Journal of Magnetism and Magnetic Materials 324 (2012) 369–381

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Review

Spin transport and relaxation in graphene

Wei Han, K.M. McCreary, K. Pi [1], W.H. Wang [2], Yan Li [3], H. Wen, J.R. Chen, R.K. Kawakami *

Department of Physics and Astronomy, University of California, Riverside, CA 92521, USA

ARTICLE INFO

Article history:
Received 16 June 2011
Received in revised form
30 July 2011
Available online 7 August 2011

Keywords:
Graphene
Spintronics
Spin injection
Spin relaxation
Spin transport

ABSTRACT

We review our recent work on spin injection, transport and relaxation in graphene. The spin injection and transport in single layer graphene (SLG) were investigated using nonlocal magnetoresistance (MR) measurements. Spin injection was performed using either transparent contacts (Co/SLG) or tunneling contacts (Co/MgO/SLG). With tunneling contacts, the nonlocal MR was increased by a factor of ~1000 and the spin injection/detection efficiency was greatly enhanced from ~1% (transparent contacts) to ~30%. Spin relaxation was investigated on graphene spin valves using nonlocal Hanle measurements. For transparent contacts, the spin lifetime was in the range of 50–100 ps. The effects of surface chemical doping showed that for spin lifetimes in the order of 100 ps, charged impurity scattering (Au) was not the dominant mechanism for spin relaxation. While using tunneling contacts to suppress the contact-induced spin relaxation, we observed the spin lifetimes as long as 771 ps at room temperature, 1.2 ns at 4 K in SLG, and 6.2 ns at 20 K in bilayer graphene (BLG). Furthermore, contrasting spin relaxation behaviors were observed in SLG and BLG. We found that Elliot–Yafet spin relaxation dominated in SLG at low temperatures whereas Dyakonov–Perel spin relaxation dominated in BLG at low temperatures. Gate tunable spin transport was studied using the SLG property of gate tunable conductivity and incorporating different types of contacts (transparent and tunneling contacts). Consistent with theoretical predictions, the nonlocal MR was proportional to the SLG conductivity for transparent contacts and varied inversely with the SLG conductivity for tunneling contacts. Finally, bipolar spin transport in SLG was studied and an electron–hole asymmetry was observed for SLG spin valves with transparent contacts, in which nonlocal MR was roughly independent of DC bias current for electrons, but varied significantly with DC bias current for holes. These results are very important for the use of graphene for spin-based logic and information storage applications.

1. Introduction

Spintronics utilizes the electron spin degree of freedom for information storage and logic operations, which could decrease the power consumption, increase data processing speed, and increase integration densities [1–3]. Lateral spin valves consisting of ferromagnetic (FM) electrodes connected to a nonmagnetic spin transport channel are of special interest because of the design flexibility for multi-terminal devices and the ability to manipulate spin during transport [4–6]. Experimentally, spin injection and transport have been observed in a variety of materials including metals, semiconductors, and carbon-based materials. The first electronic spin injection and detection was performed by Johnson and Silsbee in 1985 in a single-crystal aluminum bar at temperatures of 77 K [7]. Following this work, electron spin injection in metals, such as Al, Cu, Ag and Au, has been demonstrated even up to room temperature (RT) in some cases [5,6,8–13]. Spin transport in semiconductors was first detected using ultrafast optical methods [14–16]. Recently there has been significant progress in the area of electrical injection and manipulation of spin in semiconductors such as Si, GaAs, Ge, etc. [17–24].

Carbon-based materials have attracted considerable interest because they are expected to have long spin lifetimes due to low intrinsic spin–orbit and hyperfine couplings [25,26]. Spin injection into carbon nanotubes has been performed using Co and other FM electrodes [27–33]. Organic semiconductors, such as Alq_3, have also been studied for spin transport due to the chemical flexibility and optoelectronic properties [34–36]. In 2004, a single atomic layer of graphitic carbon, known as graphene, was isolated by the Geim group [37]. Its carrier concentration and conductivity were found to be highly tunable with electrostatic gates, and the half-integer quantum Hall effect

* Corresponding author. Tel.: +951 827 5343; fax: +951 827 4529.
E-mail address: roland.kawakami@ucr.edu (R.K. Kawakami).
[1] Present address: Hitachi Global Storage Technologies, San Jose, CA 95135, USA.
[2] Present address: Institute of Atomic and Molecular Sciences, Academia Sinica, Taipei 106, Taiwan, ROC.
[3] Present address: Los Alamos National Laboratory, Los Alamos, NM 87545, USA.

0304-8853/$ - see front matter © 2011 Elsevier B.V. All rights reserved.
doi:10.1016/j.jmmm.2011.08.001

demonstrated the distinctive character of two-dimensional chiral Dirac fermions [38,39]. Due to high electronic mobility, gate tunability, and the potential for long spin lifetimes, graphene has drawn a lot of attention in the spintronics field [26,40–71]. Currently, several groups have demonstrated spin transport in single layer graphene (SLG) and multilayer graphene (MLG) [40,42–45,49,50,55]. The pioneering work was done by the van Wees group who demonstrated gate tunable spin transport and spin precession in nonlocal SLG spin valves at RT [41]. In that work, the electrical detection of spin precession was particularly important to prove that the observed signals indeed originated from spin transport. Similar results were reported by several groups [40,42–44,49,55]. Subsequent results include the measurement of anisotropic spin relaxation [46], local spin transport in MLG [44,45], spin drift effects [47], and bias dependence of spin injection [48,50,55].

These earlier studies identified two critical challenges, which must be overcome in order to realize the full potential of graphene for spintronics. The first important challenge was to enhance the spin injection efficiency, which was low due to the conductance mismatch between the FM metal electrodes and graphene [72]. Although it was expected that the conductance mismatch problem could be alleviated by inserting tunnel barriers into the spin injection interface [73,74], growing smooth layers on top of graphene was non-trivial because the low surface energy and high surface diffusion led to cluster formation. It was found that using a submonolayer Ti seed layer followed by MgO deposition produced atomically smooth MgO films [75]. As a result, tunneling spin injection was achieved with greatly enhanced spin injection efficiencies [58]. The second important challenge was to determine the cause of the unexpectedly short spin lifetimes measured by the Hanle effect (spin precession) in SLG (50–200 ps) [41,52–54,57]. These lifetimes were orders of magnitude shorter than expected from the intrinsic spin–orbit couplings (~μs) [25,26]. The linear scaling of spin scattering and momentum scattering in SLG [52] suggested that these were related by an Elliot–Yafet mechanism for spin relaxation (i.e. spin scattering during a momentum scattering event). Because charged impurity scattering was a strong source of momentum scattering, its effect on spin scattering was investigated through metal-doping experiments. Interestingly, it was found that for spin lifetimes on the order of 100 ps, the charged impurity scattering was not the dominant mechanism for spin relaxation in graphene [57]. Subsequently, it was observed that tunnel barriers significantly enhanced the measured spin lifetime, indicating that metal contact-induced effects were very important for spin relaxation [58]. Furthermore, with tunneling contacts to suppress the contact-induced spin relaxation, the spin lifetimes observed were as long as 771 ps at RT in SLG, 1.2 ns at 4 K in SLG, 2.0 ns at RT in bilayer graphene (BLG), and 6.2 ns at 20 K in BLG [76,77]. Very recently, Dyakonov–Perel (DP) spin relaxation was found to dominate in BLG [76–78]. Spin transport has also been demonstrated in large area epitaxial graphene fabricated by chemical vapor deposition (CVD) [79], which makes graphene a promising candidate for large scale spintronic applications [78].

In this paper, we review the contributions of our group to the study of graphene spintronics, including some of the key advances mentioned above [50,51,53,57,58,75,76]. This paper is constructed as follows: In Section 2, we discuss the nonlocal magnetoresistance (MR) measurements. In Section 3, we describe the fabrication of the graphene spin valves. In Section 4, we describe our experimental results on spin injection using either transparent contacts (Co/SLG) or tunneling contacts (Co/MgO/SLG). In Section 5, we investigate spin relaxation in SLG and BLG. Section 6 discusses the gate tunable spin transport in SLG for future spin field effect transistors and Section 7 discusses the unique opportunities of using SLG for bipolar spintronics. In Section 8, we discuss the future directions of graphene spintronics.

2. Nonlocal spin transport measurements

Typically, there are two geometries for electrical spin transport measurements. First is the conventional spin transport geometry, known as the "local" measurement, which measures the resistance across two ferromagnetic electrodes (Fig. 1a). Spin-polarized electrons are injected from one electrode, transported across the graphene, and detected by the second electrode. The spin transport is detected as the difference in resistance between the parallel and antiparallel magnetization alignments of the two electrodes. This is the geometry used for magnetic tunnel junctions and current perpendicular to the plane giant magnetoresistance (CPP-GMR) [80,81]. The second geometry is the "nonlocal" measurement [5,7], which uses four electrodes, as shown in Fig. 1b. Here, a current source is connected across Co electrodes E1 and E2 to inject spins at E2. For spin detection, a voltage is measured across Co electrodes E3 and E4, and the signal is due to the transport of spins from E2 to E3. This measurement is called

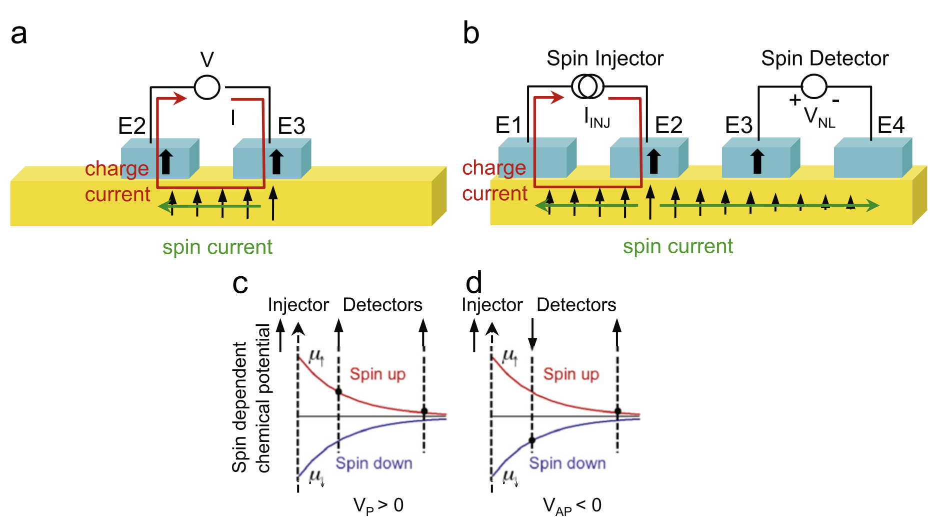

Fig. 1. Spin transport measurement. (a) Schematic of local spin transport measurements. (b) Schematic of nonlocal spin transport measurements. (c, d) Spin-dependent chemical potential for parallel and antiparallel states of spin injector and detectors in the nonlocal geometry.