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Investigation of magnetic order and spin dynamics in Mn doped 3C-SiC

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A B S T R A C T

As SiC is a technological important material which is being used for a wide variety of device applications that can work under harsh environment, inducing magnetic order in this material without altering its other physical properties can lead to the development of multifunctional magneto electronics devices, that can operate under extreme conditions such as high power, high temperature and high radiation. We have synthesized Mn doped 3C-SiC (Mn-1%, 3% and 5%) by thermal plasma technique and investigated the nature of magnetic interactions, anisotropy and its temperature dependence using DC, AC magnetometry and magnetic resonance studies. The samples exhibit ferromagnetic transition with high Curie temperature above 780 K. Isothermal DC magnetic measurement shows Hysteresis with coercivity value of 240 Oe at 5 K which reduces to 40 Oe at room temperature. There is spin reorientation below 50 K. The magnetic anisotropy decreases with increase in temperature. Detailed analysis of DC and AC magnetization measurements and temperature dependence of electron paramagnetic resonance spectra indicates that the long range magnetic order may be due to interaction among Mn^{2+} ions mediated by holes.

1. Introduction

Recent reports in literature suggests that it is possible to induce long range ferromagnetic order in Dilute Magnetic Semiconductors (DMS) [1] which has generated renewed interest in this field. DMS is a semiconductor doped with a small quantity of 3d transition metal ions carrying unpaired spin to induce magnetism in the host material which may lead to the development of novel spintronic devices. The idea of inducing magnetic order by doping transition metal ferromagnetic elements in semiconductors emerged, as there are no room temperature magnetic semiconductors available. As the attempts to induce magnetism in the most commonly used semiconductor, Si failed, the focus shifted to wide band gap semiconductors. Dietl et al. predicted through ab initio calculations that 5% Mn doped wide band gap semiconductors GaN and ZnO will be room temperature ferromagnets [2]. But the experimental results did not show clear ferromagnetic transition above room temperature [3,4]. Then the focus shifted to other wide band gap oxide semiconductors such as TiO_2, SnO_2, $BaSnO_3$, $SrSnO_3$ etc [1,5–7]. Among these the Co doped TiO_2 has been confirmed to be a room temperature ferromagnetic semiconductor based on various experimental investigations [1]. As SiC is a technologically important wide band gap semiconductor which is useful for high temperature and high power electronics application, it is worthwhile to explore the possibility of inducing ferromagnetism by doping transition metal elements. Among the various transition metal explored for inducing magnetism in 3C-SiC, the Mn substitution for SiC holds promise for spintronic device application. Though there have been some reports in literature on Mn doped 3C-SiC to the best of our knowledge there are no reports on detailed quantitative analysis of magnetic interactions, anisotropy and its temperature dependence [8–16]. We have synthesized bulk Mn doped 3C-SiC using raw rice husk in an indigenously developed plasma reactor and carried out detailed analysis of temperature dependence of Electron paramagnetic resonance, DC Magnetization and temperature and frequency dependence of AC susceptibility.

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2. Experimental procedure

The Mn doped bulk SiC polycrystalline samples were prepared by carbothermal reduction method. Rice husk was mixed with appropriate amounts of $Mn(CH_3COO)_2$.The growth temperature and time were fixed at 1600 °C and 15-mins, respectively.The detailed synthesis procedure of SiC from rice husk can be found elsewhere [15]. Typical experimental conditions are Argon gas flow-2-LPM; current-50 A, and load voltage-300 V. The possible reactions for the formation of SiC from Rice Husk can be written as follows:

SiO_2 (amorphous) + 3C (amorphous) → SiC + 2CO.

The chemical reaction for Mn incorporation in to SiC is as follows

$Mn(CH_3COO)_2 + 2\,SiO_2 + 3C \rightarrow MnSiC + SiC + 3H_2O + 5CO$

The structural characterisation was carried out using Rigaku Smart lab X-ray Diffractometer with 9 kW power generator at room temperature and Horiba Jobin- Yvon HR-800 Micro Raman spectrometer with 488 nm laser wavelength. While a Quantum Design made 9 T PPMS based - Vibrating Sample Magnetometer (VSM) was employed for the low temperature magnetization measurements, the high temperature measurements were carried out using a Lakeshore model 7400 VSM. AC susceptibility measurements were carried out using Quantum Design Physical property measurement system. The valence state of unpaired electrons, Dipolar interaction and anisotropy in the system was probed using electron paramagnetic resonance spectrometer (EPR) JEOL model JES FA 200.

3. Results and discussion

3.1. Structural investigation

From X-ray diffraction (XRD) it is clearly visible that the peak shifts towards lower angle which can be attributed to larger Mn atom substituting for smaller Si atom in SiC shown in Fig. 1. The obtained XRD pattern matched quite well with the JCPDS data for 3C-SiC (# 02–1050). The samples have crystallized in cubic

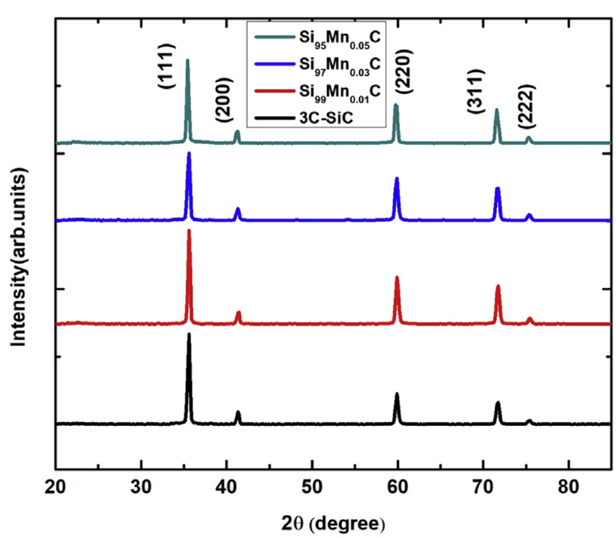

Fig. 1. XRD pattern of undoped and Mn doped 3C-SiC showing the single phase nature of the samples.

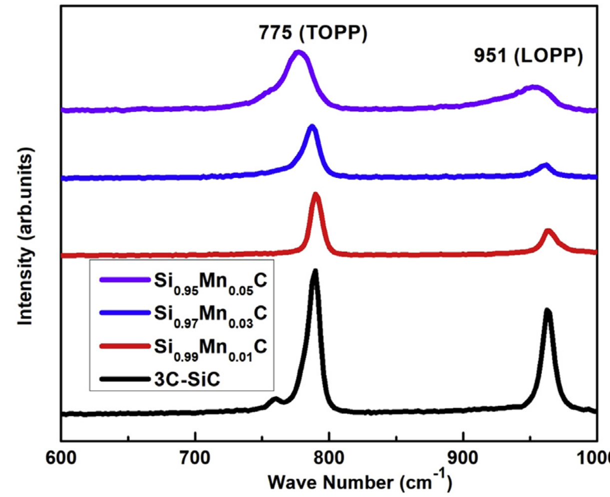

Fig. 2. Raman spectra of undoped and Mn doped SiC. Clear shift in peak position and broadening of spectra with decrease in intensity can be observed as the Mn concentration increases.

structure with *F4-3m* space group. The corresponding lattice parameter is $a = 4.30$ Å. Formation of silica is greatly reduced to insignificant level because of short duration of holding time and rapid heating during the synthesis of SiC.

The shift in peak position towards lower wave number and increase in full width at half maximum (FWHM) of the Raman spectra with increase in Mn concentration can be attributed to distribution in bond length and bond angle within the tetrahedral structure of SiC as shown in Fig. 2. Which confirms the incorporation of Mn in host SiC matrix. There is no such significant contribution from strain has been found.

3.2. Electron paramagnetic resonance (EPR) studies:

EPR measurements were carried out using a JEOL X-band (frequency −9.56 GHz) spectrometer using a rectangular cavity with 100 kHz field modulation and phase sensitive detection. The EPR absorption peaks for pure 3C-SiC is located around 338.05 mT,

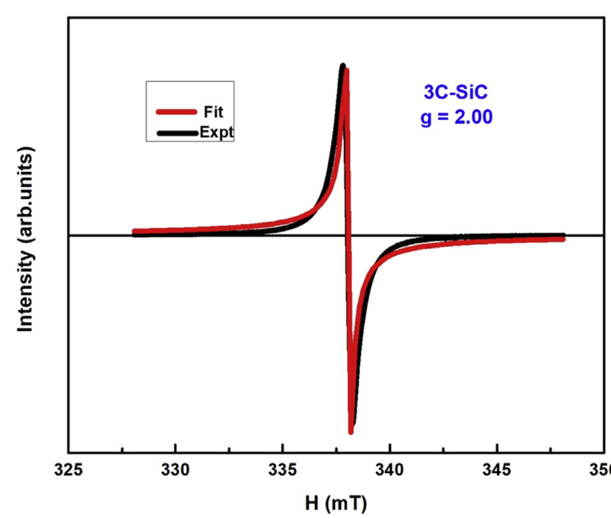

Fig. 3. EPR (X-band f = 9.56 GHz) spectra of pure 3C-SiC powder sample showing resonance at Hr = 338.05 mT with a spectral width, ΔHpp = 0.9 mT indicating the presence of weak magnetic interactions in the system.