OPTIMAL CONTACT ELECTRODE. Indeed, an ohmic contact has been demonstrated with armchair GNRs with a width of seven carbon atoms (7-aGNR), the thermal stability and the oxidation process of ambient-exposed high-performance quasi-metallic contact characteristics of the interfaces. The GNR theory study revealed the native covalent-bond nature and the atomic-level stability of both zigzag and armchair edges of GNRs against oxidation. In a joint experimental–theoretical investigation, we showed that the zigzag and armchair edges can be oxidized by thermal annealing, leading to a significantly reduced bandgap of the 7-aGNR. We mapped out the evolution of edge structures and revealed the reaction path and the produced chemical species of both for the zigzag and the armchair edges. (3) We also investigated potential GNR-based devices and proposed a novel structure that can be manufactured with atomic precision and exhibits significant negative differential resistance.

RESEARCH CHALLENGE

Electrical contact to low-dimensional (low-D) materials is a key to their electronic applications. Traditional metal contacts to low-D semiconductors create gap states that can pin the Fermi level ($E_F$). However, low-D metals possessing limited density of states at $E_F$ can enable gate-tunable work functions and contact barriers. Moreover, a seamless contact with native bonds at the interface that does not introduce in-gap states can act as an optimal contact electrode. Indeed, an ohmic contact has been achieved in 2D transition metal dichalcogenides at the interface of the metallic 1T- phase and the semiconducting 2H-phase. We demonstrated an all-carbon staircase contact to ultra-narrow graphene nanoribbons (GNRs), a wide bandgap 1D semiconductor. A combined scanning tunneling microscopy and density functional theory study revealed the native covalent-bond nature and the quasi-metallic contact characteristics of the interfaces. The GNR staircase points constitute a promising route to high-performance graphene nanoelectronics.

The stability of graphene nanoribbons (GNRs) against oxidation is critical for their practical applications. We systematically studied the thermal stability and the oxidation process of ambient-exposed armchair GNRs with a width of seven carbon atoms (7-aGNR), grown on an Au(111) surface. The atomic scale evolution of the armchair edges and the zigzag ends of the GNRs after annealing at different temperatures were revealed by using scanning tunneling microscopy, Raman spectroscopy, X-ray photoelectron spectroscopy, and first-principles calculations. In the oxidation process, $O_2$ molecules first dissociate and then react with carbon atoms at the edges. Two different oxygen species were identified at the armchair edges, namely the hydroxyl pair and epoxy with one bridge oxygen bonded to two edge carbons. These oxidation species modify the electronic properties of pristine 7-aGNRs, with a bandgap reduction from 2.6 eV to 2.3 eV and 1.9 eV for the hydroxyl pair- and epoxy-terminated edges, respectively. These findings revealed that both the zigzag and armchair edges of GNRs can be oxidized after thermal annealing and that the oxidation can greatly affect the electronic properties of GNRs, which also opens an opportunity for using GNRs as high-temperature oxygen sensors.

The development of atomically precise synthesis of graphene nanostructures promises a revolution in device design to deliver novel electronic functions. A prominent example is the negative differential resistance (NDR) device, for which many designs based on graphene nanoribbons (GNRs) were proposed. However, a controllable fabrication method of such devices with atomic precision has yet to emerge. We proposed a practical device structure, based on armchair GNRs, which exhibits strong NDR effect. Our computational evaluation of the traditional resonant tunneling diode uncovers important issues at the atomic scale concerning the need to minimize the direct tunneling current between the leads while achieving high peak current. The proposed device consists of a short semiconductor GNR and its lower-gap intermediate segments, which enables high current by the alignment of levels across the segments while enlarging the tunneling distance between the leads.

METHODS & CODES

The electronic structure calculations used the Real-space-MultiGrid (RMG) and Quantum Espresso codes. The quantum transport nonequilibrium Green’s function calculations used the localized orbitals and nonequilibrium Green’s function (NEGF) branches of the RMG code.