Communication

Observation of Glassy Ferromagnetism in Al-Doped 4H-SiC

Bo Song, Huiqiang Bao, Hui Li, Ming Lei, Tonghua Peng, Jikang Jian, Jun Liu, Wanyan Wang, Wenjun Wang, and Xiaolong Chen


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Ferromagnetism (FM) at and above room temperature and spin polarization in the majority carrier band are essential requirements for practical application of diluted magnetic semiconductors (DMSs) in spintronics. However, inconsistencies in literature with respect to the practical application of diluted magnetic semiconductors (DMSs) in polarization in the majority carrier band are essential requirements for corresponding to 4H-SiC (ICDD-PDF: 29-1127, space group P63/mmc) were put in a capsule for magnetic measurement (see SI). We characterized the sample structure by high-resolution X-ray diffraction (XRD) and Raman spectroscopy. The cell parameters calculated from the XRD pattern (Figure 1a) are $a = 0.3081$ and $c = 1.0061$ (2) nm and a band gap of 3.26 eV at room temperature. Novel SiC-based DMS devices have been developed and gained considerable attention because of their excellent physical properties. Here, we report the observation of glassy FM in Al-doped 4H-SiC for the first time. The Al element serves two roles: (1) induce long-range magnetic order and (2) stabilize the 4H-SiC polytypes. This study raises some basic questions toward understanding the origin of spin order in wide-gap semiconductor and may open an alternative route toward designing high-temperature SiC-based DMSs.

In a typical run, high-purity silicon (99.999%), carbon (99.9995%), and aluminum (99.997%) powder, supplied by Alfa Aesar, were selected to synthesize the samples (see Supporting Information, SI). Inductively coupled plasma–atomic emission spectrometry (ICP–AES) was used to determine the dopant concentration of aluminum with a ~0.75 atom% in the as-prepared sample. The sample (mass 133.4 mg) was put in a capsule for magnetic measurement (see SI). We characterized the sample structure by high-resolution X-ray diffraction (XRD) and Raman spectroscopy. The cell parameters calculated from the XRD pattern (Figure 1a) are $a = 0.3083(2)$ and $c = 1.0093(6)$ nm corresponding to 4H-SiC (ICDD-PDF: 29-1127, space group P63/mmc). Raman spectra (Figure 1b) reveal that the profile of Al-doped 4H-SiC is almost the same as the pristine 4H-SiC wafer, using the folded transverse optic (FTO) mode, which is quite sensitive to the type and degree of crystallization, peaking at 796 cm$^{-1}$. No contributions from other SiC polytypes or secondary phases within the sensitivity of XRD were observed at both 200 and 300 K, indicating FM order dominates the entire temperature range. For the undoped sample, the loop was not observed even at 4 K. Zero-field-cooled (ZFC) and field-cooled (FC) temperature dependence of dc magnetization was measured (see Figure S1). In the field of 1 kOe, the curves exhibit a pronounced FC-ZFC irreversibility as indicated by arrows and a cusp in ZFC curve. A weak irreversibility occurred at ~90 K, and a strong deviation from that in FC run was observed at ~16.4 K in ZFC curve, which is usually ascribed to the freezing of transversal and longitudinal spins, respec-

Figure 1. (a) X-ray diffraction pattern of Al-doped 4H-SiC; (b) Raman spectra of Al-doped 4H-SiC (lower curve) and reference 4H-SiC wafer (upper curve).

Figure 2. Hysteresis loops for Al-doped 4H-SiC, observed in the range $-5$ kOe < $H$ < 5 kOe, at 200 K (circles) and 300 K (triangles). Inset shows the hysteresis loops at $T = 4$ K (squares) for undoped 4H-SiC obtained at the same experimental conditions.
only induced by defects since the itinerant FM could originate substantially from some special structural defects. In fact, the doping Al in SiC may introduce large scales of defects into the lattice, such as vacancies and interstitials. The possible contribution of these defects for FM in Al-doped 4H-SiC needs to be investigated further. In addition, we note that other doping elements (such as Fe, Co, Ni, etc.) can only lead to the mixture of 6H-SiC (main phase) and other SiC polytypes phases under the same conditions. Here, it is speculated that Al element serves as the key role to stabilize the SiC crystal structure inherent to 4H and the true mechanism is unclear.

In conclusion, we synthesized Al-doped 4H-SiC, and magnetic properties measurement showed a typical glassy FM feature. We propose a possible explanation for the origin of FM order: the coefficient of $sp^3/sp^2$ configuration along with the structural defects. Our work demonstrates that the aluminum is a novel dopant in SiC that exhibit two striking features, (1) induces FM order; (2) stabilizes crystal structure, and has exposed the exciting possibility of doping nonmagnetic atoms to control the spin moments in wide-gap semiconductor.

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**Supporting Information Available:** Experimental process, low temperature magnetic measurements. This material is available free of charge via the Internet at http://pubs.acs.org.

References

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