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Thermoelectric Properties of Doped Zigzag Silicene Nanoribbons

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Thermoelectric properties of silicene nanoribbons doped with magnetic impurity atoms are investigated theoretically for both antiparallel and parallel orientations of the edge magnetic moments. Spin density distribution and transport parameters have been determined by *ab-initio* numerical methods based on the density functional theory. Doping with magnetic atoms considerably modifies the spin density distribution, leading to a ground state with a non-zero magnetic moment. Apart from this, the spin thermopower can be considerably enhanced by the impurity atoms.

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1. Introduction

Recently, one can observe an increasing interest in silicene – a two-dimensional crystal of Si atoms on a honeycomb lattice. Unlike graphene, atomic structure of silicene is buckled, with the two triangular sublattices slightly shifted in vertical direction. Silicene is a zero-gap semiconductor with two Dirac cones at the K and K' points of the hexagonal Brillouin zone. Due to the buckled structure, transverse electric field can open an energy gap [1]. Ab-initio numerical calculations show that the electrical field also leads to two gapped Dirac cones corresponding to spin polarized states. Accordingly, an effective silicene-based spin filter has been proposed, in which current spin polarization can be controlled with an electric field [2]. Thus, silicene may be of some importance for future applications in electronic and spintronic devices.

Quasi-one-dimensional silicene nanoribbons (SiNRs) have also been fabricated [3], and their electronic as well as magnetic and mechanical properties have been extensively studied by *ab-initio* methods [4, 5]. Silicene nanoribbons can exhibit semiconducting or metallic character, similarly to graphene ones. Theoretical investigations have shown that nanoribbons of armchair type (aSiNRs) are nonmagnetic, while those of zigzag type (zSiNRs) exhibit a magnetic ground state, with opposite magnetic moments at the two edges (AFM state). This is similar to zigzag graphene nanoribbons, where magnetic moments at one edge are opposite to those at the other edge [6]. Ferromagnetic state (FM) in which edge moments are parallel, has a slightly higher energy, but it can be stabilized by an external magnetic field or by the proximity effect. Generally, zSiNRs with AFM ordering have semiconducting properties, with a relatively wide energy gap, whereas zSiNRs in the FM state are metallic. Accordingly, a giant magnetoresistance associated with transition from the FM to AFM state has been predicted [7]. The influence of doping on electronic structure and magnetic properties of zSiNRs has been intensively studied using the ab-initio approach based on the density functional theory (DFT) [4, 8]. Spin gapless semiconductor behavior with practically 100% spin polarization has been predicted for zSiNRs doped with B/N atoms at the edge positions [9]. Introduction of a B-N pair into zSiNRs also leads to spin gapless semiconductor [10]. Apart from this, first-principle calculations show that the presence of Co atoms can suppress the edge moments in the vicinity of Co impurities [11].

Very recently, thermoelectric phenomena in SiNRs have been studied in both pristine [5] and doped nanoribbons [8]. The subject is of current interest due to the possibility of converting dissipated heat to electrical energy at nanoscale. Thermoelectric properties, especially the Seebeck coefficient S describing thermally-generated electrical voltage, significantly depend on the electronic band structure and are enhanced when the Fermi level is in an energy gap. Accordingly, a very remarkable thermopower S, exceeding 1 mV/K, has been predicted for semiconducting zSiNRs in the AFM state [5]. In turn, the thermopower in the FM state is strongly reduced, so the transition from the AFM to FM state leads to a considerable magnetothermopower [5]. In ferromagnetic materials, in which the two spin channels are different and independent, spin related thermoelectric phenomena resulting from an interplay of charge, spin and heat transport can be observed in addition to the conventional ones. In particular, it is possible to observe spin Seebeck effect, which corresponds to the thermal generation of a spin voltage [12]. Ab-initio calculations performed for zSiNRs doped with Al/P atoms show that the spin Seebeck coefficient can be remarkably enhanced by im-

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purities [8]. This enhancement depends on the impurity position with respect to the nanoribbon edges. In the present paper we consider the effect of a magnetic impurity on transport and thermoelectric phenomena in zSiNRs with edges terminated with hydrogen atoms. Spin related thermoelectric effects are also studied, and the spin Seebeck coefficient is determined numerically.

2. Computational details

Electronic transport through zSiNRs was investigated by *ab-initio* DFT calculations within the Siesta code [13]. The spin-resolved energy-dependent transmission, $T_\sigma(E)$, through the zSiNRs was calculated in terms of the non-equilibrium Green function method (NGF) as implemented in the Transiesta code [14]. All structures were optimized until atomic forces converged to 0.02 eV/A. The generalized gradient approximation (GGA) with Perdrew-Burke-Ernzerhof parameterization was used for the exchange-correlation part of the total energy functional [15]. The width of a zSiNR is characterized by the number N of zigzag chains in a ribbon.

In systems in which spin channels are not mixed in the nanoribbon, the conventional S_c and spin S_s thermopowers can be defined as $S_c = -\Delta V/\Delta T$ and $S_s = -\Delta V_s/\Delta T$, respectively. S_c and S_s can be determined under the conditions that charge and spin currents are equal to zero. Then, in the linear response regime S_c and S_s are given by $S_{c/s} = -\frac{1}{2|e|T}(L_{1\uparrow}/L_{0\uparrow} \pm L_{1\downarrow}/L_{0\downarrow})$ Here e is the electron charge, while $L_{n\sigma}$ $(n = 0, 1, 2)$ are defined as $L_{n\sigma} = -\frac{1}{h}\int \mathrm{d}E\, T_\sigma(E)\,(E-\mu)^n \frac{\partial f}{\partial E}$, with $f(E)$ being the Fermi-Dirac distribution function corresponding to the chemical potential μ and temperature T [12].

3. Results

Numerical calculations have been performed for zSiNRs with $N = 6$ and doped with Fe atoms placed at one of the edges (PE configuration). The ground state of the system corresponds to ferrimagnetic ordering, with antiparallel magnetic moments of the two edges. However, the edge moments do not compensate each other as a large moment appears on the Fe atom and also on Si atoms in its close vicinity (Fig. 1a). The system exhibits then semiconducting character with a relatively wide energy gap. The charge thermopower is strongly enhanced due to the gap and is close to 1mV/K. The dependence of the thermopower on the chemical potential is very similar to that obtained for zSiNRs doped with Al/P atoms in the edge positions [8], so we do not present it here.

Energy of the FM state (see Fig. 1b) is slightly higher than that of the ferrimagnetic state. Magnetic moments in this state are parallel and they are mainly localized on the edge atoms. Similarly as in the ferrimagnetic case, a large moment appears on the Fe atom and the considerable moments are also distributed in its vicinity. Below we discuss in detail thermoelectric properties of the FM state. This configuration can be stabilized by an external magnetic field, by exchange coupling to a ferromagnetic substrate or by coupling to ferromagnetic contacts. The

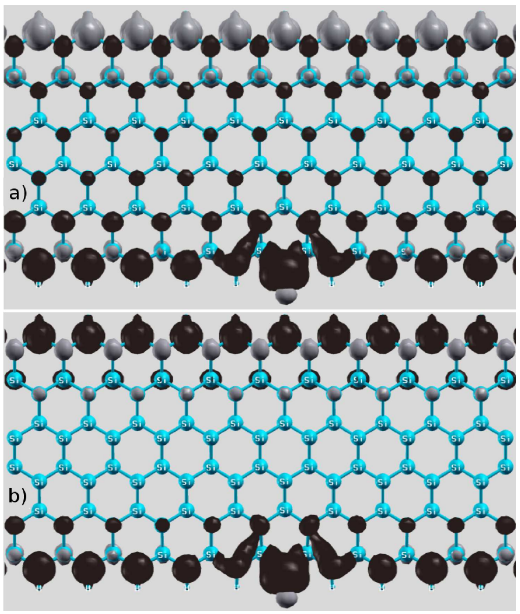

Fig. 1. Spin density in the low energy state (a) and in the FM state (b), calculated within GGA approximation for zSiNRs with $N = 6$. The black and gray dots correspond to spins oriented in the opposite directions.

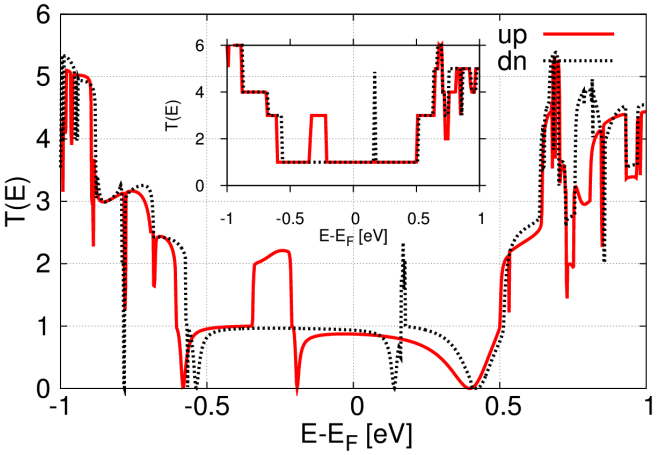

Fig. 2. Spin-dependent transmission in the FM state of the zSiNR with $N = 6$, doped with Fe atoms. The inset shows the transmission calculated for the corresponding pristine zSiNR.

spin-dependent transmission function, $T_\sigma(E)$, calculated for the FM state is presented in Fig. 2 as a function of energy (measured from the Fermi energy E_F of the corresponding pristine nanoribbon). The system shows metallic character with almost constant and spin-degenerate transmission in the vicinity of the Fermi energy. The pronounced spin dependence appears for energies lower than E_F, where a dip with zero transmission, followed by a broad maximum, appears in the spin-up channel, whereas the transmission is constant for the spin-down channel. Similar behavior can be observed in the spin-down channel for $E > E_\mathrm{F}$, but a very narrow peak appears instead of the broad maximum. For higher values of

energy, wide dips with zero transmission appear in both spin channels, but they are slightly separated in energy. The calculated transmission, especially the appearance of dips in both spin channels for $E > E_F$, is similar to that obtained for zSiNRs doped with Al in the PE configuration [8]. Thus, the Fe atom in Si lattice can behave like impurity of p-type. Detailed calculations show that the good consistency of the results obtained for zSiNRs with Fe and Al impurities results from the good matching of the spin-dependent band structure in a wide range of energy near E_F for both types of impurities.

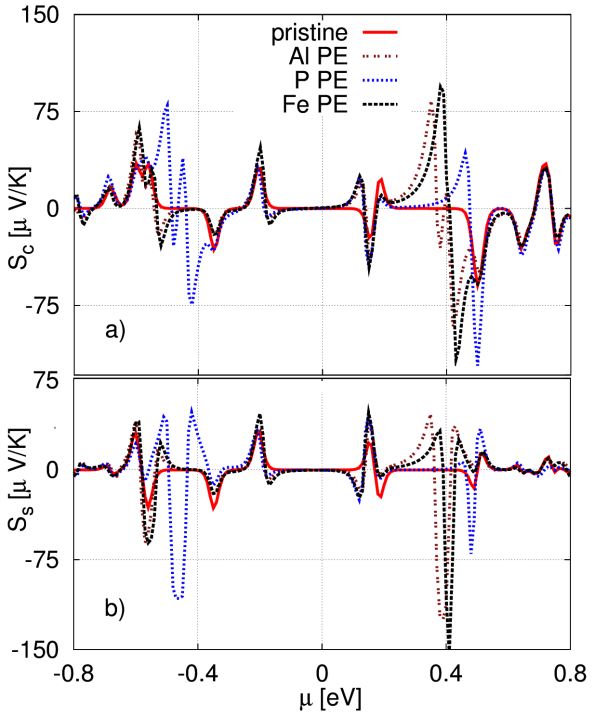

Fig. 3. Charge (a) and spin (b) thermopowers as a function of the chemical potential μ in the FM state of zSiNRs doped with Fe impurities, compared with the corresponding results for pristine zSiNRs and those doped with Al and P impurities obtained in [8].

Now, assuming that spin relaxation processes are weak and do not mix the two spin channels, we discuss the spin effects in the thermopower. Conventional S_c and spin S_s thermopowers vs chemical potential are shown in Fig. 3. For comparison, the curves calculated for the pristine zSiNR and zSiNRs doped with Al and P atoms in edge positions are presented there. Due to metallic character of zSiNRs, both thermopowers are strongly reduced for small values of $|\mu|$. Considerable enhancement of S_c/S_s is obtained for $\mu \approx 0.4$ eV. This enhancement is correlated with the appearance of well defined dips in the transmission function, and results from steep changes of the transmission near the dip edges. Similar changes of S_c/S_s can be observed for Al impurity, as in both cases transmission exhibits similar behavior. However, peaks in S_c/S_s for zSiNRs with Fe impurity are more pronounced than in the case of Al impurities.

4. Concluding remarks

The presented results show that both transmission and charge/spin thermopowers are not crucially dependent on the impurity atom. Though, introduction of the Fe atom strongly changes the spin density, it does not affect the thermoelectric phenomena considerably. In particular, the spin thermopower is similar to that obtained for Al impurities. In both cases the spin thermopower is enhanced and exhibits a well pronounced peak for positive values of the chemical potential. This weak dependence on the type of impurity results from the good matching in both systems of the spin-dependent electronic bands in a wide region near E_F.

Acknowledgments

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