Graphene as a Versatile Spin Tunnel Barrier

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Abstract
The quantum phenomenon of tunneling enables novel charge-based devices with ultra-low power consumption, and is key to the emerging field of spintronics. The tunnel barrier is critical, and the intrinsic characteristics of 2D materials such as graphene and h-BN provide many of the desired attributes: they provide discrete monolayer control of thickness and resist pinhole formation, they are chemically inert and prevent interdiffusion, and are highly stable. We describe the use of single layer graphene as a spin tunnel barrier for magnetic tunnel junctions and spin injection into silicon wafers and nanowires, and functionalized graphene for spin injection into an adjacent graphene transport channel.

1. Introduction
Graphene has been widely studied for its high in-plane charge carrier mobility and long spin diffusion lengths. In contrast, the out-of-plane charge and spin transport behavior of this atomically thin material have not been well addressed. Tunnel barriers to date have relied upon oxides which often exhibit defects, trap states and interdiffusion which compromise performance and reliability. We show here that while graphene exhibits metallic conductivity in-plane, it serves effectively as an insulator for transport perpendicular to the plane. We fabricate graphene-based magnetic tunnel junctions, and demonstrate electrical spin injection/detection in silicon and graphene using graphene as a tunnel barrier.

2. Measurements and Results
Graphene is grown by chemical vapor deposition on copper foil and incorporated as the tunnel barrier by physical transfer and standard lithographic processes to form Co / graphene / NiFe magnetic tunnel junctions (MTJs) [1]. Non-linear I-V curves and weak temperature dependence of the zero-bias resistance provide clear evidence for tunneling. The magnetic field dependence exhibits the classic signature of MTJ behavior, and the structures exhibit tunneling magnetoresistance (TMR) to 425 K, in good agreement with theory [2].

Single-layer graphene also successfully circumvents the classic issue of conductivity mismatch between a metal and a semiconductor for electrical spin injection and detection. Hanle spin precession measurements performed on devices with NiFe / single layer graphene / Si contacts exhibit the classic Lorentzial lineshape due to spin injection and dephasing in Si above room temperature. We show that the spin lifetimes correlate with Si carrier concentration, and the contact resistance–area products are two to three orders of magnitude lower than those achieved with oxide tunnel barriers on silicon substrates with identical doping levels, as shown in Figure 1 [3].

Fig. 1 Single layer graphene provides a tunnel barrier that enables spin injection from a ferromagnetic metal into a semiconductor such as silicon. Calculation of the local (two-terminal) magnetoresistance (MR) as a function of the conventional resistance–area product of the contact and the Si electron density for the device geometry shown in the inset, using the theory of Fert and Jaffres. Data points are the resistance–area products measured for our ferromagnetic metal/tunnel barrier/Si contacts using 2 nm SiO2 (triangles), 1.5 nm Al2O3 (diamond) and monolayer graphene (circles) tunnel barriers prepared from identical Si wafers in our laboratory. The ferromagnetic metal/graphene resistance–area products fall within the window of useful magnetoresistance values. Parameters: W=w=11 nm.
This reduction of contact resistance enables spin injection and quantitative measurements of spin lifetimes in silicon nanowires, as shown in Figure 2 [4]. We report the first observation of spin precession via the Hanle effect in both local three-terminal and non-local spin-valve geometries, providing a direct measure of spin lifetimes and confirmation of spin accumulation and pure spin transport. The use of graphene as the tunnel barrier provides a low-resistance area product contact and clean magnetic switching characteristics, because it smoothly bridges the nanowire and minimizes complicated magnetic domains that otherwise compromise the magnetic behavior.

Finally, we demonstrate a homoepitaxial tunnel barrier structure in which graphene serves as both the tunnel barrier and the high mobility transport channel. We fluorinate [5] or hydrogenate [6] the top layer of a graphene bilayer to decouple it from the bottom layer, so that it serves as a single monolayer tunnel barrier for both charge and spin injection into the lower graphene channel. We demonstrate high spin injection efficiency with record tunneling spin polarization > 45%, lateral transport of spin currents in non-local spin-valve structures, and determine spin lifetimes with the Hanle. The characteristic spin-valve signal is shown in Figure 3. In contrast with most oxide tunnel barriers on graphene, fluorinated graphene provides much larger tunneling spin polarization efficiency that we attribute to interface spin filtering, a more uniform and well-controlled barrier, and allows the observation of the theoretically predicted Hanle voltage and spin lifetime on gate voltage.

3. Conclusions
Utilizing intrinsic two-dimensional layers such as graphene or hexagonal boron nitride as tunnel contacts on semiconductors offers many advantages over conventional materials deposited by vapor deposition, enabling a path to highly scaled electronic and spintronic devices.

Fig. 3 Non-local spin valve signal obtained using a top fluorinated graphene layer as a tunnel barrier for electrical spin injection and detection of spin transport in the underlying graphene transport channel.

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References