Journal of Magnetism and Magnetic Materials 446 (2018) 260–263

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Research articles

On magnetic structure of $CuFe_2Ge_2$: Constrains from the ^{57}Fe Mössbauer spectroscopy

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ARTICLE INFO

Article history:
Received 23 June 2017
Received in revised form 16 August 2017
Accepted 18 September 2017
Available online 20 September 2017

Keywords:
Mössbauer spectroscopy
Magnetic order
Hyperfine parameters

ABSTRACT

^{57}Fe Mössbauer spectroscopy measurements were performed on a powdered $CuFe_2Ge_2$ sample that orders antiferromagnetically at ~ 175 K. Whereas a paramagnetic doublet was observed above the Néel temperature, a superposition of paramagnetic doublet and magnetic sextet (in approximately 0.5:0.5 ratio) was observed in the magnetically ordered state, suggesting a magnetic structure similar to a double-*Q* spin density wave with half of the Fe paramagnetic and another half bearing static moment of $\sim 0.5 - 1\mu_B$. These results call for a re-evaluation of the recent neutron scattering data and band structure calculations, as well as for deeper examination of details of sample preparation techniques.

1. Introduction

The recent discovery of superconductivity in iron-based compounds [1], followed by a flare of experimental and theoretical studies of related materials [2–5], restored interest in Fe-based intermetallics with either magnetic order or enhanced Fermi-liquid properties and possible strong magnetic fluctuations, renewing, for example, interest in the RFe_2Ge_2 (R = rare earth) series [6–12]. Among other materials the electronic structure and magnetism of $CuFe_2Ge_2$ [13], were investigated in some detail [14,15].

Unlike many other so-called 122 compounds, $CuFe_2Ge_2$ crystallizes in an orthorhombic structure (space group 51, *Pmma*), with a 2*a* site for Cu, 2*d* and 2*f* sites for Fe and 2*e* and 2*f* sites for Ge [13]. Band structure calculations [14] suggested that $CuFe_2Ge_2$ has a magnetic ground state that is ferromagnetic along *a* direction and antiferromagnetic in other directions. Calculated magnetic moments on two Fe sites differ by less than 5%.

Magnetization measurements in $CuFe_2Ge_2$ [15] showed an onset of a ferromagnetic-like transition at ≈ 228 K. On further cooling, multiple experimental techniques, including powder neutron diffraction, [15] identified a commensurate antiferromagnetic ordering below $T_N \approx 175$ K. The commensurate structure was described by the propagation vector (0, 1/2, 0), so that the moments are aligned antiferromagnetically along *b*, with chains of Fe(1) atoms ferromagnetically coupled along *a* and antiferromagnetically coupled with Fe(2) atoms. [15] The magnetic moments evaluated from the neutron diffraction data refinement were 0.36(10) μ_B on Fe(1) and 0.55(10) μ_B on Fe(2) at 135 K. An incommensurate spin density wave structure was reported to set in below ≈ 125 K with a coexistence of two structures between approximately 70 and 125 K. The incommensurate structure at 4 K was described by the propagation vector (0, 1/2, 0.117) with magnetic moments of 1.0(1) μ_B on Fe(1) and 0.71(10) μ_B on Fe (2). The direction of the moments in both commensurate and incommensurate magnetic phases was suggested to be along the *c*-axis direction.

$CuFe_2Ge_2$ was identified as a metallic compound with competing magnetic ground states, that are possibly strongly coupled to the lattice and easily manipulated using temperature and applied magnetic felds. [14,15] Additionally, powder neutron diffraction data allowed for some ambiguity in the modeling of the data. [15] All this suggested that further studies, in particular with other local probes, would be desirable to gain understanding of magnetism in this compound.

In this work we use ^{57}Fe Mössbauer spectroscopy to perform a study of $CuFe_2Ge_2$, over a large temperature range that includes the paramagnetic and suggested magnetically ordered states.

2. Synthesis and general characterizatoion

Polycrystalline samples of $CuFe_2Ge_2$ were prepared by arc melting high purity elements on a water cooled copper hearth under ~ 10 mTorr of Ar atmosphere, followed by annealing. The weight loss after arc melting was $\sim 2\%$. The arc melted sample was put in an alumina crucible, sealed in an amorphous silica tube under a partial Ar atmosphere, and then annealed at 600 °C for 168 h

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https://doi.org/10.1016/j.jmmm.2017.09.046
0304-8853/

and furnace-cooled. Given that Ref. [15] emphasized the importance of annealing at this temperature, we made every effort to reproduce their annealing procedure. Note that in this work the bulk arc-melted button was annealed, whereas in Ref. [15] the arc-melted sample was ground and cold pressed, and then annealed. This is the only apparent difference between the sample preparation procedures.

Room temperature powder X-ray diffraction was performed using a Rigaku MiniFlex II diffractometer and zero diffraction, silicon sample holder. The results were analyzed using the GSAS software package [16]. The results (Fig. 1) suggest that the sample is a single phase, the refined lattice parameters are $a = 4.980$Å, $b = 3.970$ Å, and $c = 6.795$ Å, in agreement with the literature values [13].

Temperature dependent resistivity measurements were performed using a conventional four-probe technique and a Quantum Design Physical Property Measurement System ACT option ($f = 16$ Hz, $I = 3$ mA). Electrical contacts to the sample were made with Epo-Tek H20E conductive epoxy and were lower than 1Ω. The results of the measurements are shown in Fig. 2. The $RRR = \rho_{300\ K}/\rho_{1.8\ K}$ is about 3.8. The transition at ~ 175 K is clearly seen both in resistivity data and in its derivative. These data are consistent with the results of Ref. [15], Supplementary Information.

Temperature dependent magnetization was measured on bulk and powdered polycrystalline samples between 1.8 and 300 K for several values of applied magnetic field using a Quantum Design Magnetic Property Measurement System (MPMS 3) SQUID magnetometer. No discernible difference was observed between these two sets of data suggesting that if there is a texture (preferential orientation) in the bulk polycrystalline sample, it is insignificant. The results for powdered sample are shown in Fig. 3 (a). The feature associated with the transition at ~ 175 K is seen in all curves. The measurement at $H = 10$ Oe suggest presence of a ferromagnetic component below ~225–230 K, and the 1 kOe data also suggests a similar ferromagnetic component. The presence of this ferromagnetic component is evident in the magnetization loops shown in Fig. 3 (b). Distinct from the data in Ref. [15] we do not observe any apparent feature at $T_2 \approx 125$ K and the low temperature magnetization tail in our measurements is smaller. These differences could be due either to (different) preferential orientation/ texture of the polycrystalline samples, or to slightly different chemical compositions or small secondary magnetic phases possibly associated with the apparently larger low temperature tail in Ref. [15].

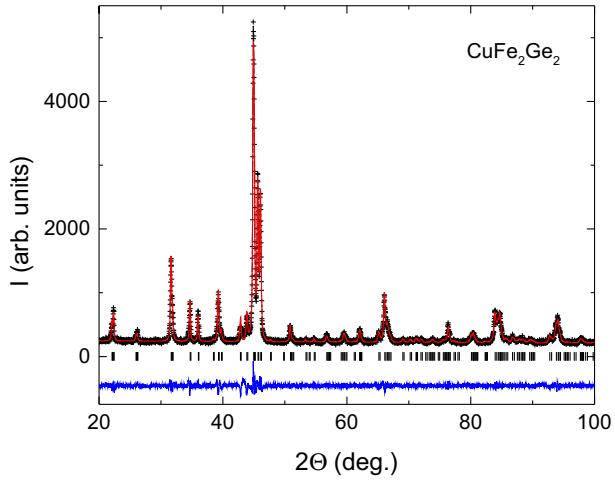

Fig. 1. Powder X-ray diffraction data at room temperature. Data (crosses), fit (red line), calculated peak positions (vertical bars) and the difference between experimental and calculated spectra (blue line) are shown. The refined lattice parameters are $a = 4.980$ Å, $b = 3.970$ Å, and $c = 6.795$ Å. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

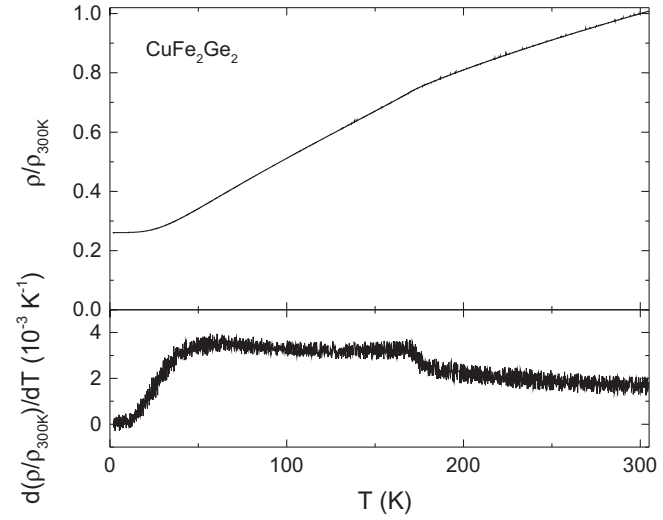

Fig. 2. Temperature-dependent resistivity of $CuFe_2Ge_2$ normalized to its value at 300 K (top panel) and its temperature derivative (bottom panel).

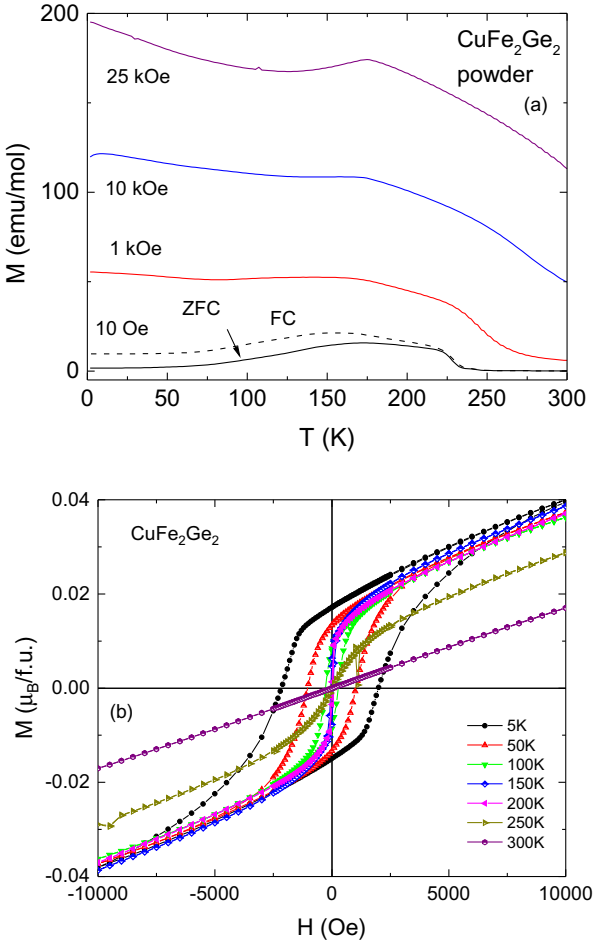

Fig. 3. (a) Temperature-dependent magnetization of powdered polycrystalline $CuFe_2Ge_2$ measured at four different values of applied magnetic field, 10 Oe (zero field cooled -ZFC and field cooled - FC) 1 kOe, 10 kOe and 25 kOe. (b) $M(H)$ magnetization loops measured at seven different temperatures on bulk piece of $CuFe_2Ge_2$