Observation of transferred-electron oscillations in diamond

Cite as: Appl. Phys. Lett. 115, 192101 (2019); https://doi.org/10.1063/1.5126058
Submitted: 29 August 2019 . Accepted: 22 October 2019 . Published Online: 04 November 2019

N. Suntornwipat, S. Majdi, M. Gabrysch, I. Friel, and J. Isberg

Lock-in Amplifiers ... and more, from DC to 600 MHz
Observation of transferred-electron oscillations in diamond

Cite as: Appl. Phys. Lett. 115, 192101 (2019); doi: 10.1063/1.5126058
Submitted: 29 August 2019 · Accepted: 22 October 2019 · Published Online: 4 November 2019

N. Suntornwipat,1 S. Majdi,1 M. Gabrysch,1 I. Friel,2 and J. Isberg1,a)

AFFILIATIONS
1Division for Electricity, Department of Engineering Sciences, Uppsala University, Box 534, 751 21, Uppsala, Sweden
2Element Six Innovation, Fermi Avenue, Harwell Oxford, Didcot, Oxfordshire OX11 0QR, United Kingdom

a)Author to whom correspondence should be addressed: jan.isberg@angstrom.uu.se

ABSTRACT
The transferred-electron oscillator (TEO), or Gunn oscillator, is a device used in microwave applications, which utilizes the negative differential mobility (NDM) effect to generate continuous oscillations. Recently, NDM was observed in intrinsic single-crystalline chemical vapor deposition (SC-CVD) diamond. The occurrence was explained by the electron repopulation between its different conduction band valleys. This paper presents the results of constructing a diamond TEO based on the NDM effect. A series of experiments have been performed for varying voltages, temperatures, and resonator parameters on three SC-CVD diamond samples of different thicknesses. For the temperature range of 90–300 K, we observe transferred-electron oscillations in diamond.

© 2019 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/). https://doi.org/10.1063/1.5126058

Transferred-electron devices (TEDs) or Gunn diodes are widely used in microwave applications as local oscillators and drivers for amplifier chains.1,2 The TED is one type of negative resistance oscillator that uses negative differential mobility (NDM), caused when electrons are scattered from the central conduction band valley with low effective electron mass to satellite valleys with high effective electron mass, to generate continuous oscillations. Oscillations in InP and GaAs devices were first reported by Gunn3,4 in 1963, but he did not give an explanation for the phenomenon at the time. However, it turned out that oscillations had already been predicted by Hilsum5 in the previous year, based on a suggestion by Ridley and Watkins6 that certain materials should exhibit the NDM effect. Gunn oscillations are usually only observed in direct bandgap materials such as GaAs, InP, InAs, CdTe, and ZnSe3–9 and are not normally associated with elemental or indirect-bandgap semiconductors.

Diamond exhibits many exceptional material properties such as high thermal conductivity, extreme mechanical strength, high carrier mobilities, and chemical inertness. Together with the possibility to synthesize high-purity single-crystalline chemical vapor deposition (SC-CVD) diamond,12 it is a very interesting material for semiconductor devices. Since diamond is an indirect semiconductor without a central (Γ-point) valley in the conduction band, one might not expect that this material exhibits the NDM effect. Nevertheless, the first observation of NDM occurring in intrinsic SC-CVD diamond was reported in Ref. 11 at a temperature range of 110 to 140 K for an applied electric field of 300 to 600 V/cm in the [100] direction.

Even though diamond has energetically equivalent valleys, NDM occurs because of the anisotropic transport properties of electrons in different valleys, i.e., by different electron effective masses in different directions. The six conduction band valleys in diamond are situated at 76% of the distance from the Γ-point on the A-axis in the Brillouin zone (BZ), as illustrated in Fig. 1(a).12–14 When applying an electric field in one direction (in the present case, along [100]), electrons in the two valleys aligned parallel [on the (100) axis] to the field and the four valleys aligned orthogonally [on the (010) and (001) axes] to the field respond with different effective masses. The longitudinal electron effective mass (mL*) is 1.56m0, and the transverse electron effective mass (mT*) is 0.28m0.15 In contrast, in the case of GaAs, NDM is explained by the repopulation of electrons between a central conduction band valley (Γ-point) and satellite valleys (L-point), with different energy minima (about 300 meV).16 This is shown in Fig. 1(b).

The possibility of using the NDM effect in diamond devices has been investigated with a theoretical model to establish if it is feasible to demonstrate a diamond based transferred-electron oscillator (TEO).17 In this model, the TED operates with an electric field applied in such a way that the internal space charge distribution and the internal electrical field distribution become unstable and cause oscillations in a resonant circuit. This paper presents experimental results of an actual
cryostat. The amplified current is recorded with a digital oscilloscope (pHEMT) based amplifier cooled to sample temperature inside the cryostat or a custom-built pseudomorphic high-electron-mobility transistor amplifier placed outside the cryostat (Mini-Circuits ZFL1000LN mounted on the cold finger of a vacuum cryostat. The current through the front in order to make the electrons travel through the sample and to extract the holes at the illuminated side. The circular contacts are situated along {111} directions. Gunn oscillations in GaAs instead arise from electron repopulation between central and satellite valleys. (c) A diamond Gunn oscillator where the oscillations in current are the results of electrons repeatedly depleting and accumulating inside an intrinsic diamond film.

FIG. 1. The band structure of (a) diamond and for comparison (b) GaAs. The valleys in diamond are located along [100] directions near the zone boundary and have the same minimum energy of 5.47 eV above the valence band maximum. The 3D pictures show equal-energy surfaces in the first Brillouin zone surrounding the conduction band minima. When applying an electric field along the [100] direction, the two valleys aligned in parallel to the field and the four valleys aligned orthogonal to the field respond with different electron effective masses and mobilities. In GaAs, the central valley (T) lies about 300 meV below the ellipsoidal satellite valleys (L) situated along [111] directions. Gunn oscillations in GaAs instead arise from electron repopulation between central and satellite valleys. (c) A diamond Gunn oscillator where the oscillations in current are the results of electrons repeatedly depleting and accumulating inside an intrinsic diamond film. (Tektronics TDS 684C, 5 G/s). A doubly shielded casing is used to carefully isolate the sample and the pHEMT amplifier from any radio frequency noise generated in the outside environment. To stabilize the bias voltage, a capacitor of 33 nF with NP0 dielectric is added in close proximity to the sample. A calibrated GaAlAs diode sensor (TG-120-CU-HT-1.4H) in contact with the sample holder is used for temperature monitoring in conjunction with a Lake Shore 331 temperature controller. Liquid nitrogen is used as the coolant. The schematic of the measurement setup is shown in Fig. 2.

At low temperatures, intervalley scattering caused by short-wavelength lattice vibrations are absent in diamond due to the extreme rigidity of the lattice. This makes it possible to observe electrons moving with different velocities depending on their valley state. At low electric fields, all valleys are roughly equally populated and the electron drift velocity is proportional to the field. As the applied electric field is increased, the electrons in orthogonal valleys are heated up more than the electrons in parallel valleys due to the difference in their effective masses along the direction of the field. The electrons from the orthogonal valleys reach the energy threshold for the longitudinal acoustic (LA) phonon emission of 120 meV first, which leads to intervalley scattering and a strong repopulation of conduction band electrons from orthogonal to parallel valleys. This phenomenon causes the observed negative differential mobility. As the electric field is increased further, electrons in the parallel valleys will reach a sufficient energy to scatter into the orthogonal valleys, which tends to equalize the valley population. If NDM is present and the carrier concentration exceeds a certain threshold, instabilities in the spatial carrier distribution can occur as fast electrons catch up with slow electrons. This causes a build-up of an accumulation layer and at the same time leaves a depleted region behind. The internal space charge distribution of such accumulation and depletion of electrons results in an oscillating current.19 Figure 1(c) shows a Gunn oscillator where the oscillations in current are the results of electrons repeatedly depleting and accumulating inside intrinsic diamond.

Experiments were performed at a temperature range of 90 to 300 K using three high-purity SC-CVD diamond samples from Element Six Ltd. with sample thicknesses of 307, 390, and 420 μm. The details of the samples used for this study are summarized in Table I. The nitrogen impurity concentration in all the chosen samples is below
the detection limit of EPR. For samples with a nitrogen concentration well above $10^{14}$ cm$^{-3}$, no TEO could be observed.

The samples were illuminated by the deep ultraviolet (DUV) laser, and the induced current was measured. Several different experiments were performed with inductance and capacitance values of external LC circuit varying between 1.8 to 10 $\mu$H and 0 to 4.7 pF, respectively. For $C = 0$, the resonance relies on the internal capacitance of the sample (approximately 0.5 pF) and stray capacitance in the sample holder (approximately 1 pF). For the temperature range where NDM had previously been reported, we observed oscillations in the frequency range of 20 to 50 MHz depending on the chosen inductor and capacitor values. The resonant frequency $f$ can be estimated by

$$f = \frac{1}{2\pi\sqrt{L(C + C_{\text{sample}} + C_{\text{stray}})}}.$$

Figures 3–5 show the measured current from three such tests. Here, the 307 and 390 $\mu$m samples were separately connected in series with an inductor of 5.6 $\mu$H (devices A and B, respectively), while the 420 $\mu$m sample was connected in series with an inductor of 10 $\mu$H (device C). No external capacitor was connected in either case, and bias voltages of 0 to 30 V were applied across the devices. The observed resonant frequencies agree well with simulated values and also roughly with the simple expression above. The current waveforms at the temperatures of 90 and 110 K with bias voltages of 0 and 20 V are shown for device A in Fig. 3. With a voltage applied, current oscillations are observed having a much larger amplitude at 110 K compared to 90 K.

A comparison of the current waveforms in the frequency domain at different voltages and at temperatures of 90, 110 and 290 K is presented for device A in Fig. 4. The oscillations are most pronounced at

| Sample | Description     | Size (mm) | Thickness ($\mu$m) | Impurity conc. ($\text{cm}^{-3}$) |
|--------|----------------|-----------|-------------------|-------------------------------|
| #A     | SC-Intrinsic   | 2.8 x 2.8 | 307               | $<10^{14}$                    |
| #B     | SC-Intrinsic   | 3.5 x 3.5 | 390               | $<10^{14}$                    |
| #C     | SC-Intrinsic   | 4.5 x 4.5 | 420               | $<10^{14}$                    |
detailed model that includes a full description of the population observed also at temperatures above 150 K, and we suggest that a differential carrier mobility. Surprisingly, weak oscillations have been observed by electron density instabilities occurring in situations with negative capacitor values of the LC circuit. These oscillations can be explained by oscillations of the three devices. The reason for this is not clear and requires further investigations with more samples of different thicknesses. One possibility is that the actual penetration depth of the illumination is somewhat smaller than the value used for the simulations in Ref. 17. It should be noted that the oscillations are observed outside the NDM region (110 to 140 K) but rapidly diminish as the temperature is lowered below 100 K.

In all three cases, the oscillator strength is at its maximum at 110 to 130 K, as would be expected from simulations.\textsuperscript{11} This can be explained by the negligible rate of intervalley phonon scattering at low temperatures, which effectively inhibits valley repopulation.\textsuperscript{12} Oscillations are, however, also observed well above 150 K, and their amplitude only slowly diminishes with increasing temperature. Even at room-temperature, weak oscillations can be observed. Clearly, the model presented in Ref. 17 does not explain the high-temperature behavior. In this model, the population ratios are defined by first order rate equations, and we believe that this approach may be too simplistic to capture the detailed “dynamics” of the valley population. Presumably a more comprehensive model that includes a full description of the population dynamics and the scattering rates could give an explanation for this observation.

The authors would like to thank the Swedish Research Council (VR, Grant No. 621-2012-5819, 621-2014-6026, and 2018-04154) for financial support. In addition, the Swedish Energy Agency (Grant No. 44718-1) is gratefully acknowledged by the authors. S.M. would like to thank Professor S. Shikata (AIST) for providing the opportunity to conduct part of the research in his group and express his gratitude to the Japan Society for the Promotion of Science (JSPS, Grant No. PE 13542) for financial support.

**REFERENCES**

1. D. Q. Wu, R. Jia, and Y. Bai, Adv. Mater. Res. 684, 299 (2013).
2. S. M. Sze and M. K. Lee, Semiconductor Devices: Physics and Technology, 3rd ed. (John Wiley & Sons, Inc., 2012).
3. J. B. Gunn, Solid State Commun. 1, 88 (1963).
4. J. B. Gunn, IEEE Trans. Electron Devices 23, 705 (1976).
5. C. Hilsenrath, Proc. Inst. Radio Eng. 50, 185 (1962).
6. K. K. Ridley and T. B. Watkins, Proc. Phys. Soc. 78, 293 (1961).
7. W. Allen, M. Shyam, and G. L. Pearson, Appl. Phys. Lett. 9, 39 (1966).
8. M. R. Oliver and A. G. Foyle, IEEE Trans. Electron Devices 14, 617 (1967).
9. G. W. Ludwig, R. E. Halsted, and M. Even, IEEE Trans. Electron Devices 13, 671 (1966).
10. J. Isberg, J. Hammersberg, E. Johansson, T. Wikström, D. J. Twitchen, A. J. Whitehead, S. E. Cox, and G. A. Scarbrough, Science 297, 1670 (2002).
11. J. Isberg, M. Gabrysch, S. Majdi, and D. Twitchen, Appl. Phys. Lett. 100, 172103 (2012).
12. J. Isberg, M. Gabrysch, J. Hammersberg, S. Majdi, K. K. Kovi, and D. J. Twitchen, Nat. Mater. 12, 760 (2013).
13. N. Suntornwipat, M. Gabrysch, S. Majdi, D. J. Twitchen, and J. Isberg, Phys. Rev. B 94, 035408 (2016).
14. J. Hammersberg, S. Majdi, K. K. Kovi, N. Suntornwipat, M. Gabrysch, D. J. Twitchen, and J. Isberg, Appl. Phys. Lett. 104, 232105 (2014).
15. N. Naka, K. Fukai, Y. Handa, and I. Akimoto, Phys. Rev. B 88, 035305 (2013).
16. M. Lundstrom, Fundamentals of Carrier Transport, 2nd ed. (Cambridge University Press, 2000).
17. N. Suntornwipat, S. Majdi, M. Gabrysch, and J. Isberg, Appl. Phys. Lett. 108, 212104 (2016).
18. C. D. Clark, P. J. Dean, and P. V. Harris, Proc. R. Soc. London, Ser. A 277, 312 (1964).
19. D. Shiri, A. Verma, R. Nekovei, A. Isacsson, C. R. Selvakumar, and M. P. Anantram, Sci. Rep. 8, 6273 (2018).