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Energy gap in graphene and silicene nanoribbons: a semiclassical approach

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Abstract. We present a semiclassical approach based on recently reported Fermi velocity either graphene [1] and silicene [2] to explore the energy gap and band dispersion when graphene (silicene) is laterally confining at nanometer size, i.e., graphene nanoribbons (GNRs) and silicene nanoribbons (SiNRs). The modeling approach matches previous experimental and theoretical works for GNRs featuring widths ranging from 1 to 20 nm. Furthermore, the proposed approach results coherent with experimental measures on GNRs up to 100 nm wide. For this reason, we extend the modeling approach to SiNRs. A remarkable feature, in SiNRs, is an energy gap opening which is lower concerning GNRs due to a reduced Fermi velocity in silicene. SiNRs are expected to be a good competitor with GNRs for digital electronics.

I. INTRODUCTION

Graphene, a one-atom-thick two-dimensional (2D) carbon sheet, has attracted much interest since its successfully single-layer isolation from graphite, in 2004 [3] and due to its unique and [4]. The properties of graphene arise from its semimetallic band structure just at the Fermi level [5], with the valence (π), and conduction (π^*) bands exhibiting a conical dispersion vs. the in-plane momentum at the six corners of the hexagonal first Brillouin zone, i.e., the Dirac K or K' points [4]. This key feature makes graphene a zero-energy gap material at room temperature. The gapless band structure of graphene is a significant hindrance since it limits on/off ratio to enable digital electronics [6]. Nevertheless, graphene appears as a promising nanomaterial for future generations of nanoelectronics [7, 8], radio frequency devices [9], thermally and electrically conductive reinforced composites [10], sensors [11], transparent electrodes for displays or solar cells [12, 13], among others.

It is worth noting that there exists incompatibility of carbon-based materials (e.g., graphene [14, 15], graphene oxide [16, 17, 18, 19], carbon nanotubes [20] and fullerenes [21, 22]) with the current silicon-based electronics. This incompatibility of technologies makes it infeasible to use graphene in high-tech industrial applications. For this reason, other materials, especially those in the IV group, e.g., silicon and germanium in a one-atom-thick 2D lattice, named silicene and germanene, nowadays are further investigated [23, 24, 2]. Similarly to graphene, silicene and germanene show a conical dispersing π and π^* bands, crossing at the Dirac points, where charge-carriers behave as massless Dirac's fermions. It is worth heeding that silicene has four principal advantages over graphene: *i*) obvious compatibility with current electronics, *ii*) a large spin-orbit induced energy gap of 1-10 meV [25, 26], with respect to the 10^3 meV value predicted for graphene [27], *iii*) tunable energy gap in the presence of a perpendicular electric field or by chemical functionalization [28, 29], and *iv*) synthesis on silver substrates with slight affected semimetallic properties [30]. For those reasons, silicene is expected to grow its participation in the electronics market and goes on par with carbon-related materials.

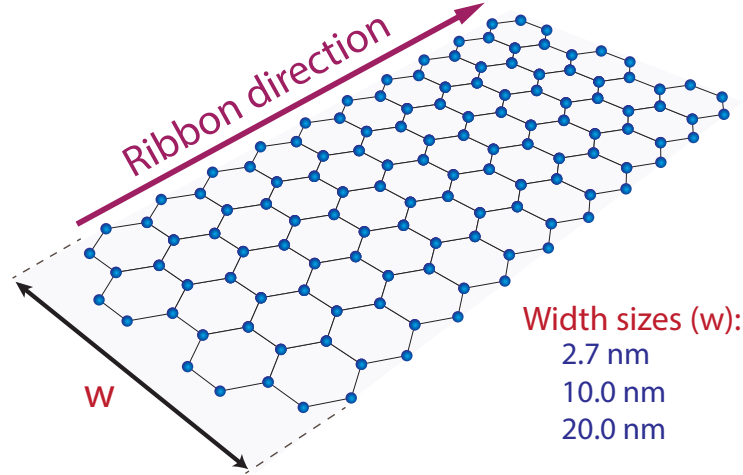


FIGURE 1. Schematic of a nanoribbon. Ribbons of 2.7 nm, 10 nm and 20.0 nm wide are selected to analyze the energy gap and band dispersion in graphene and silicene nanoribbons.

Despite that both graphene and silicene have attracted significant attention in recent years; a non-zero energy gap is necessary, on these honeycomb-like structures, for manufacturing electronic devices [31]. Nevertheless, when graphene (or silicene) is laterally confining at nanometric scales (< 20 nm), see Figure 1, quasi-one-dimensional structures called graphene nanoribbons (GNRs) appear. Considering the atomistic and structural conformation, GNRs show zigzag or armchair shaped-edges [32, 33, 34, 35]. At first approximation, GNRs could be seen as rectangular ultra-thin strips with which it is possible to vary their widths without considering the edges. Some earlier studies show an energy gap opening inversely proportional to the ribbon width [32, 36, 37]. This idea has motivated efforts to develop scalable methods of producing homogeneous and ultra-narrow GNRs [38, 39], such as by cutting mechanically exfoliated graphene [40], or by patterning epitaxial grown graphene [41].

From the theoretical point of view, tight-binding model within the Huckel approximation [42] has predicted that all GNRs to be metals. Whereas, density functional theory (DFT) calculation showed that GNRs with armchair shaped-edges have a finite energy gap, and GNRs with zigzag shaped-edges have a non-zero energy gap when spin polarization is considered [32, 43]. However, it is well-known that DFT calculation is inadequate to predict the energy gaps of nano-sized semiconductors [44], but appropriate for describing the electronic properties of (semi)metallic systems [45, 46, 47]. On the other hand, the first-principles many-electron Green's function approach within the GW approximation [48] has been shown to be reliable for obtaining energy gaps of GNRs up to 2.4 nm wide, taking into account the armchair or zigzag structural conformation.

The approaches mentioned above cannot be extended to wider ribbons due to the vast numbers of atoms involved, and since those calculations consider each atom, its create a time-cost computational problem challenging to address. Motivated by the lack of a viable theoretical framework to analyze the energy gap of recently synthesized GNRs [38, 39], in this work, we show a simple semiclassical approach [49], using a newly reported velocity of charge-carriers in graphene (and silicene) in literature, that help to predict the energy gap of GNRs. Our results are in agreement with previous works on both experimental and theoretical, for GNRs ranging from 1 to 20 nm wide, and reasonably describes the experimental observation for GNRs up to 100 nm wide. Thus, we use this method to compare and predict the energy gap and band dispersion of silicene nanoribbons (SiNRs) with similar widths to GNRs.

II. SEMICLASSICAL APPROACH

When graphene (silicene) is laterally patterned at nanometric scale, the charge-carriers are confined in quasi-one-dimensional structures, where only a few sub-bands are involved in the charge-transport. This case can be interpreted

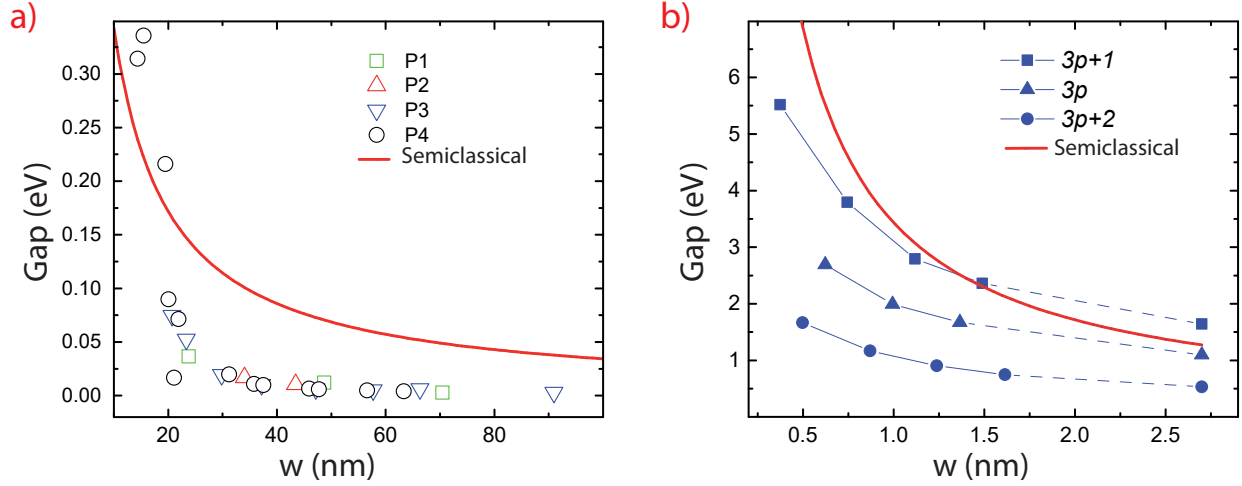


FIGURE 2. Energy gap (eV) vs ribbon width (nm). (a) Semiclassical approach compared to the energy gaps (P1-P4) measured on parallel GNRs [37]. (b) Semiclassical approach compared to the energy gaps predicted in three families of GNRs by using GW approximation. [48]. The dashed lines represent an extrapolated data.

as a quasi-hard-wall boundary condition [49, 48] which allows the energy gap opening in GRNs, independent of chirality (armchair or zigzag shaped-edges).

Previous works already demonstrated that the energy gap in GNRs increase as decrease ribbon width. In fact, for a relatively wide GNR, the charge-carriers move through the ribbon direction, Figure 1, characterized by the velocity of the π and π^* graphene electrons [50, 49]. For that, the energy gap for ribbons of graphene and silicene reduces to:

$$\Delta E = h \frac{V_F}{w}, \quad (1)$$

where the Planck's constant h equals the relation of Eq. 1, V_F is the Fermi velocity of charge-carriers in graphene or silicene, and w represents the ribbon width.

Recently, the V_F of graphene has been (re)evaluated, by DFT calculation, showing a value of 0.83×10^8 cm/s [1], which is lower than the previously reported value of $\sim 1.09 \times 10^8$ cm/s [4] and $\sim 0.85 \times 10^8$ cm/s [51]. Similarly, the V_F of silicene has been predicted with a value of 0.64×10^8 cm/s [2]. It is worth noting that the armchair or zigzag conformation is not considered in Equation 1. This expression is an analytical relation which confirms that all GNRs are semiconductors [see Figure 2 (a), (b)], as it was observed in *GW* approaches [48].

Following with the idealized case of a quasi-hard-wall condition [50, 49], the electronic band dispersion of a one-dimensional system is:

$$E = \pm \frac{\Delta E}{2} \sqrt{n^2 + \frac{h^2 k^2}{2\pi^2 m^* \Delta E}}, \quad (2)$$

upper and lower signs refer to the occupied (valence) and unoccupied (conduction) bands, n is the band index ($n = 1$, for the first conduction band and last valence band, and so on), k is parallel component of wave-vector \mathbf{k} , and the effective mass m^* is

$$m^* = \frac{\Delta E}{2V_F^2}. \quad (3)$$

Considering the important role of Fermi velocity of graphene and silicene, the application of this simple semiclassical approach, can be of helpfully interpreting the energy gap measurements on the currently synthesized GNR and SiNR structures.

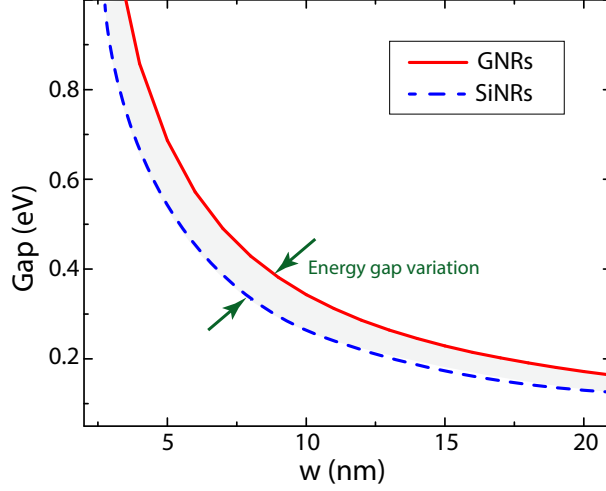


FIGURE 3. Variation of the energy gap (eV) vs ribbon width (nm). The red line for GNRs and blue dashed line for SiNRs.

III. RESULTS AND DISCUSSIONS

Energy Gap

Figure 2 shows the comparison between the data presented in the previous works, experimental in Figure 2 (a) [37] and theoretical in Figure 2 (b) [48], with the plotting curve of Equation 1, where the energy gap of GNRs is a function of w . Our results are plenty consistent with the experiments [37] on parallel GNRs (P1-P4) up to ~ 20 nm wide, see Figure 2 (a). But GNRs with widths above to > 20 nm show a variation of ~ 60 meV. The main reason for this discrepancy is because V_F of graphene is fixed at 0.83×10^8 cm/s, in Equation 1. We expect that this value to be lower in GNRs, by the following assumption. The V_F of graphene decreases as diminishing the ribbon width, which is a consequence of changing from a conical band dispersion (in graphene) to parabolic band dispersion (in GNRs). Experimental measurements could corroborate this premise on ultra-narrow GNRs.

Figure 2 (b) show the energy gaps computed with the semiclassical approach and GW approximation. The main difference between the presented approach and the methodology of [48] lies on the physical treatment of the system. Accordingly, we employ the V_F of graphene obtained from DFT computation [1], instead of a many-body treatment. Accounting the shaped-edge of GNRs, GW approximation shows the energy gap of three ribbon-families, which are classified as $3p+1$, $3p$, or $3p+2$ (p is a positive integer number), with widths from 0.4 to 2.7 nm. Our semiclassical energy gap values match perfectly with the predicted region, from 1.5 to 2.7 nm, within GW approximation and seem to be consistent with the $3p$ family. In addition, for a ribbon of 2.7 nm wide, the semiclassical energy gap value is 1.27 eV, very close at the experimental energy gap [38] measured on recently synthesized GNRs. Thus, the application of the semiclassical approach to GNRs is appropriate for ribbons ranging from 1.5 to 20 nm wide, and furthermore, this approach can be extended to characterize the energy gap in SiNRs of the similar width.

In Figure 3, we show a comparison of energy gaps between GNRs and SiNRs, up to 22 nm wide. Besides, the semiconducting feature of GRNs and the predicted in SiNRs by using Equation 1; a difference of energy gaps is observed (shaded area in Figure 3), which is a direct consequence of reduced velocity of charge-carriers in silicene, i.e., its $V_F = 0.64 \times 10^8$ cm/s, respect to graphene ($V_F = 0.83 \times 10^8$ cm/s). It is worth noting that increasing the ribbon width (e.g., from 2.5 to 22 nm, Figure 3) the energy gap difference decreases from ~ 0.3 eV to ~ 30 meV. The latter can be explained as an effect inversely proportional to the ribbon width, slightly affected by the respective V_F . Additionally, Equation 1 could also be used to estimate the energy gap (and band dispersion) for wider ribbons. However, it is necessary to consider the *relative* discrepancy of our results (above discussed) with the experimental measurements, see Figure 2 (a).

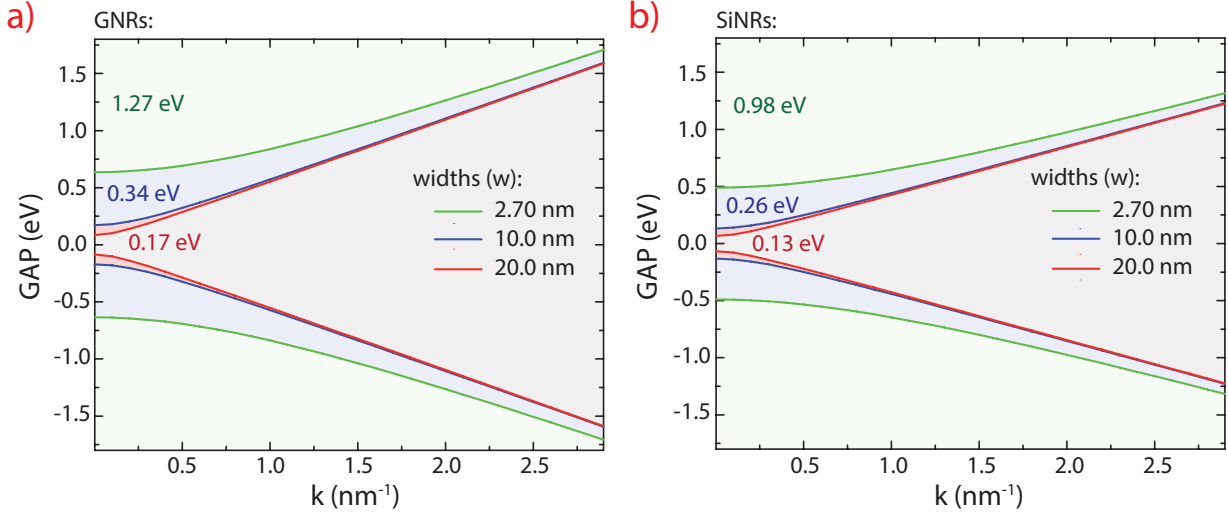


FIGURE 4. Band dispersion (eV) vs parallel component k (nm^{-1}). For three distinct ribbon widths. (a) GNRs and (b) SiNR.

Band Dispersion

On the other hand, solving Equation 3, the effective mass m^* is employed in Equation 2 to compute the band spectrum for any GNR or SiNR. Thus, the band dispersion as a function of parallel component k , is reported in Figure 4. Note, we have considered only the bottom conduction band and top valence band in each case, i.e., $n = 1$. Three distinct ribbon widths (2.7, 10 and 20 nm) are selected, in which the semiclassical approach has demonstrated a good agreement with the experimental and theoretical observations (previously discussed). The reported energy gap values for GNRs in Figure 4 (a), and SiNRs in Figure 4 (b), correspond to the effective electron masses ($\sim 0.2 - 0.02$) m_0 , where m_0 is the free-electron mass. In particular, for a GNR of 2.7 nm wide its effective mass reduces as $m^* \approx 0.16m_0$, which is very close to the effective mass of GNRs calculated in [48].

As shown in DFT [32] and GW approximation [48], the presented semiclassical approach exhibits two important characteristics in GNRs and SiNRs, Figure 4 (a)-(b): (i) a parabolic band dispersion similar to DFT- or GW-based picture, and (ii) a direct energy gap, which can be tuned through the variation of ribbon width. From the technological point of view, a remarkable feature is observed in GNRs and SiNRs of 2.7 nm wide where the corresponding energy gaps are 1.27 eV and 0.98 eV, respectively. These results can be exploited in modern electronics with particular emphasis on SiNR-based devices. Further support in this direction has been the fabrication of a first silicene-based field-effect-transistor [52].

IV. CONCLUSIONS

We have presented a semiclassical approach based on recently reported Fermi velocity in graphene [1] and silicene [2]. This simplified approach is useful predicting the energy gap, and band dispersion in GNRs and SiNRs matching according previous experimental [37], and theoretical [48] works for ribbons ranging from 1.5 to 20 nm wide and reasonably describe the energy gaps in ribbons with widths > 20 nm. The presented semiclassical approach confirms two remarkable points: the tunability of the energy gap by varying the ribbon width and the parabolic band spectrum as observed in more sophisticated DFT [32] and GW [48] approaches. Particular attention is given on GNRs and SiNRs of 2.7 nm wide, where the energy gap values are consistent with the experimental measurements of [38]. Furthermore, these are within the most attractive range of 0.98-1.27 eV and give promise for applications of GNRs and SiNRs in nanoelectronics. Besides, our results support the argument that silicene-based nanomaterials are excellent options for the design of next-generation nanodevices, in competition with graphene-related materials.

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