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Electrical properties of hydrogenated diamond

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Hydrogen passivation of deep traps in diamond is demonstrated. Current-voltage (I-V) characteristics of polycrystalline thin film and bulk diamond were studied before and after hydrogenation. On hydrogenation, all the samples showed several orders of magnitude increase in conductivity. Hydrogenation was carried out under controlled conditions to study the changes in the I-V characteristics of the samples. The concentration of uncompensated traps was varied systematically by hydrogenation. The concentration of electrically active hydrogen was determined from the I-V data. It is shown that hydrogenation is an alternative to deep-level transient spectroscopy, suitable for characterization of traps in a wide-band-gap material like diamond.

In two recent letters, it has been reported that the resistivity of diamond, both bulk and film, can be reduced by several orders of magnitude by hydrogen plasma treatment.[1] Hydrogen passivation of deep donors has been suggested and the increase in conductivity is thought to be due to shallow acceptor levels. Atomic hydrogen alters the electronic and optical properties of semiconductors. In the case of Si, atomic hydrogen reduces the leakage current of *pn* junctions, neutralizes both acceptors and donors, passivates point defects, and neutralizes deep-level defects due to Au, Cu, and Ni.[2–4] Grain boundary passivation in poly-Si is also reported. Similarly, Cu-related defects in Ge are passivated by atomic hydrogen.[5] Hydrogen passivation effects are also observed in III-V compounds.[6] In view of these results, hydrogen has been proposed as a passivaton agent for defects in polycrystalline superlattices and quantum wells.[7] Similar effects are expected in diamond, which will be useful for electronic devices. However, no systematic study has been reported yet. Here we show that the current-voltage (I-V) characteristics of type Ia synthetic diamond, type IIb natural diamond, and free-standing diamond films can be altered by the passivation of deep traps by atomic hydrogen. The concentration of electrically active hydrogen introduced by the plasma can be determined from the current-voltage characteristics. Controlled hydrogenation produces systematic changes in the I-V characteristics which reveal the nature of conduction in diamond samples.

The synthetic diamond samples were type Ia and the natural diamond samples were type IIb approximately 3×3 mm square, and 1.5 and 0.25 mm thick, respectively. The diamond thin films were polycrystalline about 0.120 mm thick, grown on silicon substrates using the microwave plasma-enhanced chemical vapor deposition (MPECVD) process. All samples were annealed in vacuum at 600 °C. Electrical contacts to the samples were made using silver epoxy which has been shown to make ohmic contacts.[8] I-V characteristics were measured using a Keithly 617 electrometer and stabilized power supply with computer interface for automatic data collection.

The samples were subjected to two kinds of hydrogenation process at various times using rf (13.56 MHz) plasma in a capacitively coupled barrel system and a microwave plasma in the MPECVD system and the I-V measurements were repeated.

In Fig. 1, the I-V characteristics obtained before and after hydrogenation of type Ia diamond are shown. The rf power was 50 W at 500 mTorr of hydrogen and the maximum temperature of the sample was ~ 100 °C. The I-V curves are linear showing ohmic behavior for applied field below 1 kV/cm. A systematic increase in conductivity is seen with hydrogenation time; for the 80 min hydrogenation conductivity is increased by five orders of magnitude. Similar results are obtained for the samples treated in a microwave plasma with 750 W at 10 Torr hydrogen and 200 °C substrate temperature. Microwave plasma is known to produce more atomic hydrogen than an equivalent rf plasma; yet high-power microwave plasma hydrogenation has only the same effect as a low-power rf plasma. Hence it is reasonable to assume that the amount of hydrogen introduced is controlled by its diffusivity in diamond. In the case of polycrystalline diamond films, the conductivity is saturated by 40 min hydrogenation as shown in Fig. 2. The relatively smaller change in conductivity is perhaps due to the hydrogen remaining after the 600 °C anneal, since the film was grown in a hydrogen-rich plasma. Again, for low applied fields, the I-V characteristics are linear. Although several orders of magnitude changes in conductivity are observed in these samples, for low applied fields no turning points in the I-V characteristics could be detected due to any trap-filling effect. This is simply an artifact of the combined effect of trap density, sample thickness, and the contact area. Hence the amount of hydrogen introduced cannot be determined from these I-V characteristics.

However, in the case of type IIb diamond samples, the I-V characteristics show some interesting features as shown in Fig. 3. Before hydrogenation in the microwave plasma, the I-V curve is linear up to 11 V; then the current increases by about nine orders of magnitude for an order of magnitude increase in applied voltage. This is the classic case of a deep trap level which is located below the Fermi level. Such a band structure as shown in Fig. 4, gives rise to a I-V characteristic exhibiting an ohmic to trap-filled limit (TFL).[9]

The voltage V_{TFL} at the turning point is related to the concentration of uncompensated traps $p_{t,0}$ approximately by

$$V_{\mathrm{TFL}} = q p_{t,0} L^2/\epsilon, \tag{1}$$

where q is the electronic charge, L is the sample thickness, and ϵ is its dielectric constant. From Eq. (1), $p_{t,0}$

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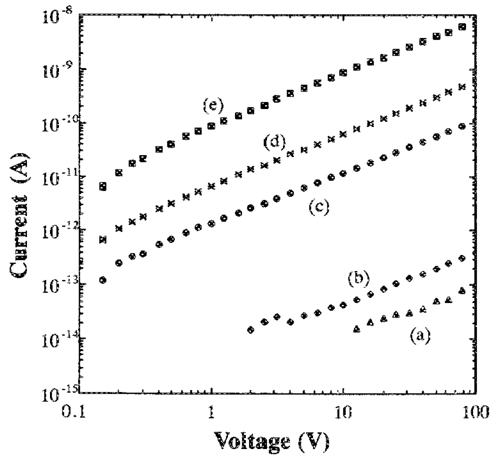

FIG. 1. I-V characteristics of type Ia diamond before and after hydrogenation with 50 W rf power at 500 mTorr. (a) Before hydrogenation, (b) 20 min, (c) 40 min, (d) 60 min, and (e) 80 min.

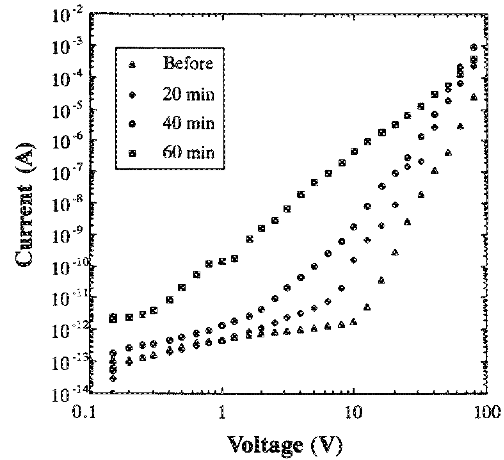

FIG. 3. I-V characteristics of type IIb diamond before and after hydrogenation with 750 W microwave power at 10 Torr.

$= 1.06 \times 10^{11}$ cm^{-3}. It has been shown[9] that

$$I(2V_{\text{TFL}})/I(V_{\text{TFL}}) = (p_{t,0}/n_0), \quad (2)$$

where n_0 is the free-carrier concentration. From Fig. 3, it is evident that $p_{t,0}$ is at least three orders of magnitude higher than n_0. Hence the conduction in this sample is dominated by the trap. On hydrogenation, V_{TFL} is found to shift to lower voltages as seen in Fig. 3. However,

$$p_{t,0} \propto \exp[-(F-E_t)/kT], \quad (3)$$

where F is the quasi-Fermi level pinned closer to the trap level E_t which is below the equilibrium Fermi level F_0, k is the Boltzmann's constant, and T is the absolute temperature. As the traps are compensated, F will move towards F_0, increasing the value of $(F-E_t)/kT$. Hence, $p_{t,0}$ and V_{TFL} will decrease on hydrogenation. This is in agreement with the experimental result shown in Fig. 3. From the shift in V_{TFL} on hydrogenation, we can deduce the amount of electrically active hydrogen introduced in diamond, which is shown in Fig. 5 as as function of hydrogenation time. For the limited experimental data, the amount of hydrogen introduced depends linearly on time.

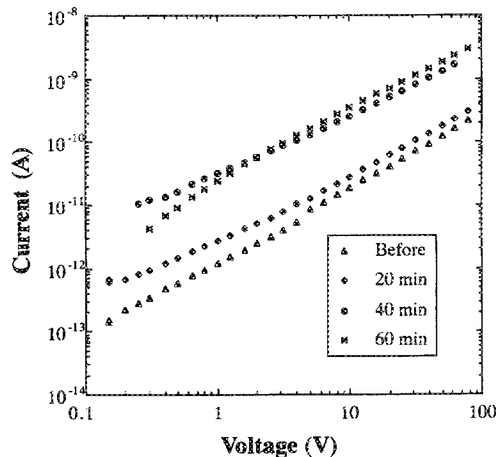

FIG. 2. I-V characteristics of polycrystalline diamond film before and after hydrogenation with 50 W rf plasma at 500 mTorr.

On closer examination of Fig. 3, we notice that the slopes of the curves in the TFL region decrease with hydrogenation, because the ratio $(p_{t,0}/n_0)$ decreases. The slope change seen in Fig. 3 is, therefore, consistent with this argument. After the complete passivation of the traps by hydrogenation, the TFL region will disappear and a trap-free square-law region should result. This is exactly what is observed as shown in Fig. 6 for the same sample hydrogenated for complete passivation under identical conditions. The crosses are experimental points and the lines correspond to ohmic and trap-free space-charge regions. The free-carrier concentration and mobility can be determined from these data. Thus, it appears that hydrogenation under controlled conditions is a suitable method for changing the density of electrically active traps and hence the I-V characteristics of the diamond samples. In this regard, this method will be much simpler than the deep-level transient spectroscopy (DLTS) technique to study the nature of traps in wide-band-gap materials such as diamond.

In conclusion, the electrical conductivity of diamond, both film and bulk, can be increased several orders of magnitude by hydrogenation using a low-power plasma. Hydrogenation seems to be independent of plasma power and hydrogen pressure. Grain boundaries in polycrystalline films are passivated by hydrogen, while deep traps in bulk dia-

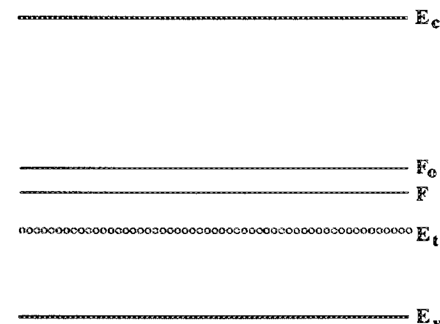

FIG. 4. Band structure for a deep trap in type IIb diamond.

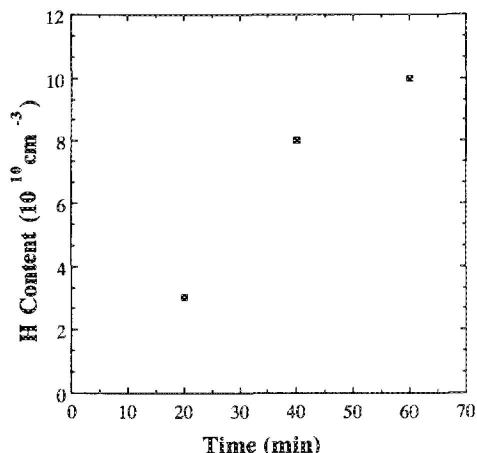

FIG. 5. Concentration of electrically active hydrogen.

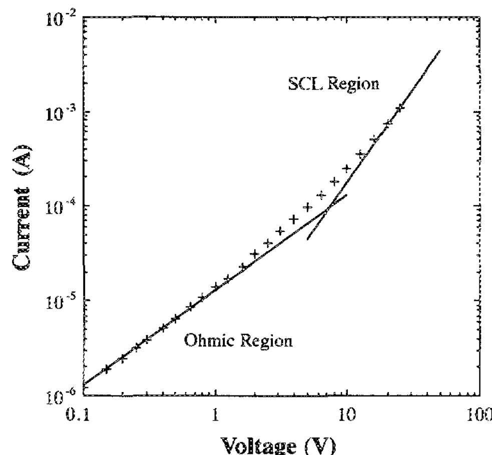

FIG. 6. *I-V* characteristics of the sample in Fig. 3, after complete passivation of deep traps by hydrogenation. The crosses are experimental data points and the lines correspond to ohmic and trap-free space-charge-limited regions of slopes 1 and 2, respectively.

mond are passivated. Controlled hydrogenation can reveal subtle changes in the *I-V* characteristics of diamond samples. The concentration of electrically active hydrogen, introduced by the plasma, can be measured from the shifts in the *I-V* characteristics of the samples containing deep traps. Hydrogenation appears to be simpler than DLTS to study the trap-related properties of wide-band-gap materials such as diamond.

Linwood Watkins is a NASA Graduate Fellow.

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