Solid-State Electronics 79 (2013) 60–64

Contents lists available at SciVerse ScienceDirect

Solid-State Electronics

journal homepage: www.elsevier.com/locate/sse

Observation and study of the negative magnetoresistance in porous silicon

B. Chouaibi [a], A. Benfredj [a], S. Romdhane [a,b,*], M. Bouaïcha [c], H. Bouchriha [a]

[a] Laboratoire Matériaux Avancés et Phénomènes Quantiques, Faculté des Sciences de Tunis, Université El-Manar, 2092 Campus Universitaire Tunis, Tunisia
[b] Faculté des Sciences de Bizerte, 7021 Zarzouna, Bizerte, Université de Carthage, Tunisia
[c] Laboratoire de Photovoltaïque, Centre de Recherches et des Technologies de l'Energie, BP 95 Hammam-Lif 2050, Tunisia

ARTICLE INFO

Article history:
Received 17 December 2011
Received in revised form 13 June 2012
Accepted 25 July 2012
Available online 8 October 2012

The review of this paper was arranged by S. Cristoloveanu

Keywords:
Porous silicon
Nanowire
Magnetoresistance
Electronic properties
Weak localization
Diffusive semiconductors

ABSTRACT

We investigate the effect of a static magnetic field on the electrical properties of thin films of porous silicon. We observe a relative variation of the resistance with respect to intensity of the applied magnetic field. This effect called magnetoresistance (MR) has been found to be negative at room temperature in the entire range of the applied magnetic field (under 8000 G). The negative MR effect depends strongly of the applied voltage and reaches up to 5% at the value of magnetic field of 8000 G. The analysis has been based on the 1D weak localization theory (WL) in diffusive semiconductors, from which a consistent picture emerges, considering the PS film as networks of quasi-one-dimensional wires. We give an explicit expression for the WL correction depending only of the phase coherence length L_φ. A good agreement is observed between experimental data and theory.

1. Introduction

Magnetoresistance (MR) is a change of the resistance of a material system caused by an applied magnetic field. This effect in micro and nanostructured semiconductors, e.g., quantum dots, wires, and rings, is topic of intense research since the early days of mesoscopic physics. The explanation of the observed negative magnetoresistance (NMR) in disordered metals and degenerate semiconductors in weak magnetic field is based generally on the weak localization (WL) phenomenon [1,2]. The WL corresponds to the enhancement of the back scattering probability of the electrons due to the interference by partial electron waves traveling along time reversed electron-path. Constructive interference will occur even in the presence of elastic scattering since this type of scattering does not influence the phase coherence of the electrons. The scattering events, which affect the wavelength of the electronic wave function, are mainly the electron–electron and electron–phonon scattering. An externally applied magnetic field suppresses the phase coherence leading to a negative magneto-resistance.

In addition, during the last decades, there has been a rapid increase of research in the field of porous silicon (PS) and related applications [3,4]. The electronic transport properties of PS are premised on an assumed geometrical structure and here lie a major difficulty. PS is a complex material which one can characterize by a hierarchy of disorders. One feature that strongly affects the electronic transport properties of PS is the inhomogeneous nature of the material. Crystallites and pores in the PS network have a fractal like structure. The PS material is highly irregular in the sense that the crystallites which are randomly distributed have different sizes. This leads to band-gap variations from one particle to another in nanoporous silicon (nano-PS). These band gap fluctuations would cause localization of the electronic states in the band edges and also to the creation of surface states having energy distributed in the band gap. The observation of strong visible photoluminescence in PS at room temperature [5] has caused considerable interest. This phenomenon is believed to be attributed to quantum confinement effect in thinned silicon columns which probably represent the network of one-dimensional (1D) quantum wires (QW) [5,6]. Thus, simulations about PS have mainly focused on the investigation of silicon QW [7–11]. The use of the QW can well simulate the nanostructure of PS. However, the observed NMR in the present work, at room temperature can be explained well on the basis of the model of weak localization in 1D system because the existence of disorder is believed to be of key importance for the WL in PS. We are not aware of similar studies on the NMR effect in PS at room temperature.

In this work, we report the result of studying the Current–Voltage (I–V) characteristic of the aluminum/porous silicon/sili-

* Corresponding author at: Laboratoire Matériaux Avancés et Phénomènes Quantiques, Faculté des Sciences de Tunis, Université El-Manar, 2092 Campus Universitaire Tunis, Tunisia. Tel.: +216 71 256 425.

E-mail address: Samir.Romdhane@fsb.rnu.tn (S. Romdhane).

0038-1101/$ - see front matter
http://dx.doi.org/10.1016/j.sse.2012.07.019

con/aluminum (Al/PS/Si/Al) structure under application of a static magnetic field. One expects magneto-resistance to be related to porous layer of silicon. We show that the observed NMR in PS and the magnetoconductance ($\Delta G = [G(H) - G(H = 0)]$, where $G(H)$ is the conductance I/V at magnetic field H) are described by a one-dimensional WL effect.

2. Experimental

2.1. Realization structures

The PS layers were formed from (1 0 0) oriented single side polished p-type (boron doped) silicon. The resistivity of the silicon wafer is about 1 Ω cm and its thickness of 400 μm. We used the electrochemical anodization method (anodization time was fixed to 25 min) to form the PS layer on the front side of the sample. The applied current density was equal to 10 mA/cm^2. The basic anodization solution is composed by HF (20%) and pure ethanol in the proportion 1:1. After, the sample is dried under nitrogen flux. A thick aluminum (Al) layers, about 2 μm thick, was thermally evaporated on the back surface of the sample and on the PS layer. The Al contacts were fired at 500 °C, in an infrared furnace for several minutes, to ensure ohmic contacts. The PS layer thickness (around 10 μm) and its porosity (~62%) were estimated by the gravimetric method. The surface of the PS layer is about 0.5 cm^2.

2.2. Electrical characteristics under application of a static magnetic field

The *I–V* characteristics of the Al/PS/Si/Al structures were measured at room temperature using a KEITHLEY 2400 source-meter. All measurements were computer controlled. The device structure is shown in Fig. 1.

During the MR measurements, the device is placed between the pole pieces of electromagnet (BRUKER: MAGNET B–E 10) whose magnetic field intensity H can reach 1 T. The direction of the magnetic field is perpendicular to the direction of the current flow and its intensity is measured by means of a BRUKER-DIGIGAUS 2 gaussmeter.

3. Experimental results

We have achieved *I–V* measurements of the device in a transverse magnetic field with different values of the static magnetic field ranging from 0 to 8000 G. In Fig. 2, we give a set of two *I–V* characteristics corresponding to 0 G and 8000 G. Regarding these *I–V* characteristics, one can notice that for a fixed bias voltage the measured current is enhanced when the transverse magnetic field increases.

The experimental results indicate a small-magnitude negative magnetoresistance ($\Delta R/R = [R(H) - R(H = 0)]/R(H = 0)$, where $R(H)$ is the two-terminal resistance (V/I) at magnetic field H which depends heavily on the applied bias voltage. Fig. 3 shows the MR at room temperature, with magnetic field strength varied from 0 G to 8000 G of the PS/Si heterojunction, for different values of the bias voltage. The measured NMR increases with magnetic field

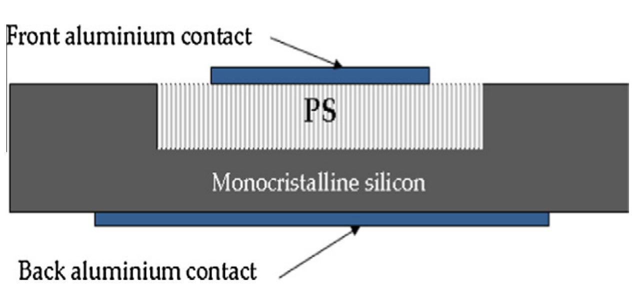

Fig. 1. Schematic representation of the used device.

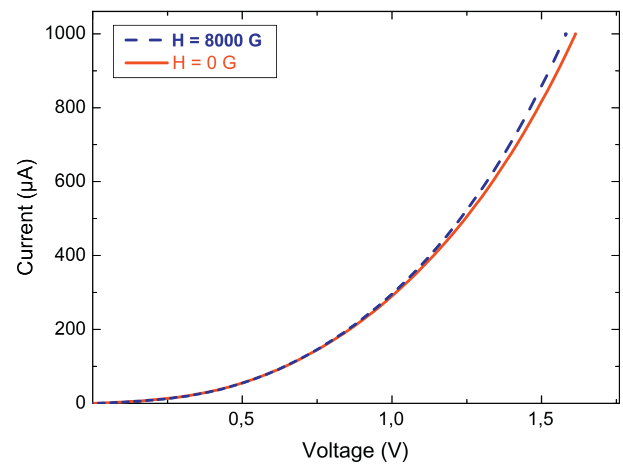

Fig. 2. *I–V* characteristics of Si/SP heterojunction at room temperature, for two values of the applied magnetic field *H*.

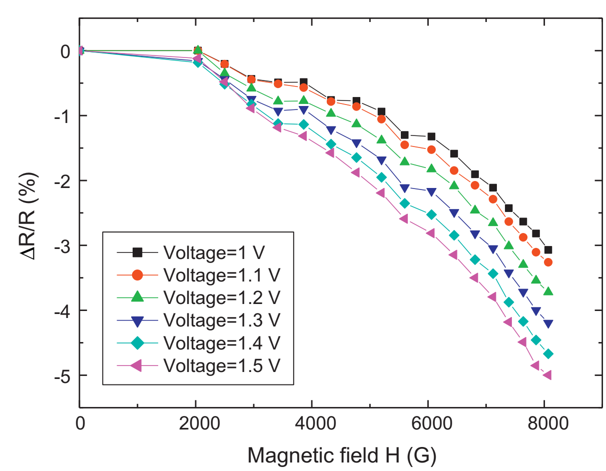

Fig. 3. Two-terminal MR line shapes of the PS/Si heterojunction, for various voltage values, at room temperature.

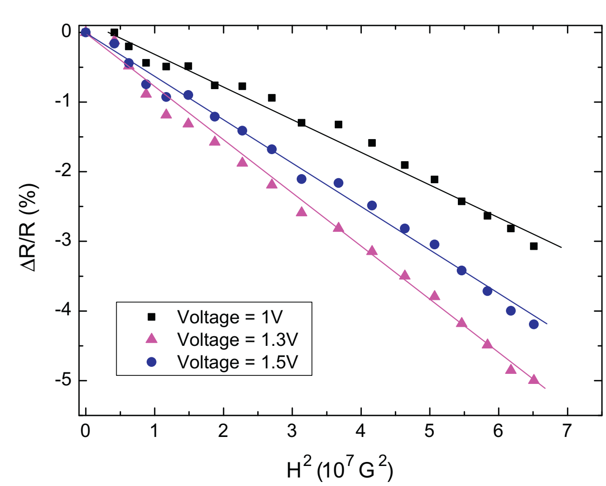

Fig. 4. $\Delta R/R$ (H^2) for applied voltage 1, 1.3 and 1.5 V.

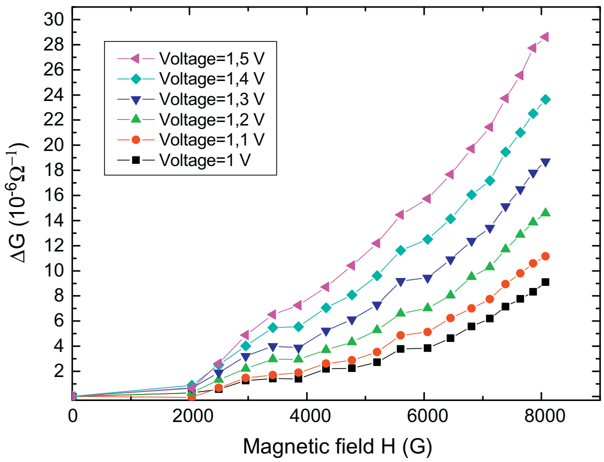

Fig. 5. $\Delta G(H)$ measured at room temperature in a PS/Si heterojunction for different voltage.

strength H and attains a maximum of 5% at 8000 G for an applied voltage of 1.5 V.

Fig. 4 shows a plot of the magnetoresistance $\Delta R/R$ as a function of H^2 where the results yield a good fit to a quadratic field dependence at low magnetic field. Therefore increasing the applied voltage brings about a decrease in the NMR magnitude.

We also represented the dependence of the magnetoconductance (MC) of the PS/Si structure on the magnetic field, at room temperature, for different voltage (Fig. 5). The experimental results show that there is always small-magnitude positive MC. The ΔG attains a maximum of $30 \times 10^{-6}\,\Omega^{-1}$ at 1.5 V for $H = 8000$ G. Fig. 5 shows that MC increases with increasing voltage.

4. Discussion

Measured I–V characteristics may be governed by either PS/Si heterojunction or PS/Al interface, or both. It is known that I–V characteristics of the metal/PS/p-Si/metal structure are essentially nonlinear [12]. Because of the band gap difference between PS and Si substrate, a heterojunction is formed at the interface [13]. The diode rectification behaviour observed for the I–V characteristics in the Al/PS/Si/Al device has been attributed to the junction between the silicon and porous silicon interface [14] because the Al/PS junction does not exhibit a Schottky junction but an ohmic contact. However, an energy band diagram around the interface between PS and bulk Si was also used to analyze PS device. It was supposed that PS has a band gap of about 1.8 eV. Furthermore, the interface of PS and bulk Si (with a band gap of about 1.12eV) has been postulated with band discontinuities occurring at both valence and conduction bands [15]. By study the conductance in the PS/Si heterojunction, it was established that the conduction is due to bulk processes in PS rather than to junction proprieties [16]. As by study the I–V characteristic corresponding to the different external bias applied to metal/PS/Si/metal structure, those authors assumed that the total current of devices was governed by the carrier transport in the high resistivity PS layers [15]. Therefore for the study of the magnetic and electrical transport through the metal/PS/Si structure, it is necessary to ensure that the current in the structure is controlled by the porous material and not by the Schottky barrier at the metal/PS interface [17,18]. Also we take into account that the conductance in PS/Si heterojunction is decreasing with increasing of the porosity of PS layer [14].

MR can be explained with two mechanisms: (i) Lorentz force and (ii) weak localization. It appears that in our study the mechanisms (i) cannot explain the observed MR effect, because it leads to exclusively positive magnetoresistance, whereas the measured effect is typically negative. We note that the observed MR traces resemble closely to MR traces due to WL in diffusive regime [19]. The WL, which is a result of constructive interference of two electron waves traveling along a closed path in opposite direction and scattering by the same impurities, leads to an additional contribution to the resistance. In the presence of external magnetic field H, the partial waves describing the buckles in the inverse sense are dephased. This occurs when the electron returns to its original point 0, by $\Delta\phi = e\phi/\hbar$ where ϕ is the flux of magnetic through the buckles, e the elementary charge and $\hbar$ the reduced Plank's constant. Then, the magnetic field destroys the phase coherence, which is the origin of constructive interference leading to localization. The application of the magnetic field leads to a delocalization and to a negative magnetoresistance (NMR). The phase coherent effect of electrons scattered by impurities plays an important role in disordered systems. The negative magnetoresistance [20], and mesoscopic effects [21] originate from the interference of electron wave. After discovery of WL, the theory of the NMR was elaborated, in which it was established that NMR occurs due to the destruction of the quantum corrections to the conductance by the external magnetic field which affects the phase coherence. In inhomogeneous system, it is possible to observe an NMR at too much higher temperatures (even to near room temperature) because the phase coherence of the carriers is preserved to higher temperatures [22].

Let us first consider the effect of phase coherence on transport mechanisms. The theory of quantum corrections to the conductivity of thin 1D wires was formulated in the mid-1980s [23]. However, to this day not all of the basic assumptions have been checked experimentally in detail. One such problem is the behavior of the resistance in magnetic field. It is well known that the effect of a magnetic field on the interference correction to the conductivity of disordered metals and degenerate semiconductors leads to a negative magnetoresistance in weak fields [1]. We consider weakly disordered system, for which the mean free path le is much larger than the distance between electrons: $k_F l_e \gg 1$, where k_F is the Fermi wave vector. Quantum interferences are responsible for a small reduction of the Drude–Boltzmann conductivity called the "WL correction". This correction is a manifestation of quantum coherence which is always limited over a certain length scale, named the phase coherence length L_φ. A way to extract this important length scale in experiments is to use the magnetic field sensitivity of the WL. The structure of this classical conductivity is given by a sum of probability intensities $\sigma_{cl} \propto \sum_j |A_j|^2$ where A_j represents some amplitude related to a diffusion process. However, one knows that in quantum mechanics one must add amplitudes instead of intensities. Thus, the structure of the conductivity has to be $\sigma \propto \sum_{i,j} A_i A_j^*$. Since the interference terms, of the form $A_i A_j^*$, have random phases they cancel in average so that the conductivity reduces to its classical value given by the diagonal terms in the sum. However, there is a class of contributions which may not cancel in average. They correspond to diffusive trajectories which form closed loops. Such a loop can be traveled in clockwise or anti-clockwise directions. If there is time reversal symmetry, both trajectories, j and its time-reversed jT, have same action, so that they interfere constructively. As a result, in addition to the classical average conductivity, there is a correction of the form $\Delta\sigma \propto \langle \sum_j A_i A_{j^T}^* \rangle$ where the sum extends over the closed trajectories. The sign of the correction is negative because the trajectories j and j^T have opposite moments. This reduction in conductivity by interference effect is called the WL correction. This is a phase coherent effect because it implies that the time reversed trajectories have the same action. This phase coherence is broken by inelastic events due to the coupling to other degrees of freedom or due to electron–electron interactions. Such coherence breakdown is temperature dependent, and can be phenomenologically described by a phase coherence length L_φ. Value of L_φ, decreases very slowly with temperature and, as long as $L_\varphi > l_e$, WL remains possible, thus allowing the NMR to be observed even at room temperature [22]. The effect of a magnetic field which breaks time-reversal symmetry is to destroy this phase-coherent contribution to the conductance, leading to an NMR (in the absence of spin–orbit scattering) which is one of the most spectacular signatures of phase coherence. In the middle of the 80's, the progresses in nanolithography allowed to realize not only wires of mesoscopic sizes (μm) but also networks of wires, whose more complicate topologies make them particularly suitable to study interference effects. Comparing the interference correction to the conductance, to the correction due to the interaction shows that in a three-dimensional (3D) system the effect due to the electron–electron interaction predominates, for a 2D system both corrections are of the same order of magnitude, and in a 1D metal the interference correction predominates [24]. Quantum corrections have been most actively investigated in 2D and 3D systems. The quantum correction to the conductance in PS can be explained within the 1D WL theory.

For an isolated quasi-1D wire, with a finite length L; the correction to the conductance, in the absence of magnetic field and expressed in term of quantum conductance e^2/h, in the WL regime [20] is given by:

$$\Delta G_{\mathrm{WL}} = -\frac{e^2}{\pi\hbar}\frac{L_\varphi}{L} \quad (1)$$

This result corresponds to the case where phase coherence length is smaller than the system size ($L_\varphi < L$). Experimentally, modification of an external parameter such as temperature or magnetic field can be used as a tool to measure this correction.

Since weak localization gives a negative contribution to the conductance, its suppression by a transverse magnetic field leads to an increase in conductance, which corresponds to a negative magnetoresistance (NMR).

The correction to the conductance in a quasi-1D wire in a transverse magnetic field H is [2,20]:

$$\Delta G_{\mathrm{WL}}(H) = -\frac{e^2}{\pi\hbar L}\left(L_\phi^{-2} + \frac{W^2}{3l_H^4}\right)^{-1/2} \quad (2)$$

where $l_H = \sqrt{\frac{\hbar}{eH}}$ is the magnetic length and W is the radius of the wire.

Since $G(0)$ (zero field conductance) is field independent, the magnetoconductance $\Delta G(H) = G(H) - G(0)$ is the difference between the weak localization corrections $\Delta G_{WL}(H) - \Delta G_{WL}(0)$. Using relations (1) and (2), the magnetoconductance becomes:

$$\Delta G(H) = \frac{e^2}{\pi\hbar L}\left[L_\phi - \left(L_\phi^{-2} + \frac{W^2}{3l_H^4}\right)^{-1/2}\right] \quad (3)$$

In PS layer, we have assumed the conductance to be determined by carrier diffusion along quasi-1D trajectories bounded by the walls of pores. Furthermore, we assume that all QWS are the same size.

The charge transport in porous silicon is ensured through several interconnections of QWs. The electrical conductivity is carrier concentration dependent. Due to the broadening of the gap, the carrier concentration may be given by:

$$p_{Qw} = N_V exp\left(-\frac{E_F - E_V}{k_BT}\right) \quad (4)$$

where E_F is the Fermi level, E_V is the new position of the valence band in QW, k_B is the Boltzmann constant, T the temperature and N_V is the holes effective density of states.

Following the quantum confinement theory and assuming that in QW the holes mobility is the same as in bulk silicon, the QW electrical conductance can be given by:

$$G_{QW} = ep_{QW}\mu_{Si} = G_{Si}\exp\left(-\frac{\Delta E_V}{k_BT}\right) \quad (5)$$

where $\Delta E_V = E_V - E'_V$.

The total broadening of the band gap in the PS layer can be expressed as:

$$\Delta E = \Delta E_C + \Delta E_V \quad (6)$$

where ΔE_C and ΔE_V are the displacements of the conduction and valence band edge.

Several models have been proposed to explain the increase of the band gap of porous silicon. The original and most prevalent has been the quantum confinement model. Hence, the increase in the band gap is due to the confinement of holes in Si nanocrystallites in PS. In a first-order approximation, the increase in valence and conduction band energies is given by [6]:

$$\Delta E_{C,V} \propto \frac{h^2}{4m^*_{e,h}d^2} \quad (7)$$

where $m^*_{e,h}$ is the effective masses of electrons and holes, $d = 2$ W is the crystallite diameter and h is the Plank constant. We assume equality between electron (τ_e) and hole (τ_h) life times [14], and we suppose that they are not affected by the quantum confinement effect, such simplifications yield to:

$$G_{QW} = G_{Si}\exp\left(-\frac{\Delta E}{4k_BT}\right) = \lambda G_{Si} \quad (8)$$

The used PS layer has a porosity of 62%, the corresponding [25] ΔE value is 0.75 eV.

Taking into account the quantum confinement (8) and the WL correction (3), MC is then given by:

$$\Delta G(H) = \lambda\frac{e^2}{\pi\hbar L}\left[L_\phi - \left(L_\phi^{-2} + \frac{W^2}{3l_H^4}\right)^{-1/2}\right] \quad (9)$$

At each voltage, a value of the phase coherence length can be extracted from the fit to Eq. (9). To fit experimental curves, we have considered the scheme of Fig. 6 as a simple architecture to model the PS structure. Expression (9) was obtained for an isolated wire with length $L = 10$ μm (thickness of the porous layer) and radius $W = 1.5$ nm.

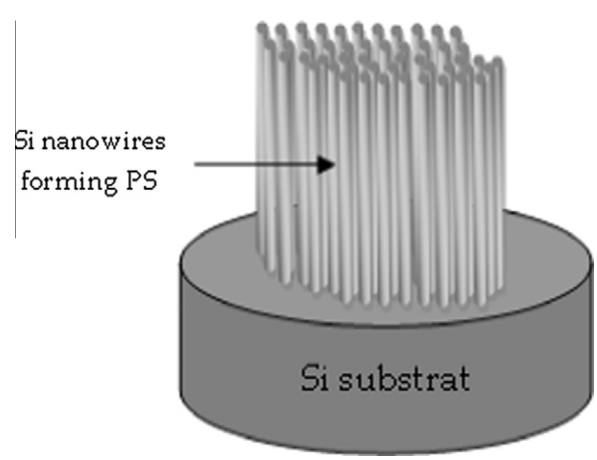

Fig. 6. Adopted architecture to model the PS structure.

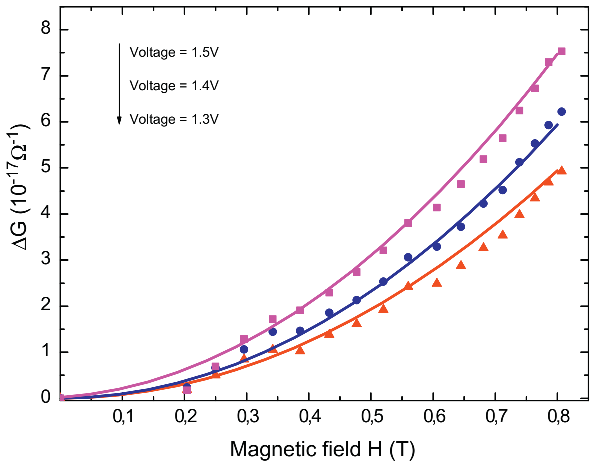

Fig. 7. $\Delta G(H)$ of one nanowire at voltages 1.3, 1.4 and 1.5 V. Solid colored curves are fits to Eq. (9). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Taking for the total conductance of the porous silicon and a large number of wires, we find that approximately 3.8×10^{11} wires per $7 \times 10^{-2}\,\text{cm}^2$ connected in parallel participate in the conduction. In Fig. 7, we show the average of the MC of one wire in PS (through dividing by the number of wires), solid lines represent good fits to 1D weak localization theory for 1.3, 1.4 and 1.5 V bias voltage at room temperature. The fit gives$L_\phi = 3.15 \pm 0.04$ nm, $L_\phi = 3.33 \pm 0.05$ nm and $L_\phi = 3.61 \pm 0.03$ nm respectively for 1.3, 1.4 and 1.5 V. These values obtained for L_ϕ do indeed satisfy the criterion for the interference correction to be a 1D character in the diffusive regime:.$L > L_\phi > 2W$.

5. Conclusion

We have described measurement of variation in the resistance of PS/Si heterojunction as a function of an applied magnetic field. We observed an NMR in PS elaborated in p-type boron doped monocrystalline silicon. The magnitude of this effect increases to 5%, at room temperature for magnetic field up to about 8000 G. Production of semiconductor materials with NMR value in the room temperature represents a special interest from the viewpoint of designing new class of magnetic devices. It was found, quite surprisingly, that for porous silicon system, an NMR could be observed even at room temperature, and that the MC curves for all samples, fit with good precision the conventional 1D weak localization theory. This makes the object attractive for investigations of electric and magnetic phenomena in low-dimensional disordered structures.

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