Scientists at the U.S. Naval Research Laboratory (NRL) have created a new type of tunnel device structure in which the tunnel barrier and transport channel are made of the same material, graphene. They show that dilutely fluorinated graphene, a single atomic layer of carbon atoms arranged in a two-dimensional (2D) honeycomb array, acts as a tunnel barrier on another layer of graphene for charge and spin transport. They demonstrate tunnel injection through the fluorinated graphene, and lateral transport and electrical detection of pure spin current in the graphene channel. They further report the highest spin injection values yet measured for graphene, providing evidence for the enhancement of tunnel spin polarization theoretically predicted to occur for certain ferromagnetic metals on graphene. This discovery opens an entirely new avenue for making highly functional, scalable graphene-based electronic and spintronic devices a reality.

The research results are reported in a paper published in the journal *Nature Communications* on January 21, 2014.

The coupled imperatives for reduced heat dissipation and power consumption in high-density electronics have rekindled interest in devices based on tunneling, a quantum mechanical phenomenon in which electrons transit through a potential barrier rather than going over it. Because the tunnel barrier and transport channel are typically very different materials, such devices require mating dissimilar materials, raising issues of heteroepitaxy, layer uniformity, interface stability and electronic defect states that severely complicate fabrication and compromise performance.

"2D materials such as graphene and hexagonal boron nitride obviate these issues and offer a new paradigm for tunnel barriers", explains Dr. Berend Jonker, Senior Scientist and project leader. In bulk form, these materials are comprised of well-defined layers which exhibit very strong atomic bonding in-plane, but relatively weak bonding between the layers, known as van der Waals bonding. Single layers can be readily separated from the bulk, or grown directly over large areas by a variety of techniques. These layers thus have a strong tendency to be very uniform in thickness down to a single atom, have very few defects, and do not intermix readily with other materials—these are key characteristics for a tunnel barrier, in which the tunnel current depends exponentially on the barrier thickness.

The NRL scientists fluorinate 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. They deposit ohmic (gold) and ferromagnetic permalloy (red) contacts as shown in the figure, forming a non-local spin valve structure. When a bias current is applied between the left two contacts, a spin-polarized charge current tunnels from the permalloy into the...
graphene transport channel, generating a pure spin current that diffuses to the right. This spin current is detected as a voltage on the right permalloy contact that is proportional to the degree of spin polarization and its orientation. The vectorial character of spin (compared to the scalar character of charge) provides additional mechanisms for the control and manipulation needed for advanced information processing.

The NRL team demonstrated the highest spin injection efficiency ever measured for graphene (63%), and determined spin lifetimes with the Hanle effect. In contrast with most oxide tunnel barriers on graphene, fluorinated graphene provides much larger tunnelling spin polarization efficiency, attributed to interface spin filtering and a more uniform, well-controlled barrier, and allows the observation of the theoretically predicted Hanle voltage and spin lifetime on gate voltage.

These results identify a new route towards high quality, next generation graphene electronic/spintronic devices including spin-based transistors, logic, and memory. In addition, the process is completely scalable and easily accomplished. "In the near future," predicts Dr. Adam Friedman, lead author on the project, "We will be able to write entire spintronic circuits in situ on grown, large areas of bilayer graphene simply by selectively chemically modifying the top layer of graphene." Fluorographene/graphene enables realization of homoepitaxial few-layer carbon structures for versatile electronic devices.

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