*2 defect of $\sim1.2\times10^{16}$ cm^{-3}. Neutron irradiations were performed using the Kyoto University Reactor (KUR), which is a light-water–moderated research reactor. Samples were irradiated with fast (fluence equals 3.7×10^{18} cm^{-2}) and thermal neutrons (1.3×10^{19} cm^{-2}) at fluxes of 1.4×10^{13} and 4.7×10^{13} cm^{-2}sec^{-1} for each neutron. The detail situation of the neutron irradiation has been described in our previous paper.[11,12] The annealing of irradiated samples was performed by placing the two GaAs wafers in N_2 flow for 30 min at a temperature ranging from 250 to 850 °C. To eliminate the decomposed layer by incongruent evaporation of arsenic, a few μm of material were removed from the surface by chemical etching after each annealing stage.[11] The Ohmic contacts with a gap of 1 mm were fabricated by sintering Au-Ge-Ni alloy for 1 min at 480 °C. Conductance measurements were carried out at a temperature ranging from 80 to 300 K in dark before and after the illumination of the quenching light. The photoquenching was performed using the light-emitting diodes (LED) with various peak wavelengths of 850 nm (photon energy and full width at half maximum; 1.46 eV and 40 nm), 890 nm (1.39 eV and 80 nm), and 940 nm (1.32 eV and 40 nm). The intensity of the light was 6, 10, and 12 mW for each LED.

Figure 1 shows the resistivity as a function of annealing temperature for NTD GaAs irradiated with fast-neutron fluences of 3.7×10^{18} and $\leq10^{14}$ cm^{-2}, which has been described in our previous paper.[12] The resistivity of the unannealed sample is reduced from 2×10^{7} to 8×10^{5} Ω cm by neutron irradiation. This reduction is based on the tunneling-assisted hopping conduction[13] between defect clusters involving the isolated $\mathrm{As_{Ga}}$ defect. This defect produced by neutron irradiation was estimated to be 3.3×10^{18} cm^{-3} by the ESR measurements at 77 K.[11] The hopping conduction observed here obeys the relationship[14]

$$\sigma=\sigma_0\exp(-b/T^{1/4})\,, \qquad (1)$$

where b is a constant and correlated with the defect concentration associated with the hopping conduction. The hopping conduction was observed at an annealing temperature up to 500 °C. The drastic decrease in resistivity around 600 °C is corresponding to the annihilation of the isolated $\mathrm{As_{Ga}}$ defects. This behavior is in good agreement with ESR measurements.[15,16] The decrease in resistivity

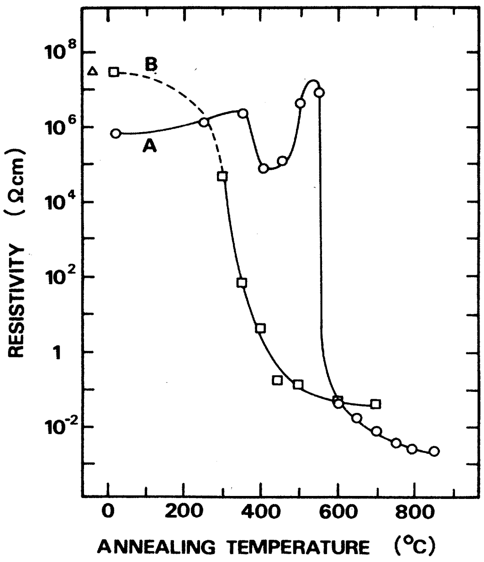

FIG. 1. Room-temperature resistivity as a function of annealing temperature for NTD GaAs irradiated with two different neutron fluences: Curve A for fast-neutron fluence (Φ_f) of 3.7×10^{18} cm^{-2} and thermal-neutron fluence ($\Phi_{\rm th}$) of 1.3×10^{19} cm^{-2}, and curve B for $\Phi_f < 10^{14}$ cm^{-2} and $\Phi_{\rm th}=2.2\times10^{17}$ cm^{-2}. The triangle represents the starting sample.

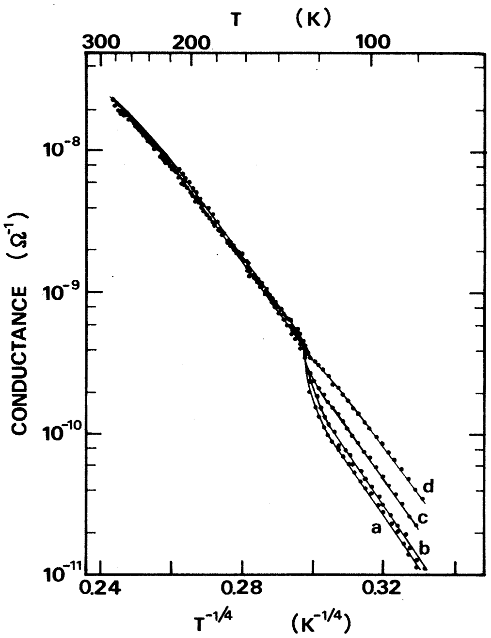

FIG. 2. Temperature dependence of conductance for the samples preilluminated with lights of $\lambda=940$ nm (curve a), 890 nm (curve b), and 850 nm (curve c). Curve d represents the conductance without preillumination. Samples presented here were annealed at 250 °C for 30 min.

around 400 °C is associated with the activation of the NTD impurities. The activated impurities provide the electrons to the defect levels originating the hopping conduction because in the irradiation with a small amount of fast neutrons the transmuted impurities start to activate around 300 °C (see Fig. 1). Consequently the hopping conduction is enhanced by the increase in electron density in defect levels.

Figure 2 shows the temperature dependence of conductance for NTD samples preilluminated with lights of $\lambda=850$, 890, and 940 nm for 20 min. The samples used here were annealed at 250 °C for 30 min. The conductance without preillumination shows obviously the hopping conduction, while the preillumination with lights of $\lambda=890$ and 940 nm for 20 min induces the abrupt change in conductance at 125 K and the hopping conduction remains below 125 K. This behavior suggests that a part of the defects associated with the hopping conduction are quenched by preillumination. Furthermore, for the preillumination with a light of 1.46 eV (850 nm) for 20 min, the noticeable photoquenching of the conductance was observed. This photon energy is less than the band gap ($\sim$1.51 eV at 85 K) at a temperature during the illumination. The variation of quenching behavior for various wavelengths suggests that the origin of the quenchable component exists in the forbidden band. The slight deviation from $\exp(-b/T^{-1/4})$ above $\sim$200 K must be originating from the electrons in the conduction band excited thermally from the defect levels associated with the hopping conduction.

Using slope b of the conductance curve in Fig. 2, we estimated the defect concentration ($N_{\rm HC}$) associated with the hopping conduction. A constant b in Eq. (1) is correlated with $N_{\rm HC}$ as follows:[13,14]

$$b=A(\alpha R)^{3/4}(W/k)^{1/4}, \tag{2a}$$

$$N_{\rm HC}=R^{-3}\times 3/(4\pi), \tag{2b}$$

where A is a constant, 2.95, α^{-1} is the tunneling length $[\alpha=(2m^*E_g/2)^{1/2}/\hbar]$, W is the width of the defect levels, R is the distance between the hopping sites, and the other terms have the usual meaning. W is estimated to be 0.083 eV from Arrehenius plot of the conductance without preillumination. For the preillumination with a light of $\lambda=940$ nm for 20 min, the slopes of quenched and recovered conductances were estimated to be 85 and 76, respectively. These slopes coincide with those of the illumination with other wavelengths within the experimental errors. These values give the defect concentration of 9.6×10^{17} and 1.4×10^{18} cm^{-3} for quenching and recovery, respectively. This indicates that the concentration of the quenchable defects is 4.4×10^{17} cm^{-3}.

We discuss the origin of the quenchable component observed in the hopping conduction. The $EL2$ and isolated $\rm As_{Ga}$ defects have been known as the defects with the metastability induced by photoillumination. The recent ESR measurements[9,10] indicate that the isolated $\rm As_{Ga}$ defect introduced by neutron irradiation does not show the photoquenching phenomenon, while the native $\rm As_{Ga}$ defect in as-grown crystal has been identified as the quenchable component. The concentration of the native $\rm As_{Ga}$ defect is less than 2×10^{17} cm^{-3} in our ESR measurements at 77 K.[11] Therefore, the native $\rm As_{Ga}$ defects are not identified as a main origin for the quenching of the hopping conduction, since the quenchable defect is 4.4×10^{17} cm^{-3}, as discussed earlier.

A possible alternative origin is the *EL*2 defects. However, the quenchable component is not observed in as-irradiated NTD GaAs (see Fig. 3). This result supports the suggestion[1] that the high-dose neutron irradiation decomposes completely the atomic structure of the *EL*2 defect. Figure 3 shows the conductances recorded by preillumination with a light of $\lambda = 940$ nm for as-irradiated and annealed samples. The sample annealed at 450 °C as well as annealing at 250 °C shows clearly the quenching behavior with an irregularity around 125 K. Therefore, the appearance of the quenchable component may originate from the formation of the *EL*2 (or *EL*2-like) defects due to the interaction of $\mathrm{As_{Ga}}$ and any point defect such as $\mathrm{V_{Ga}}$ and As_i induced by neutron irradiation. The artificially induced isolated $\mathrm{As_{Ga}}$ defect cannot be annihilated by annealing below 550 °C.[11] The Ga vacancy presumably included in the defect cluster is annealed out at around 450 °C,[15,16] and Goltzene, Meyer, and Schwab[15] have proposed the $\mathrm{As_{Ga}}$-divacancy model.[7] The divacancies consisting of $\mathrm{V_{Ga}}$ and $\mathrm{V_{As}}$ may be more mobile than single Ga vacancies in GaAs crystal because they can migrate by single-atom nearest-neighbor hopping through the divacancy cavity; they can thereby avoid both hopping the second-nearest-neighbor distance and the necessity to make several antisite defects. Another mobile defect at 250 °C is identified with As_i.[17] Therefore, the *EL*2 (or *EL*2-like) defects are likely to be generated by the interaction between the isolated $\mathrm{As_{Ga}}$ and either As_i or divacancy through the annealing process. We cannot evaluate whether or not the thermally created *EL*2 defect is identical with the native *EL*2 defect at present. The *EL*2-like defect mentioned above may be correlated with the "*DL*2" defect proposed by Manasreh and Fischer,[1] but *DL*2 defect is generated by annealing for 6 min at 600 °C.

In conclusion, we found the quenching phenomenon of the tunneling-assisted-hopping conduction in neutron-irradiated GaAs annealed above 250 °C, while this phenomenon was not observed for the as-irradiated sample. It was suggested that the *EL*2 (or *EL*2-like) defect is generated by the interaction of the isolated $\mathrm{As_{Ga}}$ defect and the mobile defect such as As_i and divacancy ($\mathrm{V_{As}} + \mathrm{V_{Ga}}$) at annealing temperatures above 250 °C. We cannot confirm whether As_i is more mobile than divacancy at lower temperature at present. However, we believe that our observation provides a new approach for the atomic model of *EL*2.

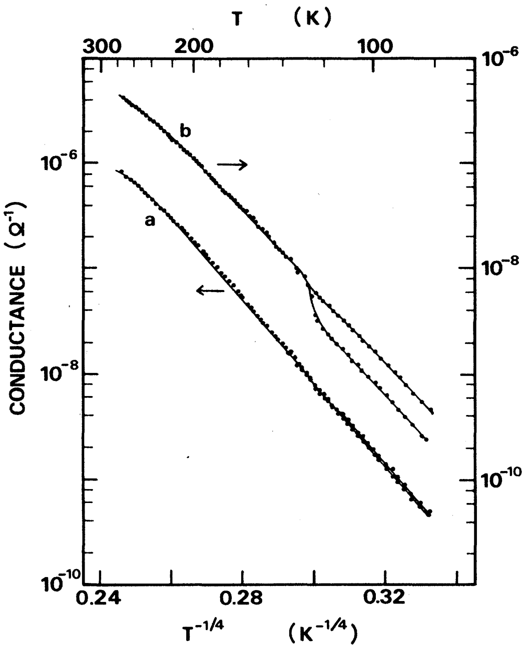

FIG. 3. Temperature dependence of conductances for (curve *a*) as-irradiation and (curve *b*) annealing at 450 °C for 30 min before and after illumination with a light of $\lambda = 940$ nm for 20 min. See Fig. 2 for annealing at 250 °C. The annealed sample shows the decrease in conductance below 125 K, indicating the photoquenching.

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[1] M. O. Manasreh and D. W. Fischer, Phys. Rev. B **39**, 3239 (1989).

[2] G. Vincent, D. Bois, and A. Chantre, J. Appl. Phys. **53**, 3643 (1982).

[3] G. M. Martin, Appl. Phys. Lett. **39**, 747 (1981).

[4] T. Hariu, T. Satoh, H. Komori, and K. Matsushita, J. Appl. Phys. **61**, 1068 (1987).

[5] H. J. von Bardeleben, D. Stievenard, D. Deresmes, A. Huber, and J. C. Bourgoin, Phys. Rev. B **34**, 7192 (1986); H. J. von Bardeleben, J. C. Bourgoin, and D. Stievenard, *ibid.* **39**, 1966 (1989).

[6] J. F. Wager and J. A. Van Vechten, Phys. Rev. B **35**, 2330 (1987); **39**, 1967 (1989).

[7] Zou Yuanxi, Wang Guangyu, A. Benakki, A. Goltzene, and C. Schwab, Phys. Rev. B **38**, 10953 (1988).

[8] M. O. Manasreh, D. W. Fischer, and B. C. Covington, Phys. Rev. B **37**, 6567 (1988).

[9] M. O. Manasreh, P. F. McDonald, S. A. Kivlighn, J. T. Minton, and B. C. Covington, Solid State Commun. **65**, 1267 (1988).

[10] A. Goltzene, B. Meyer, and C. Schwab, Appl. Phys. Lett. **54**, 907 (1989).

[11] M. Satoh, H. Kawahara, K. Kuriyama, T. Kawakubo, K. Yoneda, and I. Kimura, J. Appl. Phys. **63**, 1099 (1988).

[12] M. Satoh, K. Kuriyama, M. Yahagi, K. Iwamura, C. Kim, T. Kawakubo, K. Yoneda, and I. Kimura, Appl. Phys. Lett. **50**, 580 (1987); M. Satoh, K. Kuriyama, and Y. Makita, J. Appl. Phys. **65**, 2248 (1989).

[13] R. Coates and E. W. J. Mitchell, J. Phys. C **5**, L113 (1972); Adv. Phys. **24**, 593 (1975).

[14] N. F. Mott, J. Non-Cryst. Solids **1**, 1 (1968).

[15] A. Goltzene, B. Meyer, and C. Schwab, J. Appl. Phys. **54**, 3117 (1983).

[16] R. B. Beall, R. C. Newman, and J. E. Whitehouse, J. Phys. C **19**, 3735 (1986).

[17] J. C. Bourgoin, H. J. von Bardeleben, and D. Stievenard, J. Appl. Phys. **65**, R65 (1988).