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Observation of periodical negative differential conductivity in nanocrystalline silicon/crystalline silicon heterostructures

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Abstract
We report the observation of periodical negative differential conductivity (NDC) in hydrogenated nanocrystalline silicon/crystalline silicon diode heterostructures by low-temperature current–voltage measurements. The NDC-related series of spike-like current peaks is found to result from the accumulation and depletion of electrons tunnelling through the nanodot layers in the neutral region. The observation has been further supported by the facts of onset-voltage blueshift, number variation in current peaks, and inter-Landau-level tunnelling in the temperature- and magnetic field-dependent experiments. We also discuss the differences between the present periodical NDC and the electric field domain in superlattice structures.

Instability in semiconductor material is frequently related to the occurrence of negative differential conductivity (NDC), which has proven to be very useful in the generation, amplification, switching and processing of microwave signals [1]. Among all the NDC effects, periodical NDC is one of the most special ones; it has only been observed in weakly coupled semiconductor superlattices (SLs) due to the existence of the electric field domain and domain-related current oscillations, where the latter are of more practical interest, because the oscillation frequency may vary in a wider range of the electromagnetic spectrum from several kHz to tens of GHz [2–6]. Phenomenally, periodical NDC will embody itself as a series of current peaks in the current–voltage (I–V) characteristics; which actually contain adequate information to reflect the spacing of the electronic subbands and the number of periodic layers in SLs. These interesting vertical transport phenomena have been much investigated both theoretically and experimentally during the past years [7–10].

So far, discussions of the periodical NDC effect have been limited to man-made SLs, and much less attention has been paid to other structures. This is probably because the SL has a uniform periodic arrangement of potential in the material, and the periodicity can be artificially controlled during the growth process. However, if this beautiful effect could be extended to different structures other than SLs, especially to those of naturally grown materials, the academic research as well as the underlying applications will be more attractive and valuable. Recently, considerable interest has been given to semiconductor nanostructures because of their potential applications in optoelectronic devices, such as single electron transistors [11] and single photon detectors [12]. Among them, natural quantum dots of silicon are found to have the advantage of greater fabrication simplicity over artificial quantum dots without sacrificing the low-dimensional tunnelling characteristics [13].

In this paper, we report on the observation of the periodical NDC phenomenon in hydrogenated nanocrystalline silicon (nc-Si:H) thin films grown on crystalline silicon (c-Si) substrates, where the natural quantum dots of nc-Si:H are embedded in thin amorphous Si (a-Si) barriers. The physical process of periodical NDC has been revealed and well confirmed by the temperature- and magnetic field-dependent I–V experiments. These exciting findings break the limitation that periodical NDC could only be demonstrated in artificial SLs and may open a way to investigate this effect in natural nanodot systems.

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The nc-Si:H/c-Si diode heterostructure studied was prepared in an rf (13.56 MHz) capacitive-coupled plasma-enhanced chemical vapour deposition system from silane (SiH_4) and hydrogen (H_2) at a temperature of 250 °C [13, 14]. The percentage content of silane ($SiH_4/SiH_4 + H_2$) was about 1.0%. The n-type nc-Si:H thin film was doped with phosphine (PH_3/SiH_4) $\sim$ 0.8% and grown on slightly doped p-type c-Si substrate ($N_A \sim 10^{15}$ cm^{-3}). The electron concentration N_D in the nc-Si:H thin film was about 5×10^{17} cm^{-3}; this was extracted from the magnetic field-dependent Hall effect experiment. High-resolution transmission electron microscopy revealed an ordered nc-Si:H microstructure on the c-Si substrate. X-ray diffraction and Raman measurements also showed the good quality of the nc-Si:H thin film with an average grain size of ~5 nm. Indium was deposited as metal electrodes on both the top and bottom sides of the diode. The temperature- and magnetic field-dependent I–V curves were obtained by using a computer-controlled Keithley 2400 sourcemeter under an Oxford Instruments superconductive magnet (temperature from 10 to 300 K, and magnetic field up to 15 T).

The electronic energy band diagram along the growth direction of the nc-Si:H/c-Si p–n heterostructures studied is considered as a superposition of the nanodot structure and heterojunction energy band, as plotted in figure 1(a) for the conduction band profile, where the heterojunction energy band can be calculated by self-consistently solving the Schrödinger and Poisson equations [15]. Figure 1(b) shows a typical I–V curve of the nc-Si:H/c-Si sample at a temperature of 10 K. Besides the clear diode-like characteristic, two important structures are observed in the reverse-bias region, one of which is a step-like structure appearing within the voltage range of −11 to −13 V, while the other is a series of spike-like current peaks in the voltage range from −17 to −19 V.

The two structures above have been enlarged in figures 1(c) and (d) for clarity, both of which directly reflect the unusual electron transport behaviour in the nc-Si:H/c-Si p–n heterostructure system under the reverse bias. The electrons as minority carriers are swept out from the p-type c-Si substrate to the n-type nc-Si:H thin film under the electric field force. Because of the conduction band offset between c-Si and a-Si, electrons accumulate at the interface of the substrate and the thin film, forming a two-dimensional electron gas (2DEG), as shown in figure 1(a), and the voltage is almost completely applied on the barrier region of the junction at this time. On increasing the reverse voltage, electrons accelerated from the c-Si substrate can first tunnel to the states in the 2DEG, and afterwards to the thin film when the Fermi level in the 2DEG is higher than that of the zero-dimensional (0D) electronic state in the nc-Si:H nanodots. This transport process across the 3D–2D–0D system leads to the observed step-like resonant structure in the I–V curve (−11 to −13 V). More detailed theoretical analysis has been successfully achieved via the self-consistent calculation together with a transfer matrix procedure. We have displayed in figure 1(c) the calculated resonant tunnelling structure (dotted curve), which shows a good accordance with the experimental one (solid curve).

With further increase of the reverse bias, the energy band of thin film within the barrier region gets steeper. When the ground electronic state in one nanodot aligns to the excited state in the adjacent nanodot, most of the electrons can transport through the nanodots in the barrier region and arrive at the nanodot layer lying at the boundary of the barrier and neutral regions (dashed line in figure 1(a)). In such a high applied voltage along the growth direction, the 3D quantum confinement in the nanodots may turn into 1D nanodot layers along the electric field direction because of the field-induced localization [16]. As electrons tunnel through the nanodot layers in the neutral region, they may experience a process of accumulation and depletion. The electron accumulation comes from the ground state mismatch between the neighbouring layers, while the depletion occurs when the excited state in the first layer aligns to the ground state in the next one, leading to the appearance of a current peak. Therefore, the periodical NDC (spike-like current peaks in figure 1(d)) appears as expected.

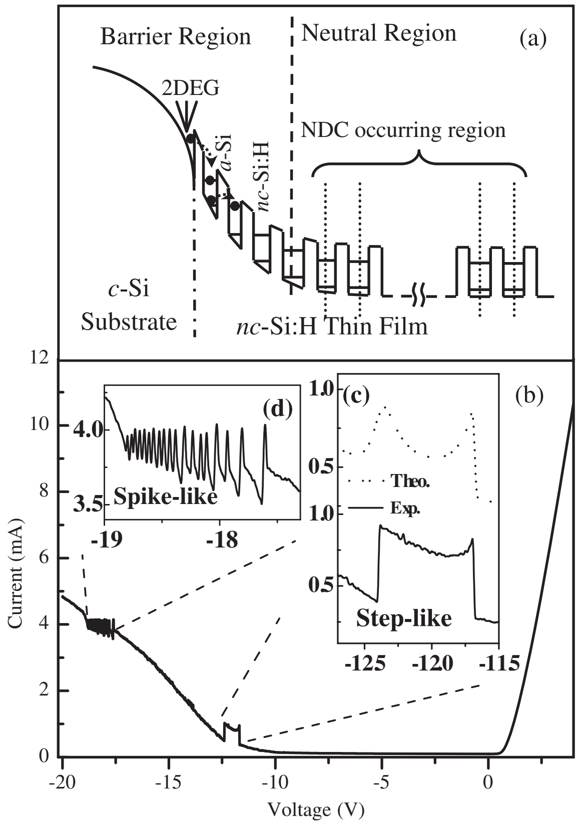

Figure 1. (a) Schematic conduction band diagram of the nc-Si:H(n)/c-Si(p) diode heterostructure with an applied reverse bias. (b) The I–V characteristic of nc-Si:H/c-Si diode heterostructure at a temperature of 10 K. (c) Enlarged experimental (solid curve) and calculated (dotted curve) step-like structures. (d) Enlarged experimental spike-like structure.

According to the analysis above, the separation between two current peaks should correspond to the spacing of the electronic subbands in nanodot layers. In our observation, the separation between the neighbouring current peaks is not so uniform, especially at the first several current peaks. On increasing the voltage, the separation becomes smaller and finally reaches a value of about 38–40 meV. The nonuniformity comes from the fact that the existence of a size distribution in our sample is inevitable, which leads to the different electronic state spacings at different nanodot layers. This confirms the

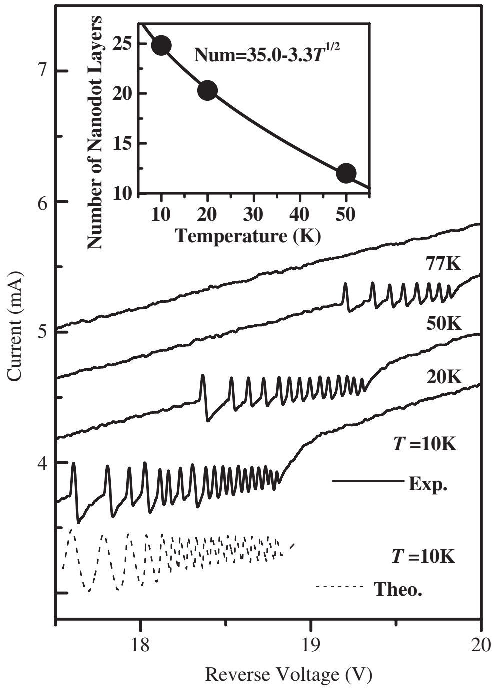

Figure 2. Temperature-dependent spike-like structures in the nc-Si:H/c-Si diode heterostructure, with the curves shifted by 0.4 mA for clarity. The dotted curve is the calculated spike-like structure at a temperature of 10 K. The inset plots the relationship between the temperature and the number of nanodot layers in the neutral region.

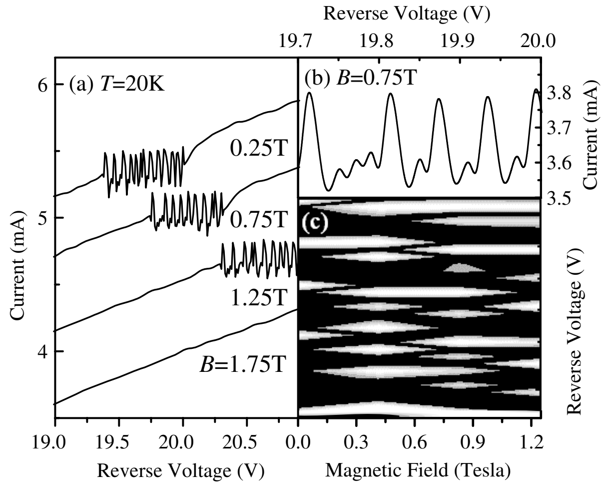

Figure 3. (a) Magnetic field-dependent spike-like structures in the nc-Si:H/c-Si diode heterostructure at a temperature of 20 K. (b) Enlarged current peaks at $B = 0.75$ T, where inter-Landau-level tunnelling is clearly observed. (c) Greyscale plot of the differential conductance $G = \mathrm{d}I/\mathrm{d}V$ as a function of bias voltage and magnetic field.

argument that the periodical NDC does originate from repeated electron accumulation and depletion in nanodot layers. Larger peak energy spacing at the beginning implies that the initial accumulation and depletion of electrons occurs across several nanodot layers simultaneously, induced by elastic scattering effects such as interface roughness and/or impurity scattering existing at the boundary of the barrier and neutral regions. Therefore, if we divide the whole NDC energy range by the average stable separation, one may deduce that the number of nanodot layers in the neutral region is about 25 at 10 K, more than the observed 19 current peaks in the I–V curve.

Experimental evidences of the periodical NDC in our nanodot system are also clearly manifested in the temperature- and magnetic field-dependent results. Figure 2 displays the I–V characteristics at different temperatures with the spike-like behaviour appearing up to $T = 50$ K. When the temperature is above 50 K, the electrons are thermally activated into the extended state above the barriers among the nanodots, resulting in the disappearance of the periodic tunnelling peaks in the I–V curves. The decrease in the amplitude of the peaks with increasing temperature may be qualitatively explained by an enhancement of electron–phonon scattering [2]. On increasing the temperature, two other interesting observations are revealed. One is the explicit blueshift of the onset voltage when NDC appears; the other is the dramatic decrease of the number of observed peaks.

As we know, with the increase in temperature, the built-in potential in diodes becomes larger [17], while the self-consistent calculation indicates that the energy band bending in the barrier region is gentler. Therefore, much greater applied voltage should be used, with a larger width of the barrier region at higher temperature. This, together with the larger voltage division by the slightly doped c-Si substrate, results in the explicit blueshift of the onset applied voltage. Furthermore, the decreasing number of current peaks comes from the reduction of nanodot layers in the neutral region, which is linked with the increase of the barrier region width. Through estimation, the number of nanodot layers in the neutral region can be deduced as 20 and 12 at temperatures of 20 and 50 K, respectively. Considering the $\sqrt{T}$ dependence of the barrier region width in p–n junctions [17], we may use a simple formula of $b - a\sqrt{T}$ to fit the relationship between the temperature T and the number of nanodot layers in the neutral region, where b represents the total number of nanodot layers in the thin film, and $a\sqrt{T}$ is the number of nanodot layers in the barrier region, with a an overall parameter related to the nanodot properties as well as the band structure. The inset of figure 2 plots the experimental data together with the fitting curve, which yields $b = 35$ and $a = 3.3$, indicating that there are in total about 35 nanodot layers along the growth direction in the thin film. The observed temperature dependence of the number in the current peaks confirms the argument that periodical NDC occurs when electrons tunnel through the nanodot layers in the neutral region.

More evidence comes from the magnetic field-dependent experiments with B applied parallel to the direction of current flow. Figure 3(a) shows the spike-like I–V characteristics at 20 K for $B = 0.25, 0.75, 1.25$, and 1.75 T, respectively. When $B = 1.75$ T, the spike-like structure may lie out of our measurement range since a larger reverse voltage will make the diode break down. Compared to the results without magnetic field, the most striking feature is a multiplication of the number of current peaks, as shown enlarged in figure 3(b). For a clearer illustration, we plot in figure 3(c) the numerically obtained differential conductance $G = \mathrm{d}I/\mathrm{d}V$ as a function of bias voltage and magnetic field, where only the first five main peaks are included. White lines reflect regions of strong NDC, i.e., the positions of the sharp current drops that follow the peaks in the I–V data. According to figure 3(c), we find that in the presence of a magnetic field, clear additional resonances appear between each pair of original resonances presented at $B = 0$ T.

The extra resonances originate from the formation of Landau levels (LLs) $E_{i,n} = E_i + (n+1/2)\hbar\omega_c$, where $i = 1, 2$ is the subband index, $n = 0, 1, 2 \ldots$ is the Landau quantum number, and $\omega_c = eB/m^*$ is the cyclotron frequency. In the absence of scattering, tunnelling requires the conservation of the Landau index. Inter-LL tunnelling is forbidden, since the in-plane wave functions of the LLs are orthogonal harmonic oscillator eigenfunctions [10]. However, elastic scattering processes such as interface roughness or impurity scattering as well as optical-phonon emission lead to inter-LL tunnelling [18, 19]. In the case of strong elastic scattering, as expected for our doped nanodot material, resonant inter-LL tunnelling $E_{1,0} \rightarrow E_{1,m}$ at the domain boundary results in the extra resonances between each pair of original resonances at bias voltages of $\Delta V_m = m(\hbar\omega_c)/e\alpha$, where α denotes the voltage-to-energy conversion coefficient. The cyclotron energy at $B = 0.75$ T is calculated as 0.002 eV, while the experimental observed magnetic field-dependent ΔV_m is about 0.02 V. Thus we obtain $\alpha \approx 0.1$, which is close to the value reported in [13]. In addition, the observed ΔV_m in figure 3(c) does become larger with increasing magnetic field. Moreover, we notice that the resonance moves to higher bias with increasing magnetic field in figure 3(a). This is because $E_{1,0}$ increases with the magnetic field, where the applied voltage has to be increased to sustain the electron flow in the nanodot layers.

The above experimental phenomena clearly demonstrate the existence of periodical NDC due to the process of electron accumulation and depletion. However, since the size of nanodots is unable to reach a high level of uniformity with a growth technique like self-assembly, the induced energy level discrepancy may prevent us from observing this kind of NDC. Nevertheless, it is the p–n junction that compensates this nonuniformity problem. In particular, the energy level of nanodots can be fine tuned with the aid of the potential dividing function of the barrier region within the p–n junction. At the beginning of voltage sweeping, the very small reverse bias added across the neutral region facilitates ground-state alignment in between the layers. When the electrons in each nanodot layer begin to repeatedly experience accumulation and depletion, the increased bias is entirely added to the neutral region as the resistivity across this region is much larger than that across the others. However, the compensation of the p–n junction will not be active under reversal sweeping from -20 to 0 V; the I–V characteristics are of course not reversible.

Moreover, the p–n junction in our system also leads to a special injection of electrons into the nanodot layers. Two segments of conductivity have been clearly observed in the positive differential conductivity region, especially in the first several tunnelling peaks (see figure 2). This phenomenon can be explained as follows: at the boundary of the barrier and neutral regions, some of the electrons in one nanodot layer tunnel from the former neighbouring nanodot layer, while another portion of the electrons, with kinetic energy higher than the a-Si barrier height, will drift directly through the border of the barrier and neutral regions. After continuous scattering by the atoms in the neutral region, these high-energy electrons gradually release their energies and are trapped into electronic states in the nanodot when the energy is lower than the barrier height. As a result, these two different electron transport processes lead to the observed two-segment conductivities in the positive differential conductivity region, whereas for the nanodot layers away from the barrier/neutral boundary, since almost all the electrons have already been trapped into the first several nanodot layers, only resonant tunnelling from the neighbouring nanodot layer is available.

Finally, we will discuss the differences between the periodical NDC observed in our nanodot system and the formation of electric field domains in SLs, both of which manifest similar I–V characteristics. For comparison, a rates equation transport model [20] is applied to an assumed c-Si/a-Si SL structure with the equivalent structure parameters of our sample. Two electronic states at energies $E_1 = 15.4$ meV and $E_2 = 54.8$ meV are calculated through the transfer matrix method, and the energy spacing can be deduced as $\Delta E = E_2 - E_1 = 39.4$ meV. The profile of the theoretical result (dotted curve in figure 2) is found to be close to the experimental one. However, differences should be noticed in such aspects as the separation of two neighbouring peaks and the region of positive differential conductivity. These differences are actually a reflection of the nonuniformity of the nanodot size and the role of the p–n junction in our sample. In addition, in our special 3D–2D–0D nc-Si:H/c-Si p–n heterostructure system, the number of current peaks is variable either with the temperature or magnetic field, which may help us realize current peak number controllable devices through external fields instead of growth conditions in SLs.

In summary, we have shown that the spike-like current characteristics in nc-Si:H/c-Si diode heterostructures is the result of a periodical NDC effect due to the accumulation and depletion of electrons tunnelling through the nanodot layers in the neutral region. A wide range of experimental results, including the spike-like current profile, onset-voltage blueshift and peak number variation with the increase of temperature, as well as the inter-Landau-level tunnelling under external magnetic field, are all consistent with our theoretical analysis and assignment. Compared with the similar NDC in SL structures, which usually occurs under very strict conditions, it is the p–n junction that compensates the nonuniformity of the nanodot size distribution. The present achievement in naturally grown quantum dots of nc-Si:H indicates a significant step towards the investigation and application of the periodical NDC effect in nanodot materials.

Acknowledgments

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