



Spin relaxation and proximity effect in WS₂/graphene/fluorographene non-local spin valves

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ABSTRACT

The mechanisms leading to spin relaxation in graphene and its heterostructures continue to be debated. Control of the spin relaxation in graphene-based structures is necessary to achieve the envisioned utility of graphene in future spintronic devices beyond Moore's law. Proximity induced spin relaxation caused by contact to a high spin-orbit material, such as WS₂, offers a promising avenue to manipulate the spin lifetime. We demonstrate the operation of WS₂/graphene/fluorographene non-local spin valves and extract the spin lifetimes for a range of carrier concentrations by Hanle effect measurements. Four-terminal charge transport measurements allow us to calculate the momentum relaxation time as a function of carrier concentration and compare it to the spin lifetime. These data show that the D'yakonov-Perel' mechanism is the dominant spin relaxation mechanism for WS₂/graphene/fluorographene devices, while, for reference graphene/fluorographene devices, linear scaling between the spin and momentum lifetimes points to spin-flip scattering during strong elastic scattering events where the scattering event is strongly coupled to the electron spin. We attribute the change in spin relaxation type in part with the inclusion of WS₂ as a substrate to proximity induced spin-orbit coupling due to the adjacent WS₂ layer, and we compare our data to the literature.

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1. Introduction

Graphene, composed entirely of sp² bonded carbon atoms, has negligible intrinsic spin-orbit coupling [1]. This characteristic combined with graphene's high mobility should result in extremely long spin lifetimes and diffusion lengths, suitable for next-generation spintronic-based devices [2–7]. However, as a single monolayer, it is greatly affected by extrinsic impurities [8,9], interaction with the contacts used for spin injection/detection [10,11], and spin absorption [12,13], which can dominate spin relaxation and make control of it challenging. Consequently, there are many contradictory reports of spin lifetimes and relaxation mechanisms in graphene samples [14–17]. Control over the spin relaxation is necessary for any implementation in modern device architectures [18].

Two mechanisms of electron spin relaxation are generally

considered in the literature as the key role players in spin-dependent phenomena in graphene: the Elliot-Yafet (EY) and the D'yakonov-Perel' (DP) mechanisms [19]. The EY mechanism produces spin relaxation through spin-flip scattering arising from momentum scattering events where the scatterer does not interact with the electron spin [20–22]. At first, experimental results considering both exfoliated and grown graphene layers were thought to support this mechanism [21,23]. However, some debated this conclusion because the EY mechanism does not apply for cases of strong elastic scatterers that are expected to exist in great number in graphene devices such as covalently bonded impurities, ripples, or adsorbed residues where the scatterer interacts locally with the electron spin. In the general EY mechanism formula, the spin relaxation time is proportional to the momentum relaxation time, with the proportionality constant being the probability of a spin-flip during a scattering event, implying a linear scaling between the two quantities [22]. However, for graphene, the spin-flip probability is strongly dependent on the carrier concentration, thus indicating that a linear fit does not necessarily implicate EY [22]. The general consensus is that vacancies and grain

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boundaries in grown material can induce an effective spin-orbit coupling that results in EY. However, the more likely candidate for most graphene devices is spin-flip scattering during momentum scattering events where the scattering potential couples with the electron spins, as can be caused by adatoms, ripples, and impurities [22]. The contacts may also play a role in the measured spin lifetime, especially for exfoliated films and those with transparent oxide tunnel barriers [10,11]. However, contact effects mainly result in an underestimation of the spin lifetime as measured. Therefore, they will not be a focus of this study where we will be more concerned with qualitative behavior in the channel of a heterostructure and, as will be discussed, we do not employ transparent, oxide-based tunnel barriers. The DP mechanism produces spin relaxation arising from precession in a random effective magnetic field within the lattice. Here, the spin relaxation time is inversely proportional to the momentum relaxation time [19]. Suspended graphene, which is less affected by extrinsic impurities and scattering caused by the substrate, has shown evidence of DP spin relaxation, mostly attributable to an effective spin-orbit coupling caused by defects and vacancies in the graphene [24].

Previous studies attempted to control spin relaxation in graphene by creating spin-orbit coupling in the graphene channel itself through the introduction of impurities using dilute functionalization, e.g. by hydrogenation or fluorination [25–28]. While this method does indeed create a spin-orbit coupling, on order of meV, the possibility exists of simultaneously causing unintentional defects and electronic scatterers in the graphene, which erode its high-quality charge transport properties, thereby compromising the desired properties. Another, perhaps more promising, method of controlling the spin relaxation in graphene is to purposely induce a higher spin-orbit coupling using proximity effects [6,7,29]. Theoretical studies found that by placing graphene in proximity to large spin-orbit 2D materials such as transition metal dichalcogenides (TMDs) like WS_2 , a spin-orbit coupling as high as 33 meV can be induced in the graphene [30]. As the electronic wavefunctions of 2D materials extend out-of-plane, and each individual layer is atomically thin, two stacked layers can interact through strong van der Waals interactions and/or electronic hybridization through wavefunction coupling, which can allow for new properties in the resultant heterostructure. Moreover, an enhancement in the spin-orbit splitting in monolayers of these materials due to loss of inversion symmetry is expected and can add to their viability for proximity effect induced control layers [31].

Although TMDs such as WS_2 may not be suitable as spin channels on their own due to their low conductivities [32], difficulty in creating low-resistance contacts [33], and intervalley scattering effects [34], there is an opportunity to couple them to graphene, creating a van der Waals heterostructure. In such a structure, the relatively low TMD conductivity is an advantage, because it will not produce parallel conductance paths that would complicate the transport. As an additional incentive to use other 2D materials for substrates for graphene, such heterostructures have shown enhancement of graphene's charge transport properties over the ubiquitous SiO_2/Si substrate material, owing mostly to a decrease in phonon scattering [35]. Although the W-based 2D materials were not shown to produce as much mobility enhancement as h-BN [36], it was nonetheless found to result in higher device metrics than SiO_2/Si .

Several proximity induced effects have recently been reported in graphene films [37–41]. For example, in ref. [41] it was determined that a variable increase in interface resistance caused by a WS_2 tunnel barrier led to a reduction in back-flow of spin, and that it could be qualitatively determined that the reduced measured spin-relaxation (relative to graphene alone) was caused by

proximity of graphene to WS_2 . However, no attempt was made to determine the type of spin relaxation, the quantitative effect of the WS_2 on spin relaxation, or to extract an effective spin-orbit coupling strength. In ref. [39], a charge transport-based study of the weak antilocalization at millikelvin temperatures of a graphene/ WS_2 heterostructure revealed a ~ 5 meV spin-orbit coupling by proximity in the graphene and a spin relaxation governed by intervalley scattering. However, no spin valve devices or direct spin-based measurements were made. In ref. [40], proximity interaction between graphene and WS_2 in a heterostructure was shown to result in a strong interface Coulomb interaction and drive a drag-like effect in the graphene charge transport.

In this study, we observe that DP type spin relaxation dominates in graphene in WS_2 /graphene/fluorographene lateral spin valves, and attribute it to a combination of proximity effects at the WS_2 /graphene interface and reduced elastic scattering due to the use of WS_2 as the substrate. In contrast, we observe a different mechanism for structures without WS_2 , which we conclude arises from spin-flip scattering caused by momentum scattering events where the spin is coupled to the scatterer, caused by strong scatterers such as ripples, adatoms, and impurities. We first demonstrate spin transport through the operation of a four-terminal non-local spin valve and determine spin lifetimes from corresponding Hanle effect measurements. We find that the spin lifetime is inversely proportional to the momentum relaxation time in WS_2 /graphene heterostructures, indicating that the DP type spin relaxation dominates. In contrast, in devices that do not include the WS_2 , the spin lifetime is proportional to the momentum relaxation time, and there does appear to be a dependence on carrier concentration. We show that although spin-flip scattering during elastic scattering events is the likely cause of spin relaxation in devices without WS_2 , it is of the type not attributable to the EY mechanism. Rather, it is likely caused by ripples, adatoms, and impurities. We extract an effective spin-orbit coupling value for the devices that include WS_2 and compare with results in the literature. In general, studying the spin relaxation in 2D heterostructures is complicated by multiple contributions, and no study can fully account for every variable. However, much can be learned by at least fitting data to these fundamental models and qualitatively analyzing the results.

2. Methods and fabrication

Large-area films of WS_2 are synthesized by sulfurizing pre-deposited W. A deposition time of 5 s is used to sputter thin films of polycrystalline W onto $SiO_2/n^+ Si$ wafers (275 nm thermal oxide thickness). The W films are then loaded into the central heating zone of a 2-inch diameter quartz tube. A separate quartz boat containing sulfur powder is placed upstream, outside the furnace-heating zone. A gas flow of 90 sccm Ar and 10 sccm H_2 is used as the sample heats to 900 °C at ambient pressure. The temperature is ramped at a rate of 20 °C/min to 625 °C, followed by 10 °C/min to 900 °C. The temperature is held constant at 900 °C for 10 min, then allowed to cool naturally to room temperature. Fig. 1(a and b) show photoluminescence (PL) and Raman spectroscopy data of the WS_2 film taken with a 488 nm laser at room temperature and in air. The data confirm that it is a monolayer film with a small grain size. Although it is difficult to measure this grain size, previous studies using comparable growth methods have estimated it to be approximately 100 nm² to 1 μm^2 [42]. We observe PL at ~ 2 eV, consistent with a direct gap material and expected for single layer WS_2 , with no lower energy indirect peaks. We find a E_{12g}^1 to A_{1g} mode separation of ~ 61.6 cm⁻¹, consistent with that reported for monolayer WS_2 in the literature [43,44].

Graphene is grown by chemical vapor deposition (CVD) on Cu foils [45]. Films are removed from the Cu growth substrate using

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