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Engineering of ion-doping on the ground states and Bose–Einstein condensation of $Sr_3Cr_2O_8$

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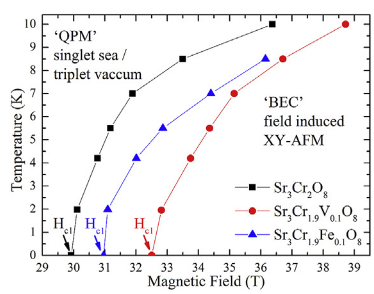
HIGHLIGHTS

- Single crystals of $Sr_3Cr_2O_8$, $Sr_3Cr_{1.9}V_{0.1}O_8$ and $Sr_3Cr_{1.9}Fe_{0.1}O_8$ were grown.
- Magnetization up to 54 T were investigated at the temperature from 2 K to 20 K.
- A magnetic field induced Bose–Einstein condensation were observed.
- We obtained the lower field phase diagrams of these three single crystals.
- The critical field of BEC and ion-dopants are correlative to each other.

GRAPHICAL ABSTRACT



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ABSTRACT

Single crystals of $Sr_3Cr_2O_8$, $Sr_3Cr_{1.9}V_{0.1}O_8$ and $Sr_3Cr_{1.9}Fe_{0.1}O_8$ were grown using the floating zone technique, and their magnetization behaviors were systematically investigated under pulsed field up to 54 T at various temperatures ranging from 2 K to 20 K. Temperature-induced transition from singlet ground state to triplet state and magnetic-field-induced Bose–Einstein condensation were observed, of which the critical temperature and the critical field are correlative to ion-dopants. The analysis of the ground state and low-field magnetization behaviors indicates that the ion-dopants lead to an enhancement of the intradimer exchange J_0, which changes the transition temperatures, and increases the critical fields H_{c1}. The magnetic Fe ion dopant makes the transition from singlet to triplet states more sensitive to the applied magnetic field, in clearly contrast to the situation of the nonmagnetic V^{5+} dopant.

1. Introduction

Over the last decades, many attentions have been focused on the studies of the Bose–Einstein condensation (BEC), the new kind BEC was first experimentally observed in $TlCuCl_3$ [1–3], which has a singlet ground state with an excitation gap. Later, this kind of BEC was observed in many quasi-2D systems, for example $BaCuSi_2O_6$ [4,5]. In a high magnetic field (H), M. Jaime et al. observed the closure of the gap in $BaCuSi_2O_6$ and a gap of Bosonic spin triplet excitations. A high magnetic field leads to the closure of the field-induced gap in the magnon spectrum of Cs_2CuCl_4 at a critical

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field, and magnetic phase transition was observed below the critical field [6]. The BEC was also observed in organic compounds, e.g. V. S. Zapf et al. [7] found that a magnetic field induced BEC of the Ni spin degrees of freedom in $NiCl_2$–4 $SC(NH_2)_2$ (DTN). B. S. Conner et al. [8] also proved the $S = 1/2$ spin-dimer compound $Pb_2V_3O_9$ to be a new material with field-induced BEC. Anomalous dynamical line shapes in a quantum magnet at finite temperature were observed in the one-dimensional dimer Heisenberg chain compound $[Cu(NO_3)_2 \cdot 2.5(D_2O)]$ [9]. The BEC has been observed in neutron scattering [10,11], nuclear magnetic resonance (NMR) [12–14], high magnetic field measurement [15] and so on. There are also a lot of models have been put forward in theory papers. R. Yu et al. developed models for both non-magnetic and magnetic dopant ions [16]. They indicated certain kinds of magnetic disorder in BEC systems can induce a Bose-Glass phase characterized by local regions of antiferromagnetic (AFM) order that are not coherent with each other. T. Roscilde made use of a local-gap model, performed quantum Monte Carlo simulations on a site-diluted bilayer system, found a sequence of three distinct quantum-disordered phases induced by the field [17].

Recently, the compounds $Ba_3Mn_2O_8$ [18–20], $Ba_3Cr_2O_8$ [21], and $Sr_3Cr_2O_8$ [22] have been reported as new spin-dimer compounds with a BEC behavior. S. Majumdar et al. have discussed the magnetic properties of spin-dimer compounds $Ba_3Mn_2O_8$ doped with La and V [23], E. C. Samulon et al. reported the heat capacity properties of $Ba_3(Mn_{1-x}V_x)O_8$ [24], and $Sr_3Cr_2O_8$ doped with V [25]. Tao Hong et al. [26] have discussed the structural and magnetic properties in the V doped $Ba_3Cr_2O_8$. These compounds have magnetic ions Mn^{5+} ($S = 1$) or Cr^{5+} ($S = 1/2$), and crystallize in a trigonal crystal structure with the space group $R\overline{3}m$. In these compounds, the spin-dimer along the hexagonal c-axis is characterized by intradimer interaction J_0 and the interdimer interactions J' (where J' is the sum of all interdimer interactions J_1, J_2 and J_3 between Cr^{5+} (or Mn^{5+}) ions). Early in 2008, T. Giamarchi, C. Rüegg and O. Tchernyshyov reviewed the BEC in magnetic insulators [27] and gave their outlook. And V. Zapf and M. Jaime reviewed experimental and theoretical work on BEC in quantum magnets in 2014, and gave a list of quantum magnets for which the three-dimensional (3D) BEC has been studied [28].

In this paper, the high magnetic field magnetization behaviors of Fe and V doped $Sr_3Cr_2O_8$ single crystals up to 54 T have been investigated. The corresponding temperatures of the minimum magnetization and the maximum one at the finite-temperature phase transition were measured. Based on the Curie–Weiss law and the Bleaney–Bowers equation for spin-1/2 dimer [22], we discussed the influence of the sizes of the doped ions and their magnetic properties on the field-induced transition temperature (especially on the condensed temperature of the spin-dimer), on the intradimer interaction J_0 and on the other transition parameters. At last, we obtained the BEC phase diagrams of $Sr_3Cr_2O_8$, $Sr_3Cr_{1.9}V_{0.1}O_8$ and $Sr_3Cr_{1.9}Fe_{0.1}O_8$.

These spin-dimer systems provide an excellent opportunity to study the critical behavior in the immediate vicinity of a BEC quantum critical point, because the high degree of homogeneity in boson density offered by crystalline solids is difficult to attain in the presence of a trap potential confining atomic gas. In this paper, we will measure the effect of the dopants, temperature and magnetic field on the magnetization behaviors of $Sr_3Cr_2O_8$, $Sr_3Cr_{1.9}V_{0.1}O_8$ and $Sr_3Cr_{1.9}Fe_{0.1}O_8$ to investigate the transition behavior from singlet to triplet phases and BEC. Since the Fe and V dopants have different sizes and measurable differences in the lattice parameters, they may cause local structural distortions around the magnetic ions. This could cause the uniaxial isotropy around the spins to change, and therefore make the BEC description more or less valid.

2. Materials and methods

Single crystal samples of parent $Sr_3Cr_2O_8$, ion-doped $Sr_3Cr_{1.9}V_{0.1}O_8$ and $Sr_3Cr_{1.9}Fe_{0.1}O_8$ were grown using an optical floating zone furnace (Crystal Systems Inc. Model FZ-T-10000-H-VII-VPO-PC). The detailed synthesized process is similar with Refs. [29,30]. Phase purity of the obtained single crystals was confirmed by powder x-ray diffraction (XRD) and electron diffraction pattern. Their XRD patterns were measured at room temperature using an X'Pert PRO (PANalytical B.V.) diffractometer with Cu-Kα radiation. The electron diffraction pattern was measured using a Tecnai G2 20 U-TWIN TEM. Low-field magnetization behaviors were measured using a commercial superconducting quantum interference device (SQUID, Quantum Design). High-field magnetization behaviors were measured utilizing a 54 T nondestructive pulsed magnet (with pulse duration of 58 ms) at Wuhan National High Magnetic Field Center (WHMFC).

3. Results

3.1. Structure

Fig. 1 (a), (b) and (c) show the XRD patterns of $Sr_3Cr_2O_8$, $Sr_3Cr_{1.9}V_{0.1}O_8$ and $Sr_3Cr_{1.9}Fe_{0.1}O_8$, respectively, which indicate all the samples are single phase with a trigonal $R\overline{3}m$ structure, and no impurities are detected. The inset of Fig. 2 shows the crystal structure of Cr^{5+} ions in $Sr_3Cr_2O_8$, and the coupling interactions

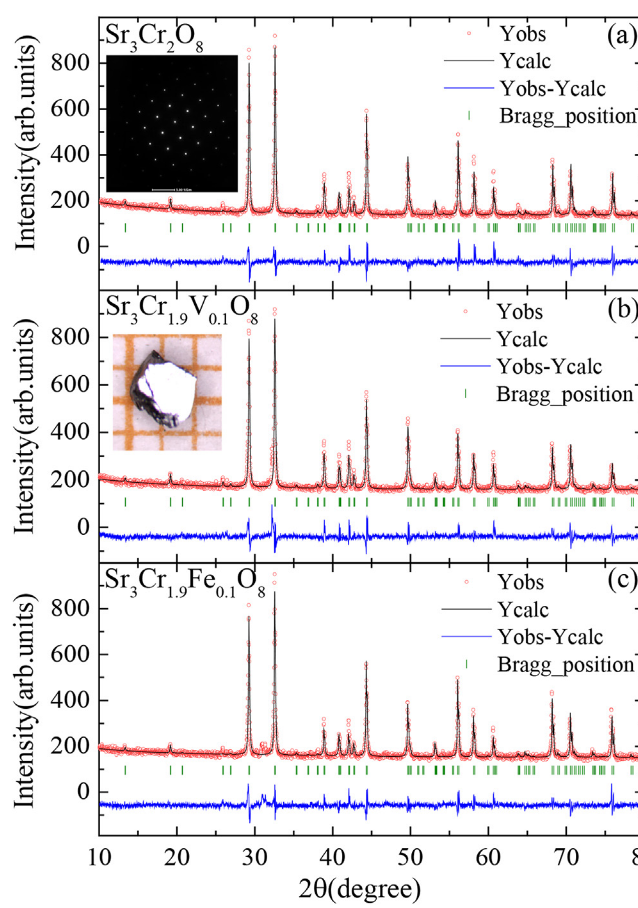

Fig. 1. X-ray diffraction patterns of (a) $Sr_3Cr_2O_8$, (b) $Sr_3Cr_{1.9}V_{0.1}O_8$ and (c) $Sr_3Cr_{1.9}Fe_{0.1}O_8$. The inset of (a) shows the electron diffraction pattern of $Sr_3Cr_2O_8$ got from TEM measurement. The inset of (b) shows one smashed single crystal of $Sr_3Cr_{1.9}V_{0.1}O_8$.