Supplementary Information for Spin density wave instability in a ferromagnet

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Here we present a summary of the crystal and magnetic structure of Fe_3Ga_4 determined from the X-ray and neutron diffraction experiments as well as a description of the refinement process to determine the magnetic structure in the intermediate temperature incommensurate phase.

Fe_3Ga_4 forms in a monoclinic ($C2/m$) crystal structure with no evidence of structural change associated with the magnetic phase transitions[1–5]. This structure is somewhat complex having 4 crystallographically unique Fe sites and 4 unique Ga sites within the unit cell (Fig. 1d and Table S1). The crystallographic parameters and atomic positions for Fe_3Ga_4 determined via single crystal X-ray diffraction at 300 K are provided in Table S1 to serve as a representative of all the data collected.

The results of the refinement of the neutron diffraction data are presented in Tables S2 and S3 which delineate both magnitude and orientation of the magnetic moments in the ferromagnetic low temperature state and the intermediate temperature spin density wave state. Previous neutron scattering investigations of the magnetic structure of Fe_3Ga_4 in polycrystalline samples were inconclusive[3,6,7].

The refinement process began with a representational analysis to determine the crystal structural symmetry allowed magnetic symmetry. The calculations were carried out using version 2K of the program SARA*h*-Representational Analysis[8]. They first involved the determination of the space group symmetry elements, g, that leave the propagation vector k invariant. Those elements form the little group $G_{\mathbf{k}}$. The magnetic representation of a crystallographic site can then be decomposed in terms of the irreducible representations (IRs) of $G_{\mathbf{k}}$:

$$\Gamma_{Mag} = \sum_{\nu} n_{\nu} \Gamma_{\nu}^{\mu} \tag{1}$$

where n_ν is the number of times that Γ_ν of order μ appears in the magnetic representation Γ_{Mag} for the chosen crystallographic site.

For Fe_3Ga_4, the crystal structure at temperatures above the phase transition is described by the space group C 2/m (#12:b1), which involves 2 centering operations and 4 symmetry operations. Only 2 operations leave the propagation k invariant or transform it into an equivalent vector. These IRs are Γ_1 and Γ_2. The decomposition of the magnetic representation Γ_{Mag} in terms of the two IRs of $G_{\mathbf{k}}$ for 7 Fe magnetic sites are examined. The basis vectors of Fe1-Fe5 sites only have contributions along the b-axis under Γ_1 and are confined in the ac-plane under Γ_2. All of the a, b, and c components are allowed on the Fe6 and Fe7 sites in both IRs. Based on our neutron diffraction results, only the Γ_2 IR works for the system. The Γ_2 IR associated basis vectors were applied to the FullProf[9] refinement and the refinement results supports the incommensurate spin density wave (ISDW) state with R_{factor} being 12.2A schematic of this magnetic state is shown in Fig. 1d and e and the magnetic moments are detailed in Table S3.

Since the incommensurate magnetic structures associated with ferromagnetic states are typically helimagnetic, we have carefully explored the possibility of the intermediate temperature state being a helical structure. A simulated annealing process was employed to search for the best fit model of a helical magnetic structure. We started from a model where the magnetic moments are confined to the ac plane, consistent with the symmetry analysis discussed above. The global ground state found under these conditions after 90 iterations retained a large R_{factor} of 40.2. Fi-

nally, we searched for a helical magnetic structure without constraining the spin direction. A model magnetic structure was found with magnetic moments rotating in the ab-plane having an R_{factor} of 13.2, comparable to that of the ISDW model. However, this state is not compatible with either the magnetic representation symmetry or the anisotropy we measure in the bulk magnetization (see Fig. 3a) so that it is not considered an acceptable magnetic structure for Fe_3Ga_4. Thus, the refinement procedure leads to the conclusion that the intermediate temperature state of Fe_3Ga_4 is an ISDW state which is consistent with our bulk measurements.

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Label	**Wyckoff Pos.**	**x**	**y**	**z**	$U_{eq}(\text{Å}^2)^a$
Fe1	2a	0	0	0	0.00314(18)
Fe2	4i	0.49454(7)	0	0.31074(9)	0.00373(14)
Fe3	4i	0.22477(7)	0	0.63799(9)	0.00320(14)
Fe4	8j	0.13570(5)	0.20036(6)	-0.15173(6)	0.00356(12)
Ga1	4i	0.26982(6)	0	0.08459(7)	0.00667(13)
Ga2	4i	0.04142(6)	0	0.34795(7)	0.00608(13)
Ga3	8j	0.60842(4)	0.20288(5)	0.15120(5)	0.00586(11)
Ga4	8j	0.35199(4)	0.18656(5)	0.44954(5)	0.00703(11)

Table S1: **Atomic Positions of Fe_3Ga_4 at 300 K determined from single crystal X-ray diffraction**: Space Group $C2/m$, a = 10.0949(16) Å, b = 7.6603(16) Å, c = 7.8703(14) Å, β = 106.286(2)°

Atom	**Atomic Coordinates**			**Moment(μ_B)**
	a	**b**	**c**	m_c
Fe1	0	0	0	1.54(28)
Fe2	0.4935(6)	0	0.3096(7)	1.20(22)
Fe3	0.2240(7)	0	0.6376(8)	1.24(21)
Fe4	0.1352(5)	0.2019(5)	0.8481(6)	1.49(12)
Average Moment				1.37(22)

Table S2: **Magnetic structure data for the low temperature ferromagnetic state**: magnetic moments determined from the refinement of the neutron scattering data at 5 K for the four in-equivalent crystallographic Fe sites. Moments were constrained to lie along the c-axis.

Atom	Atomic Coord.			Magnetic Moments			$m_{mean}(\mu_B)$				$m_{max}(\mu_B)$			
Site	a	b	c	a	b	c	a	b	c	$\|m\|$	a	b	c	$\|m\|$
1	0	0	0	-2.31(6)	0	0.58(2)	1.47(4)	0	0.37(1)	1.51(4)	2.31(6)	0	0.58(1)	2.38(6)
2	0.4935(6)	0	0.3096(7)	-1.55(4)	0	0.38(1)	1.02(2)	0	0.25(1)	1.05(4)	1.61(4)	0	0.40(1)	1.65(4)
3	0.5065(6)	0	0.6904(7)	-0.96(3)	0	0.24(1)	1.02(2)	0	0.25(1)	1.05(4)	1.61(4)	0	0.40(1)	1.65(4)
4	0.2240(7)	0	0.6376(8)	-0.95(3)	0	0.24(2)	1.16(3)	0	0.29(1)	1.20(5)	1.83(4)	0	0.46(1)	1.88(5)
5	0.7760(7)	0	0.3624(8)	-1.79(5)	0	0.45(2)	1.16(3)	0	0.29(1)	1.20(5)	1.83(4)	0	0.46(1)	1.88(5)
6(2)	0.1352(5)	0.2019(5)	0.8481(6)	-0.22(1)	0.06(1)	-0.05(1)	1.32(2)	0.37(4)	0.33(1)	1.41(8)	2.08(3)	0.58(6)	0.52(1)	2.22(8)
7(2)	0.8648(5)	0.2019(5)	0.1519(6)	-2.00(3)	-0.56(7)	0.50(1)	1.32(2)	0.37(4)	0.33(1)	1.41(8)	2.08(3)	0.58(6)	0.52(1)	2.22(8)
m_{mean}				0	0	0	1.24(3)	0.16(2)	0.31(1)	1.30(4)	-	-	-	-
m_{max}				-	-	-	-	-	-	-	2.31(6)	0.58(7)	0.58(2)	2.38(8)

Table S3: **Details of the magnetic structure of the spin density wave state**: Magnetic structural data for the spin density wave state at 100 K from refinement of our neutron scattering data. Data presented in this table include atomic coordinates determined from the structure refinement at T= 5 K, the magnetic moment in a representative single primitive cell labeled 'magnetic moments', the mean magnetic moment, m_{mean}, averaged over a large number of unit cells, and the amplitude, m_{max}, for each of the different crystallographic directions. The symmetry reduction in the incommensurate spin density wave state results in in-equivalent magnetic moments on 7 different Fe sites (6 and 7 have 2 fold symmetry).