# Magnetic order in defective reduced graphene oxides (rGO) investigated using $\mu$SR

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Several studies have investigated graphene-based materials with a focus on molecular-based magnets. In particular, reduced graphene oxide (rGO) is a fascinating system that has numerous defects and functional group on its sheets; hence, it is ideal to generate the magnetism of intrinsically nonmagnetic graphene. rGO have been reported to exhibit various magnetic states including paramagnetic,[1] weak super-paramagnetic,[2] and room temperature ferromagnetic.[3–5] These magnetic features are believed to be related to the defect states in the rGO sheet.[3,4,6] A sufficient number of defects, especially in the form of vacancies and chemisorbed hydrogen, can lead to the onset of magnetic ordering. Therefore, we intend to further investigate a possible magnetic ordering in rGO prepared by the green synthesis method.[7]

The presence of significant number of defects and different types of oxygen functionality in the obtained rGO have been confirmed by Raman, Fourier-transform infrared (FTIR), and photoemission spectroscopies. The defect concentration increases, while that of the oxygen functional group decreases when rGO is thermally reduced at 1000°C (rGO-1000). Furthermore, an enhancement in magnetization was observed when the number of defect increases, confirming defect-induced-magnetism in rGO. It was verified that the magnetic impurities did not contribute toward enhancement of the magnetization. Thus, muon spectroscopy ($\mu$SR) could help investigate the possible onset of magnetism in rGO.

$\mu$SR measurements were performed on the obtained samples, rGO and rGO-1000, under zero-field (ZF) and longitudinal-field (LF) conditions. Figure 1 (a) shows the ZF time spectra of rGO and rGO-1000 at 2 K. No clear muon-spin precession was observed. Muon spin depolarization is prominent up to 4 $\mu$s, then it is considerably slower in the range of 5–7 $\mu$s. Further, spin depolarization is observed after 8 $\mu$s. These features denote the appearance of the oscillation component on the decaying signal; it is a typical feature of graphene-based compounds.[8,9] Instead of magnetic ordering, the oscillation possibly indicates the nuclear dipolar interaction between muon and proton.[8] The changes in the initial asymmetry and background along with a small increase in the relaxing amplitude for rGO-1000 compared with that of rGO could be caused by

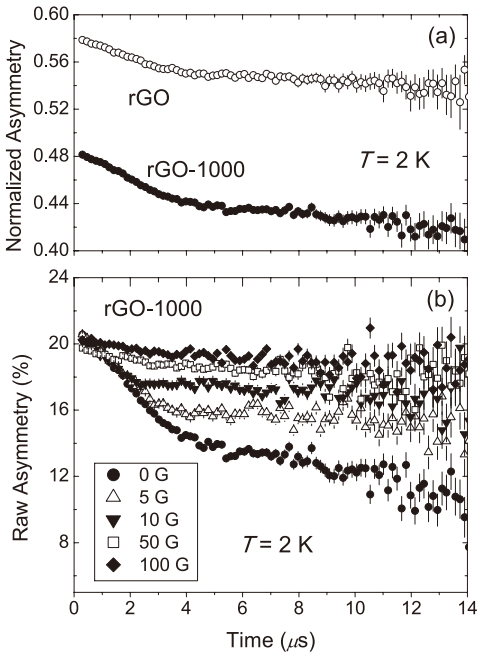


Fig. 1. (a) Zero-field (ZF) time spectra of rGO and rGO-1000 at 2 K. (b) Longitudinal-field (LF) time spectra of rGO-1000 at 2 K.

the increase in defect concentration. A missing fraction can occur in the samples when the positive muon strongly interacts with an electron (hyperfine interaction) either due to the formation of muonium or after the adduction reaction of muonium to form a radical. To confirm the cause, LF-$\mu$SR measurements were performed to decouple muon spin from the electron spin. Figure 1 (b) displays the LF spectra of rGO-1000. The decoupling is likely to occur owing to the applied field of 100 G, which is much smaller than the field required to recover the free muonium signal (1580 G). This low value demonstrates the formation of a radical complex that was not detected in bare graphene.[9] Further fitting analysis concomitant with calculation work are required to verify these suggestions.

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