Critical phenomenon of the near room temperature skyrmion material FeGe

Lei Zhang¹, Hui Han¹-², Min Ge³, Haifeng Du³, Chiming Jin¹, Wensen Wei¹, Jiyu Fan⁴, Changjin Zhang¹, Li Pi¹-³ & Yuheng Zhang¹,³

The cubic B20 compound FeGe, which exhibits a near room temperature skyrmion phase, is of great importance not only for fundamental physics such as nonlinear magnetic ordering and solitons but also for future application of skyrmion states in spintronics. In this work, the critical behavior of the cubic FeGe is investigated by means of bulk dc-magnetization. We obtain the critical exponents \( \beta = 0.336 \pm 0.004, \gamma = 1.352 \pm 0.003 \) and \( \delta = 5.267 \pm 0.001 \), where the self-consistency and reliability are verified by the Widom scaling law and scaling equations. The magnetic exchange distance is found to decay as \( J(r) \approx r^{-4.9} \), which is close to the theoretical prediction of 3D-Heisenberg model \( r^{-5} \). The critical behavior of FeGe indicates a short-range magnetic interaction. Meanwhile, the critical exponents also imply an anisotropic magnetic coupling in this system.

In recent years, skyrmion state, which is a topologically protected nanoscale vortex-like spin structure, has attracted great interest due to its potential application in spintronic storage function. It has been demonstrated that the skyrmion phase is thermodynamically stable magnetic vortex state in magnetic crystals. Writing and deleting single magnetic skyrmion have been realized in PdFe bilayer on Ir(111) surface. These findings pave a significant path to design quantum-effect devices based on the tunable skyrmion dynamics. The room-temperature skyrmion materials hosting stable skyrmion phase are paid considerable attention. The cubic FeGe belongs to the space group \( P2_31 \), in which the non-centrosymmetric cell results in a weak Dzyaloshinskii-Moriya (DM) interaction. The competition of DM interaction between the much stronger ferromagnetic exchange finally causes a long modulation period of a helimagnetic ground state. A bulk FeGe sample exhibits a long-range magnetic order at Curie temperature \( T_C = 278.2 \) K, and displays a complex succession of temperature-driven crossovers in the vicinity of \( T_C \). The skyrmion phase emerges in a narrow temperature range just below \( T_C \) in the filed range from 0.15 to 0.4 kOe. The existence of the near room temperature skyrmion phase in FeGe, to our knowledge the highest \( T_C \) in B20 skyrmion compounds, makes it one of the most promising candidates of the next generation spintronic devices. Recently, more stable skyrmion phase has been realized in FeGe thin film, and it has been claimed that the skyrmions can be tuned by the crystal lattice. On the other hand, multiple and complex magnetic interactions have also been found in FeGe. An inhomogeneous helimagnetic state has been discovered above \( T_C \) due to the strong precursor phenomena. More interestingly, it has been revealed that the helical axis (q-vector direction) orientates depending on temperature. At zero magnetic field, the helical axis is along the \( <100> \) direction below 280 K. With decreasing temperature, it changes to the \( <111> \) direction at 211 K.

In view of the potential application and abundant physics in FeGe, a deep investigation of its magnetic exchange is of great importance not only for fundamental physics such as nonlinear magnetic ordering and solitons but also for creation of a basic for future application of skyrmion states and other chiral modulations in spintronics. In this work, the critical behavior of FeGe has been investigated by means of bulk dc-magnetization. The critical exponents \( \beta = 0.336 \pm 0.004, \gamma = 1.352 \pm 0.003 \), and \( \delta = 5.267 \pm 0.001 \) are obtained, where the self-consistency and reliability are verified by the Widom scaling law and the scaling equations. These critical

¹High Magnetic Field Laboratory, Chinese Academy of Sciences, Hefei 230031, China. ²University of Science and Technology of China, Hefei 230026, China. ³Hefei National Laboratory for Physical Sciences at the Microscale, University of Science and Technology of China, Hefei 230026, China. ⁴Department of Applied Physics, Nanjing University of Aeronautics and Astronautics, Nanjing 210016, China. Correspondence and requests for materials should be addressed to L.Z. (email: zhanglei@hmfl.ac.cn)
behavior of FeGe indicates a short-range magnetic interaction with a magnetic exchange distance decaying as $J(r) \approx -r^{-4.9}$. The obtained critical exponents also imply an anisotropic magnetic coupling in FeGe system.

**Results and Discussion**

It is well known that the critical behavior for a second-order phase transition can be investigated through a series of critical exponents. In the vicinity of the critical point, the divergence of correlation length $\xi$ leads to universal scaling laws for the spontaneous magnetization $M_s$ and initial susceptibility $\chi_0$. Subsequently, the mathematical definitions of the exponents from magnetization are described as:

$$\varepsilon = -\frac{\varepsilon}{\xi} < 0, T < T_C$$  \hspace{1cm} (1)

$$\chi_0^{-1}(T) = \left(\frac{h_0}{M_0}\right)\varepsilon^\gamma, \varepsilon > 0, T > T_C$$  \hspace{1cm} (2)

$$M = DH^{1/\beta}, \varepsilon = 0, T = T_C$$  \hspace{1cm} (3)

where $\varepsilon = (T - T_C)/T_C$ is the reduced temperature; $M_0/h_0$ and $D$ are the critical amplitudes. The parameters $\beta$ (associated with $M_s$), $\gamma$ (associated with $\chi_0$), and $\delta$ (associated with $T_C$) are the critical exponents. Universally, in the asymptotic critical region ($|\varepsilon| < 0.1$), these critical exponents should follow the Arrott-Noakes equation of state:

$$\frac{H/M}{H/M} = \frac{(T - T_C)/T_C}{T_C} + \frac{(M/M_0)^{1/\beta}}{T_C}$$  \hspace{1cm} (4)

Therefore, the critical exponents $\beta$ and $\gamma$ can be obtained by fitting the $M_s(T)$ and $\chi_0^{-1}(T)$ curves using the modified Arrott plot of $M^{1/\beta}$ vs $(H/M)^{1/\beta}$. Meanwhile, $\delta$ can be generated directly by the $M(H)$ at the critical temperature $T_C$ according to Eq. (3).

Generally, the critical temperature $T_C$ can be roughly determined by the temperature dependence of magnetization $[M(T)]$. Figure 1(a) shows the $M(T)$ curves for FeGe under zero-field-cooling (ZFC) and field-cooling (FC) with an applied field $H = 100$ Oe. The $M(T)$ curves exhibit an abrupt decline with the increase of temperature, corresponding to the paramagnetic-helimagnetic (PM-HM) transition. A sharp peak is observed at $T = 278.5$ K. The inset of Fig. 1(a) gives $dM/dT$ vs $T$, where $T_C \approx 283$ K is determined from the minimum of the $dM/dT$ curve.
Wilhelm et al. has demonstrated that a long-range magnetic order occurs below 278.2 K, however, an inhomogeneous helical state has existed above that temperature due to the strong precursor phenomena. The higher $T_C$ determined here indicates the appearance of precursor phenomena which may be caused by the strong spin fluctuation. Figure 1(b) shows the isothermal magnetization $M(H)$ at 4 K, which exhibits a typical magnetic ordering behavior. The inset of Fig. 1(b) plot the magnified $M(H)$ in lower field regime, which shows that the saturation field $H_S \approx 3000$ Oe. No magnetic hysteresis is found on the $M(H)$ curve, indicating no coercive force for FeGe.

Usually, the critical exponents can be determined by the Arrott plot. For the Landau mean-field model with $\beta = 0.5$ and $\gamma = 1.10$, the Arrott-Noakes equation of state evolves into $H/M = A + BM^2$, the so called Arrott equation. In order to construct an Arrott plot, the isothermal magnetization curves $M(H)$ around $T_C$ are measured as shown in Fig. 2(a). The Arrott plot of $M^2$ vs $H/M$ for FeGe is depicted in Fig. 2(b). According to the Banerjee's criterion, the slope of line in the Arrott plot indicates the order of the phase transition: negative slope corresponds to first-order transition while positive to second-order one. Therefore, the Arrott plot of FeGe implies a second-order phase transition, in agreement with the specific heat measurement.

Four kinds of possible exponents belonging to the 3D-Heisenberg model ($\beta = 0.365$, $\gamma = 1.336$), 3D-Ising model ($\beta = 0.325$, $\gamma = 1.24$), 3D-XY model ($\beta = 0.345$, $\gamma = 1.316$), and tricritical mean-field model ($\beta = 0.25$, $\gamma = 1.0$) are used to construct the modified Arrott plots, as shown in Fig. 3 (a–d). All these four constructions exhibit quasi-straight lines in the high field region. However, all lines show an upward curvature and are not parallel to each other, indicating that the $\beta = 0.5$ and $\gamma = 1.0$ within the framework of Landau mean-field model is unsatisfied. Therefore, a modified Arrott plot should be employed.

One can see that the $NS$ of 3D-Heisenberg model is close to '1' mostly above $T_C$. This result indicates that the critical behavior of FeGe may not belong to a single universality class.
The precise critical exponents $\beta$ and $\gamma$ should be achieved by the iteration method\(^{36}\). The linear extrapolation from the high field region to the intercepts with the axes $M_{1/3}$ and $(H/M)^{1/\beta}$ yields reliable values of $M_s(T, 0)$ and $\chi^{-1}(T, 0)$, which are plotted as a function of temperature in Fig. 5(a). By fitting to Eqs. (1) and (2), one obtains a set of $\beta$ and $\gamma$. The obtained $\beta$ and $\gamma$ are used to reconstruct a new modified Arrott plot. Consequently, new $M_s(T, 0)$ and $\chi^{-1}(T, 0)$ are generated from the linear extrapolation from the high field region. Therefore, another set of $\beta$ and $\gamma$ can be yielded. This procedure is repeated until $\beta$ and $\gamma$ do not change. As one can see, the obtained critical exponents by this method are independent on the initial parameters, which confirms these critical exponents are reliable and intrinsic. In this way, it is obtained that $\beta = 0.336 \pm 0.004$ with $T_C = 283.18 \pm 0.05$ and $\gamma = 1.352 \pm 0.003$ with $T_C = 282.87 \pm 0.08$ for FeGe. The critical temperature $T_C$ from the modified Arrott plot is in agreement with that obtained from the derivative $M(T)$ curve, indicating strong critical fluctuation before the formation of the long-range ordering in FeGe\(^{24}\). This critical fluctuation is in agreement with the precursor phenomenon reported by Wilhelm \textit{et al.}\(^{28}\). The modulated precursor states and

Figure 3. The isotherms of $M_{1/3}$ vs $(H/M)^{1/\beta}$ with (a) 3D-Heisenberg model; (b) 3D-Ising model; (c) 3D-XY model; and (d) tricritical mean-field model.

Figure 4. The normalized slopes $[NS = S(T)/S(T_C)]$ as a function of temperature.
complexity of the magnetic phase diagram near the magnetic ordering are explained by the change of the character of solitonic inter-core interactions and the onset of specific confined chiral modulations\(^1\),\(^2\).

Figure 5(b) shows the isothermal magnetization \(M(H)\) at \(T_c = 283\, K\), with the inset plotted on a \(\log - \log\) scale. One can see that the \(M(H)\) at \(T_c\) exhibits a straight line on a \(\log - \log\) scale for \(H > H_s\). We determine that \(\delta = 5.297 \pm 0.001\) in the high field region \(H > H_s\). According to the statistical theory, these critical exponents should fulfill the Widom scaling law\(^3\):

\[
\delta = 1 + \frac{\gamma}{\beta}
\]  

As a result, \(\delta = 5.024 \pm 0.005\) is calculated according to the Widom scaling law, in agreement with the results from the experimental critical isothermal analysis. The self-consistency of the critical exponents demonstrates that they are reliable and unambiguous.

Finally, these critical exponents should obey the scaling equations. Two different constructions have been used in this work, both of which are based on the scaling equations of state. According to the scaling equations, in the asymptotic critical region, the magnetic equation is written as\(^2\):

\[
\varepsilon = M(H, \varepsilon) = \beta f_+ (H/\varepsilon^{\beta+\gamma}) \gamma
\]  

where \(f_+\) and \(f_-\) are regular functions denoted as \(f_+\) for \(T > T_c\) and \(f_-\) for \(T < T_c\). Defining the renormalized magnetization as \(m \equiv \varepsilon^{-\beta} M(H, \varepsilon)\), and the renormalized field as \(h \equiv H e^{-\beta+\gamma})\), the scaling equation indicates that \(m = m(h)\) for FeGe is replotted in Fig. 6(a), where all experimental data collapse onto two universal branches. The inset of Fig. 6(a) shows how \(m^2\) vs \(h/m\), where all \(M - T - H\) curves should collapse onto two independent universal curves. In addition, the scaling equation of state takes another form\(^2\):

\[
\frac{H}{M^\beta} = k \left( \frac{\varepsilon}{H^{1/\beta}} \right)
\]  

where \(k(x)\) is the scaling function. Based on Eq. (7), all experimental curves will collapse onto a single curve. Figure 6(b) shows how \(MH^{-1/\beta}\) vs \(\varepsilon H^{-1(\beta+\gamma)}\) for FeGe, where the experimental data collapse onto a single curve, and \(T_c\) locates at the zero point of the horizontal axis. The well-rescaled curves further confirm the reliability of the obtained critical exponents.

The obtained critical exponents of FeGe and other related materials, as well as those from different theoretical models are summarized in Table 1 for comparison. One can see that the critical exponent \(\gamma\) of FeGe is close to that

---

**Figure 5.** (a) The temperature dependence of \(M_s\) and \(\chi_0^{-1}\) for FeGe with the fitting solid curves; (b) the isothermal \(M(H)\) at \(T_c\) with the inset plane on \(\log - \log\) scale (the solid line is fitted).
of 3D-Heisenberg model, while $\beta$ approaches to that of 3D-Ising or 3D-XY mode, indicating that the critical behavior of FeGe do not belong to a single universality class. Anyhow, all these three models indicate a short-range magnetic coupling, implying the existence of short-range magnetic interaction in FeGe. As we know, for a homogeneous magnet, the universality class of the magnetic phase transition depends on the exchange distance $J_r$.

M. E. Fisher et al. have treated this kind of magnetic ordering as an attractive interaction of spins, where a renormalization group theory analysis suggests

$$J_r \propto r^{-\sigma}$$

where $d$ is the spatial dimensionality and $\sigma$ is a positive constant. Moreover, there is

$$40, 41$$

Table 1. Comparison of critical exponents of FeGe with different theoretical models and related materials (MAP = modified Arrott plot; Hall = Hall effect; AC = ac susceptibility; SC = single crystal; PC = polycrystal).
\[ \gamma \approx 1 + \frac{4n + 2}{d(n + 8)}\Delta\sigma + \frac{8(n + 2)(n - 4)}{d^2(n + 8)^2} \left( 1 + \frac{2G(d)}{(n - 4)(n + 8)} \right) \Delta\sigma^2 \]

(9)

where \( \Delta\sigma = (\sigma - \frac{d}{\pi}) \) and \( G(d) = 3 - \frac{1}{2} \left( \frac{d}{\pi} \right)^2 \), \( n \) is the spin dimensionality. For a three dimensional material \( (d = 3) \), we have \( J(\tau) \approx r^{-3^{(3-\sigma)}} \). When \( \sigma \geq 2 \), the Heisenberg model \( (\beta = 0.365, \gamma = 1.386 \text{ and } \delta = 4.8) \) is valid for the three dimensional isotropic magnet, where \( J(\tau) \) decreases faster than \( r^{-5} \). When \( \sigma \leq 3/2 \), the mean-field model \( (\beta = 0.5, \gamma = 1.0 \text{ and } \delta = 3.0) \) is satisfied, expecting that \( J(\tau) \) decreases slower than \( r^{-4.5} \). From Eq. (9) \( \sigma = 1.908 \pm 0.007 \) is generated for FeGe, thus close to the short-range magnetic coupling of \( \sigma \approx 2 \). Subsequently, it is found that the magnetic exchange distance decays as \( J(\tau) \approx r^{-1.49} \), which indicates that the magnetic coupling in FeGe is close to a short-range interaction. Moreover, we get the correlation length critical exponent \( \nu = 0.709 \pm 0.008 \) (where \( \nu = \sigma/\gamma, \xi = \xi_0(T - T_c)/T_c^{1/\nu} \)), and \( \alpha = (2 - \nu\gamma) = -1.127 \pm 0.008 \). Theory gives that \( \nu = -0.115(9) \) for 3D-Heisenberg model and \( \alpha = 0.110(5) \) for 3D-Ising model\(^{43,44} \). Therefore, these critical exponents indicates that the critical behavior in FeGe is close to the 3D-Heisenberg model with short-range magnetic coupling. However, the discrepancy of the critical exponents to 3D-Ising or 3D-XY models indicates an anisotropic magnetic exchange interaction.

As can be seen from Table 1, the critical exponents of Fe\(_{0.8}\)Co\(_{0.2}\)Si and Cu\(_2\)OSeO\(_3\), which also exhibit a helimagnetic and skyrmion phase transition with similar crystal symmetry, are close to the universality class of the 3D-Heisenberg model\(^{45,46} \), indicating a isotropic short-range magnetic coupling. However, the critical behavior of MnSi belongs to the tricritical mean field model\(^{47,48} \). In macroscopic view, the magnetic ordering in cubic FeGe is a DM spiral similar to the structure observed in the isostructural compound MnSi\(^{49} \). However, in microscopic view, the magnetic coupling types in these two helimagnets are different. The critical behavior of FeGe is roughly similar to those of Fe\(_{0.8}\)Co\(_{0.2}\)Si or Cu\(_2\)OSeO\(_3\), except a magnetic exchange anisotropy. In MnSi the spiral propagates are along equivalent <111> directions at all temperatures below \( T_c = 29.5 \) K. However, it has been revealed that the helical axis \( (q-v) \text{ vector direction} \) in FeGe depends on temperature. It is along the <001> direction below 280 K, and changes to the <111> direction in a lower temperature range at 211 K with the decrease of temperature at zero magnetic field\(^{20} \). This unique change of helical axis in FeGe may be correlated with the anisotropy of magnetic exchange in this system, since the magnetic exchange anisotropy also plays an important role in determination of the spin ordering direction. In addition, it should be expounded that the magnetic exchange anisotropy is essentially different from the magnetocristalline anisotropy. The magnetocristalline anisotropy is correlated to the crystal structure, while magnetic exchange anisotropy originates from the anisotropic magnetic exchange coupling \( J \).

**Conclusion**

In summary, the critical behavior of the near room temperature skyrmion material FeGe has been investigated around \( T_c \). The reliable critical exponents \( (\beta = 0.336 \pm 0.004, \gamma = 1.352 \pm 0.003, \text{ and } \delta = 5.267 \pm 0.001) \) are obtained, which are verified by the Widom scaling law and scaling equations. The magnetic exchange distance is found to decay as \( J(\tau) \approx r^{-1.49} \), which is close to that of 3D-Heisenberg model \( (r^{-3}) \). The critical behavior indicate that the magnetic interaction in FeGe is of short-range type with an anisotropic magnetic exchange coupling.

**Methods**

A polycrystalline B20-type FeGe sample was synthesized with a cubic anvil-type high-pressure apparatus. The detailed preparing method was described elsewhere, and the physical properties were carefully checked [H. Du et al., Nat. Commun. 6, 8504 (2015)]. The chemical compositions were determined by the Energy Dispersive X-ray (EDX) Spectrometry as shown in Fig. S1 and Table S I, which shows the atomic ratio of Fe: Ge \( \approx 50.52: 49.48 \). The magnetization was measured using a Quantum Design Vibrating Sample Magnetometer (SQUID-VSM). The no-overshoot mode was applied to ensure a precise magnetic field. To minimize the demagnetizing field, the sample was processed into slender ellipsoid shape and the magnetic field was applied along the longest axis. In addition, the isothermal magnetization was performed after the sample was heated well above \( T_c \) for 10 minutes and then cooled under zero field to the target temperatures to make sure curves were initially magnetized. The magnetic background was carefully subtracted. The applied magnetic field \( H_0 \) has been corrected into the internal field as \( H = H_0 - NM \) (where \( M \) is the measured magnetization and \( N \) is the demagnetization factor) [A. K. Pramanik et al., Phys. Rev. B 79, 214426 (2009)]. The corrected \( H \) was used for the analysis of critical behavior.

**References**

1. Ro/Jer, U. K., Bogdanov, A. N. & Pfeifer, C. Spontaneous skyrmion ground states in magnetic metals. Nature (London) 442, 797–801 (2006).
2. Muhlbauer, S. et al. Skyrmion lattice in a chiral magnet. Science 323, 915–919 (2009).
3. Munzer, W. et al. Skyrmion lattice in the doped semiconductor Fe\(_{1-x}\)Co\(_x\)Si. Phys. Rev. B 81, 041203 (2010).
4. Yu, X. Z. et al. Real-space observation of a two-dimensional skyrmion crystal. Nature (London) 465, 901–904 (2010).
5. Seki, S., Yu, X. Z., Ishiwata, S. & Tokura, Y. Observation of skyrmions in a multiferroic material. Science 336, 198–201 (2012).
6. Du, H. F., Wang, W., Tian, M. L. & Zhang, Y. H. Field-driven evolution of chiral spin textures in a thin helimagnet nanodisk, Phys. Rev. B 87, 014401 (2013).
7. Neubauer, A. et al. Topological Hall effect in the A phase of MnSi. Phys. Rev. Lett. 102, 186602 (2009).
8. Du, H. F. et al. Highly stable skyrmion state in helimagnetic MnSi nanowires. Nano Lett. 14, 2026–2032 (2014).
9. Nagaosa N. & Tokura, Y. Topological properties and dynamics of magnetic skyrmions. Nat. Nanotechnol. 8, 899–911 (2013).
10. Jonietz, F. et al. Spin transfer torques in MnSi at ultralow current densities. Science 330, 1648–1651 (2010).
11. Fert, A., Cros, V. & Sampaio, J. Skyrmions on the track. Nat. Nanotechnol. 8, 152–156 (2013).
Critical phenomenon of the near room temperature skyrmion material: Zhang, L. et al. Scientific Reports 6:22397, doi: 10.1038/srep22397 (2016).

Acknowledgements
This work was supported by the State Key Project of Fundamental Research of China through Grant No. 2011CB900111, the National Natural Science Foundation of China (Grant Nos 11574322, U1332140, 11004196, U1232142, 11474290, 11104281, and 11204288), the Foundation for Users with Potential of Hefei Science Center (CAS) through Grant No. 2015HSC-UP001 the Youth Innovation Promotion Association CAS No.2015267.

Author Contributions
Y.H.Z. conducted the analyses. L.Z. conducted all of the experiments and wrote the paper. H.F.D., C.M.J. and W.S.W. synthesized the sample. H.H. collected the EDX spectrum. M.G. performed the magnetic measurements. J.Y.F., C.J.Z. and L.P. analyzed the experimental results.

Additional Information
Supplementary information accompanies this paper at http://www.nature.com/srep

Competing financial interests: The authors declare no competing financial interests.

How to cite this article: Zhang, L. et al. Critical phenomenon of the near room temperature skyrmion material FeGe. Sci. Rep. 6, 22397; doi: 10.1038/srep22397 (2016).

This work is licensed under a Creative Commons Attribution 4.0 International License. The images or other third party material in this article are included in the article’s Creative Commons license, unless indicated otherwise in the credit line; if the material is not included under the Creative Commons license, users will need to obtain permission from the license holder to reproduce the material. To view a copy of this license, visit http://creativecommons.org/licenses/by/4.0/