Polar nematic state in an iron-based superconductor LaFeAsO1-xHx

Sachiko Maki
Tokyo Institute of Technology

Jun-ichi Yamaura (✉ jyamaura@lucid.msl.titech.ac.jp )
Tokyo Institute of Technology  https://orcid.org/0000-0002-8992-9099

Soshi Iimura
Tokyo Institute of Technology

Hitoshi Abe
High Energy Accelerator Research Organization

Hajime Sagayama
High Energy Accelerator Research Organization

Reiji Kumai
High Energy Accelerator Research Organization  https://orcid.org/0000-0002-5320-0028

Youichi Murakami
KEK

Satoru Matsuishi
Materials Research Center for Element Strategy

Hideo Hosono
Tokyo Institute of Technology  https://orcid.org/0000-0001-9260-6728

Article

Keywords: Bipartite Magnetic Parent Phases, Phase Transitions, Superconductivity

DOI: https://doi.org/10.21203/rs.3.rs-77544/v1

License: This work is licensed under a Creative Commons Attribution 4.0 International License. Read Full License
Abstract

High critical temperature ($T_c$) superconductivity is generally considered to result from a fluctuation-mediated Cooper-pairing derived from a parent phase. The question of what type of fluctuation forms in materials thus plays a key role in understanding the mechanism of superconductivity. The iron-based superconductor LaFeAsO$_{1-x}$H$_x$ possesses bipartite magnetic parent phases with centrosymmetric ($x \sim 0$) and non-centrosymmetric ($x \sim 0.5$) structures. The latter is an intriguing polar-metal phase induced by temperature and carrier doping. Here, we investigate average and local structures of LaFeAsO$_{1-x}$H$_x$ using X-ray diffraction and extended X-ray absorption fine-structure measurements. We found lattice $C_4$ symmetry breaking far above the structural transition temperature, and the signature of a tiny split in As–Fe bond distances with broken spatial inversion symmetry in a wide temperature/doping range. The former reveals a nematic state, and the latter highlights a fluctuated state of polar structure which can be appropriately called polar nematic state.

Introduction

Spatial inversion symmetry is intimately involved in the physics of matter, thereby its breaking can trigger exotic quantum phenomena. For example, metallic materials with polar crystallographic structure—polar metals—offer the opportunity to explore interesting physics such as unconventional superconductivity, nonreciprocal transport, and inverse Faraday effect$^{1-4}$. While over 60 polar metals have been reported to date$^5$, a temperature-induced polar metal state as suggested by Anderson and Blount in 1965 is rare because screenings of conduction electrons hamper the formation of a macroscopic electrostatic field$^6$. Shi et al. reported an example of a ferroelectric-like transition while maintaining the metallic behaviour in LiOsO$_3$.$^7$ On rising temperature or doping carrier, such a polar metal will recover its centrosymmetric structure; thus, one would expect a fluctuating state, a quantum critical point, and a nematic state adjacent to the polar metal phase$^8-10$.

Studies of high-$T_c$ iron-based superconductors have experienced spectacular growth in condensed matter physics since their discovery$^{11-13}$. We believe that the most promising material for studying polar metals is an iron oxypnictide LaFeAsO$_{1-x}$H$_x$ which is a hydrogen-substituted version of the prototype iron-based superconductor LaFeAsO$_{1-x}$F$_x$.$^{14}$ The LaFeAsO$_{1-x}$H$_x$ exhibits a characteristic phase diagram via electron doping, as illustrated in Fig. 1. The diagram has two magnetic parent phases at $x \sim 0$ (PP1) and $x \sim 0.5$ (PP2), and two superconducting domes with $T_{c,\text{max}} = 26$ K at $x \sim 0.08$ (SC1) and $T_{c,\text{max}} = 36$ K at $x \sim 0.35$ (SC2)$^{15-18}$. Tetragonal-orthorhombic structural transitions at $T_{s1} (x \sim 0)$ and $T_{s2} (x \sim 0.5)$ precede magnetic transitions; the respective orthorhombic structures are centrosymmetric and non-centrosymmetric$^{18-20}$. The PP2 structure with the polar point group $mm2$ serves as a unique parent phase in high-$T_c$ materials as the spatial inversion symmetry is broken by temperature$^{18}$. Throughout this paper, we use the term "parent phase" to refer to the magnetic ordered state that breaks lattice $C_4$ symmetry. The source of SC2 has been discussed in terms of spin-fluctuation, orbital-fluctuation, and
orbital-selective Mott state models. However, the nematicity or fluctuating state in highly electron doped LaFeAsO$_{1-x}$H$_x$ remains unexplored and warrants investigation.

In this paper, we analyse the average and local structures of LaFeAsO$_{1-x}$H$_x$ ($0.35 \leq x \leq 0.51$) by measuring synchrotron X-ray diffraction (XRD) and extended X-ray absorption fine-structure (EXAFS). Our XRD and EXAFS results led us to identify a polar nematic state: a broken lattice $C_4$ symmetry and a polar fluctuated structure that emerge at temperatures far above the parent phase. These phenomena arise from the dynamical and short-range fluctuation of the parent phase structure.

**Results And Discussion**

We first focus on the change of crystal lattice symmetry in XRD measurements. Figure 2a shows the X-ray profiles for 220$_T$ reflection at 300, 120, and 32 K for $x = 0.51$, where the suffix "T" signifies the indexing in the tetragonal system. Though the 220$_T$ profile is supposed to split into two peaks below the tetragonal-orthorhombic structural transition at $T_{s2} \sim 95$ K, the profile was already broadened at 120 K. We regard the $T_{s2}$ transition as static and long-range order of the orthorhombic distortion, the temperature of which was estimated using the lattice constant and the resistivity anomalies. Moreover, the profile with a shoulder at 32 K exhibits inequivalent intensities of two reflections split from 220$_T$, manifesting the presence of polar structure below $T_{s2}$ (Supplementary Information, Fig. S1).

Figure 2b plots the temperature dependence of the profiles’ full width at half maximum $H$ for $x = 0.51$, 0.45, 0.42, 0.40, and 0.35. $H$ was obtained by fitting a profile to a single pseudo-Voigt function and normalising it to room temperature values. The $H$ values rise significantly when cooled past the $T_{s2}$ transition for $x = 0.51$, 0.45, and 0.42. Note that they gradually increase far above $T_{s2}$. In contrast, the $H$ values are nearly constant with temperature for $x = 0.40$ and 0.35. We here define the critical temperature $T^*$ as the point deviating from the baseline formed from the averaged values of $H$ above 250 K. The results are $T^* = 240$, 190, and 140 K for $x = 0.51$, 0.45, and 0.42, respectively. Since the $T_{s2}$ transition entails the broadening of the 220$_T$ peak, our result reveals a breaking of lattice $C_4$ symmetry for $x \geq 0.42$ despite a wide gap between $T^*$ and $T_{s2}$. Considering the XRD measurement timescale of $10^{-15}$ s, the lowering of lattice symmetry in $T_s < T < T^*$ might be viewed as a slower dynamical phenomenon. Another possible symmetry lowering for $x > 0$ above $T_{s1} = 175$ K has been suggested in the narrow temperature range of $175 K < T < 200 K$.

We proceed to examine the local structure of LaFeAsO$_{1-x}$H$_x$ from the As K-edge EXAFS measurement for $x = 0.51$, 0.45, 0.42, and 0.37. Figure 3a, b shows the representative $k^2$-weighted EXAFS oscillation $k^2\chi(k)$ and $R$-space magnitude of the Fourier transformation (FT), respectively, at 9.7 and 250 K for $x = 0.51$. In
the radial direction without phase-correction, the peak amplitudes around \( R = 2.1 \) and 2.9 Å correspond to As–Fe and As–La/As–O shells, respectively. We analysed the bond distances from the fit to the first As–Fe shell with an \( R \)-range of 1.85–2.30 Å based on the high-temperature \( P4/mmm \) structure. Figure 3c plots the temperature dependence of the As–Fe distances, which decrease on cooling within the high-temperature range (> 100 K). However, for \( x = 0.51, 0.45, \) and 0.42 the upturns in bond distances on cooling were observed at respective temperatures of 95, 90, and 30 K, which correspond closely to \( T_{s2} \). The elongation of bond distance below \( T_{s2} \) arises from the negative thermal expansion of the \( c \)-axis, as reported in previous XRD measurement\(^{18}\).

Next, we employed a Fourier-filtered back transformation in EXAFS, which enables the detection of unknown tiny distortions within a specific coordination sphere\(^{26,27}\). Figure 4 illustrates the Fourier-filtered EXAFS amplitudes for As–Fe (1.60–2.45 Å) shells of \( R \)-spectra (EXAFS oscillations in Supplementary Information, Fig. S2). This examination found inflection points or “kinks” in each of profiles at the low temperature side. The amplitudes at the lowest temperature and at 250 K are plotted in the figure insets with solid blue and dashed black lines, respectively. The data at 250 K is normalised by the peak position and height of the object profiles used for the baseline. We define the difference amplitude between the object profiles and the baseline as the phenomenological formula \( k^2 \chi_{\text{diff}}(q) = k^2 \chi(q)(T) - s_0 k^2 \chi(s_1 q)(250 \text{ K}) \), where \( s_0 \) and \( s_1 \) are the scale factors (Supplementary Information, Fig. S3). We evaluated the kink positions \( q_{\text{beat}} \) indicated by arrows as the peaks in the wavenumber derivatives of \( k^2 \chi_{\text{diff}}(q) \). The \( q_{\text{beat}} \) values were roughly 11.2, 11.0, 11.6, and 11.3 Å\(^{-1} \) respectively for \( x = 0.51, 0.45, 0.42, \) and 0.37 at the lowest temperature. The kink is due to the beat produced from the phase difference of EXAFS oscillations with the difference of bond distances \( \Delta R \) in the shell. Based on the relation \( \Delta R = \pi/(2q_{\text{beat}}) \), we can estimate \( \Delta R \) of the As–Fe distances to be 0.140, 0.143, 0.135, and 0.139 Å for \( x = 0.51, 0.45, 0.42, \) and 0.37, respectively.

The uniform As–Fe distance in the high-temperature phase splits into two long and two short distances in PP2 along with the loss of inversion symmetry\(^{18}\), leading to a small \( \Delta R \); this was not observed in PP1\(^{19,20}\). Note that \( \Delta R \) at 32 K for \( x = 0.51 \) in PP2 was previously determined to be 0.14 Å from XRD study\(^{18}\), which agrees with our EXAFS result. We therefore ascribe the kinks in the EXAFS amplitude to the presence of local polar structure, and regard the \( k^2 \chi_{\text{diff}}(q) \) as the fraction of the polar structure. The amplitude is largest at 30 K for \( x = 0.51 \) in PP2; its value drops with decreasing \( x \) and/or rising temperature, but can still be observed far outside of PP2.

Our results from XRD and EXAFS are summarised in Fig. 1 together with the previously observed phase diagram\(^{15,16,18}\). The green area signifies the local distortion with broken inversion symmetry. The phase diagram shows that the lowering of lattice symmetry appears below \( T^* \), and dynamical and short-range...
A polar structure emerges over wide ranges in temperature and doping. Taking account of the gradual changes in $H$ and beat amplitude, their transformations may be regarded as cross-over phenomena instead of a phase transition.

In the iron-based superconductors BaFe$_2$(As$_{1-x}$P$_x$)$_2$ and Sr$_{1-x}$Na$_x$Fe$_2$As$_2$, nematic states arise at specific doping levels from parent to superconducting phase, while breaking $C_4$ magnetic and/or lattice symmetries$^{28,29}$. Hence, we view the lowering of lattice symmetry at temperatures $T_{s2} \leq T \leq T^*$ revealed by our XRD measurements as a nematic state. In the same way, we suggest that our nematic state involves an electronic or magnetic nematicity. Moreover, the local polar structure was identified over a wider temperature and doping range. Since the polar structure below $T_{s2}$ entails the breaking of lattice $C_4$ symmetry, both of these phenomena that occur above $T_{s2}$ should be intertwined. We thus propose that the $T_{s2} \leq T \leq T^*$ region can be called a polar nematic state, although it is difficult to identify the vanishing temperature for beat amplitude. Sakurai et al. reported NMR measurements for LaFeAsO$_{1-x}$H$_x$ and pointed out an anisotropy of the electric-field gradient derived from a disproportionation of $d$-orbital electrons$^{30}$. This observation likely shares a close link with our result, given the possibility of the local polar structure being related to an orbital ordering or its fluctuation. Moreover, Onari et al. theoretically proposed that the second parent phase comes from a charge quadrupole order stemming from the disproportionation of $d$-orbital electrons$^{23}$.

Let us now consider the interplay between the local polar structure and the superconductivity. Lowering of lattice symmetry was unobserved via XRD for $x = 0.40$ and 0.35 in SC2, whereas the local polar structure was detected via EXAFS for $x = 0.37$. Since the orthorhombicity in PP2 rapidly reduces with decreasing $x$ from $x = 0.51^{18}$, the presence of a minute lattice distortion may have been experimentally undetectable via XRD. Regardless, the aforementioned electric-field gradient anisotropy from NMR$^{30}$ was evident even in the superconducting phase. Hence, we suggest that the polar nematic state is linked with the superconducting phase$^{30}$. We consider SC2 to be derived from PP2 by the introduction of holes, namely as $x$ decreases from ~ 0.5. Thus, in relation to superconductivity, the polar structure may have an effect on $T_c$ or the paring mechanism. As an example, noncentrosymmetric superconductors can give rise to exotic pairing states with spin-singlet and spin-triplet mixtures$^{1,31}$. In contrast, the superconductivity in LaFeAsO$_{1-x}$H$_x$ emerges after recovering inversion symmetry, where one might expect fluctuation-related phenomena instead. Intriguing theories have been proposed by Anderson and Blount$^6$, and Ydlium et al.$^{32}$ positing that ferroelectric-like soft phonons enhance $T_c$ or drives the superconductivity. Moreover, an odd-parity superconductivity derived from parity fluctuation has been predicted in the vicinity of inversion symmetry breaking$^{33,34}$. Since polar structure is also observed in SmFeAsO$_{1-x}$H$_x$ with $T_c = 55$ K$^{35,36}$, further insight awaits from the more detailed investigation of local physical properties in this system.

In conclusion, the average and local structures for highly electron doped LaFeAsO$_{1-x}$H$_x$ with bipartite parent phases were investigated using XRD and EXAFS measurements. The second parent phase ($x \sim 0.5$) entails the time-reversal and the spatial inversion symmetries broken. We have demonstrated that a
dynamical state with broken lattice $C_4$ symmetry and polar structure—a polar nematic state—emerges in a wide temperature/doping range above the parent and the superconducting phases. This observation reported in this study is the intriguing result because the electronic, magnetic, and lattice instabilities should be weak generally in the highly electron-doped region. We conclude that EXAFS serves as a good probe for the detection of local polar structure that could help map further studies of nematicity.

Methods

Powder samples of LaFeAsO$_{1-x}$H$_x$ were prepared using a high-pressure solid-state reaction as described in a previous study$^{15}$. Synchrotron XRD and As K-edge transmission EXAFS were carried out over the whole temperature range on beamlines BL-8A/8B/9C at the Photon Factory at the High Energy Accelerator Research Organization (KEK). For XRD measurements, a very fine powder sample was enclosed in a capillary with a diameter of 0.1 mm, which was irradiated with an X-ray beam. The sample was continuously rotated during the exposure. Two-dimensional XRD images were obtained using a diffractometer with a curved imaging plate (R-AXIS, Rigaku Corp.) at wavelength $\lambda = 1.0993$ Å. The images were integrated to yield $2\theta$-intensity data using the DISPLAY software (Rigaku). For EXAFS measurements, all crystalline reagents were fastidiously mixed with BN powder. This process is key to obtaining high-quality data up to the high-$k$ region. EXAFS oscillations and $R$-space magnitude of the Fourier transformation were extracted from the raw data using the Athena program. The $R$-space data were fitted to the theoretical signals calculated by FEFF8 code using IFEFIT on the Arthemis platform$^{37}$.

Declarations

Author Contributions

S.Maki, J.Y, S.I., Y.M, S. Matsuishi, H.H conceived the study. S.I. synthesised the samples. S.Maki, J.Y., H.A, H.S, R.K performed the synchrotron X-ray measurements. S.Maki, J.Y co-wrote the manuscript. All the authors discussed the results and the manuscript.

Acknowledgements

We thank Y. Kuramoto for fruitful discussions. This work was supported by the MEXT Elements Strategy Initiative to Form Core Research Center (JPMXP0112101001) and JSPS KAKENHI (No. 16K05434). The synchrotron XRD, EXAFS and neutron scattering experiments were performed with the approval of the Photon Factory Program Advisory Committee (No. 2013S2-002 and 2016S2-004) of IMSS, KEK. The crystal structures were created using the software VESTA $^{38}$.

References
1. Non-Centrosymmetric Superconductors-Introduction and Overview, edited by Bauer, E. & Sigrist, M. (Springer, Heidelberg, 2012).
2. Kim, T. H. et al. Polar metals by geometric design. Nature 533, 68–72 (2016).
3. Ideue, T. et al. Bulk rectification effect in a polar semiconductor. Nat. Phys. 13, 578–583 (2017).
4. Edelstein, V. M. Inverse Faraday Effect in Conducting Crystals Caused by a Broken Mirror Symmetry. Phys. Rev. Lett. 80, 5766–5769 (1998).
5. Benedek, N. A., Birol, T. 'Ferroelectric' metals reexamined: fundamental mechanisms and design considerations for new materials. J. Mater. Chem. C 4, 4000–4015 (2016),
6. Anderson, P. W., Blount, E. I. Symmetry Considerations on Martensitic Trans Formations: "Ferroelectric" Metals ? Phys. Rev. Lett. 14, 217 (1965).
7. Shi, Y. et al. A ferroelectric-like structural transition in a metal, Nat. Mat. 12, 1024–1027, (2013).
8. Volkov, P. A., Chandra, P. Multiband Quantum Criticality of Polar Metals. Phys. Rev. Lett. 124, 237601 (2020).
9. Rischau, C. W., Lin, X., Grams, C. P., Finck, D., Harms, S. A ferroelectric quantum phase transition inside the superconducting dome of Sr1 – xCaxTiO3–δ. Nat. Phys. 13, 643–648 (2017).
10. Kobayashi, T. C., Irie, Y., Yamaura, J., Hiroi, Z., Murata, K. Superconductivity of Heavy Carriers in the Pressure-Induced Phases of Cd2Re2O7. J. Phys. Soc. Jpn. 80, 023715 (2011).
11. Lee, A. P., Nagaosa, N., Wen, X.-G. Doping a Mott insulator: Physics of high-temperature superconductivity. Rev. Mod. Phys. 78, 17–85 (2006).
12. Paglione, J., Greene, R. L. High-temperature superconductivity in iron-based materials. Nat. Phys. 6, 645–658 (2010).
13. Hosono, H., Kuroki, K. Iron-based superconductors: Current status of materials and pairing mechanism. Physica C 514, 399–422 (2015).
14. Kamihara, Y., Watanabe, T., Hirano, M., Hosono, H. Iron-based layered superconductor La[O1 – xFx]FeAs (x = 0.05–0.12) with Tc = 26 K. J. Am. Chem. Soc. 130, 3296–3297 (2008).
15. Iimura, S. et al. Two-dome structure in electron-doped iron arsenide superconductors. Nat. Commun. 3, 943 (2012).
16. Iimura, S. et al. Heavy electron doping induced antiferromagnetic phase as the parent for the iron oxypnictide superconductor LaFeAsO1 – xHx. Phys. Rev. B 94, 024512 (2016).
17. Fujiwara, N. et al. Detection of antiferromagnetic ordering in heavily doped LaFeAsO1 – xHx pnictide superconductors using nuclear-magnetic-resonance techniques. Phys. Rev. Lett. 111, 097002 (2013).
18. Hiraishi, M. et al. Bipartite magnetic parent phases in the iron oxypnictide superconductor. Nat. Phys. 10, 300–303 (2014).
19. de la Cruz, C. et al. Magnetic order close to superconductivity in the iron-based layered LaO1 – xFxFxFeAs systems. Nature 453, 899–902 (2008).
20. Qureshi, N. et al. Crystal and magnetic structure of the oxypnictide superconductor \( \text{LaFeAsO}_1 - x\text{F}_x \): A neutron-diffraction study. \textit{Phys. Rev. B} \textbf{82}, 184521 (2010).

21. Suzuki, K. et al. Model of the Electronic Structure of Electron-Doped Iron-Based Superconductors: Evidence for Enhanced Spin Fluctuations by Diagonal Electron Hopping. \textit{Phys. Rev. Lett.} \textbf{113}, 027002 (2014).

22. Moon, C.-Y., Park, H., Haule, K., Shim, J. H. Origin of doping-induced suppression and reemergence of magnetism in \( \text{LaFeAsO}_1 - x\text{H}_x \). \textit{Phys. Rev. B} \textbf{94}, 224511 (2016).

23. Onari, S., Yamakawa, Y. Kontani, H. High-\( T_c \) Superconductivity near the Anion Height Instability in Fe-Based Superconductors: Analysis of \( \text{LaFeAsO}_1 - x\text{H}_x \). \textit{Phys. Rev. Lett.} \textbf{112}, 187001 (2014).

24. Kobayashi, K. et al. Pressure effect on iron-based superconductor \( \text{LaFeAsO}_1 - x\text{H}_x \): Peculiar response of 1111-type structure. \textit{Sci. Rep.} \textbf{6}, 39646 (2016).

25. McGuire, M. A. et al. Phase transitions in \( \text{LaFeAsO} \): Structural, magnetic, elastic, and transport properties, heat capacity and Mössbauer spectra. \textit{Phys. Rev. B} \textbf{78}, 094517 (2008).

26. Zhang, C. J., Oyanagi, H. Local lattice instability and superconductivity in \( \text{La}_{1.85}\text{Sr}_{0.15}\text{Cu}_{1-x}\text{M}_x\text{O}_4 \) \((\text{M} = \text{Mn}, \text{Ni}, \text{and Co})\). \textit{Phys. Rev. B} \textbf{79}, 064521 (2009).

27. Niemann, W. Multishell Beat-Node Method for EXAFS. \textit{Physica B} \textbf{158}, 279–281 (1989).

28. Kasahara, S. et al. Electronic nematicity above the structural and superconducting transition in \( \text{BaFe}_2(\text{As}_1 - x\text{P}_x)_2 \). \textit{Nature} \textbf{486}, 382–385 (2012).

29. Frandsen, B. A. et al. Widespread orthorhombic fluctuations in the (\text{Sr,Na})\text{Fe}_2\text{As}_2 family of superconductors. \textit{Phys. Rev. B} \textbf{98}, 180505(R) (2018).

30. Sakurai, R. et al. Quantum critical behavior in heavily doped \( \text{LaFeAsO}_1 - x\text{H}_x \) pnictide superconductors analyzed using nuclear magnetic resonance. \textit{Phys. Rev. B} \textbf{91}, 064509 (2015).

31. Bauer, E. et al. Heavy Fermion Superconductivity and Magnetic Order in Noncentrosymmetric \( \text{CePt}_3\text{Si} \). \textit{Phys. Rev. Lett.} \textbf{92}, 027003 (2004).

32. Yildirim, T. Ferroelectric soft phonons, charge density wave instability, and strong electron-phonon coupling in \( \text{BiS}_2 \) layered superconductors: A first-principles study. \textit{Phys. Rev. B} \textbf{87}, 020506(R) (2013)

33. Kozii, V., Fu, L. Odd-Parity Superconductivity in the Vicinity of Inversion Symmetry Breaking in Spin-Orbit-Coupled Systems. \textit{Phys. Rev. Lett.} \textbf{115}, 207002 (2015).

34. Wang, Y., Cho, G. Y., Hughes, T. L., Fradkin, E. Topological superconducting phases from inversion symmetry breaking order in spin-orbit-coupled systems. \textit{Phys. Rev. B} \textbf{93}, 134512 (2016).

35. Matsuishi, S. et al. Controlling factors of \( T_c \) dome structure in 1111-type iron arsenide superconductors. \textit{Phys. Rev. B} \textbf{89}, 094510 (2014).

36. limura, S. et al. Large-moment antiferromagnetic order in overdoped high-\( T_c \) superconductor \( ^{154}\text{SmFeAsO}_1 - x\text{D}_x \). \textit{Proc. Natl. Acad. Sci. USA} \textbf{114}, E4354–E4359 (2017).

37. Ravel, B., Newville, M. ATHENA, ARTEMIS, HEPHAESTUS: data analysis for X-ray absorption spectroscopy using IFEFFIT. \textit{J. Synchrotron Rad.} \textbf{12}, 537–541 (2005).
38. Momma, K., Izumi, F. VESTA†3 for three-dimensional visualization of crystal, volumetric and morphology data. *J. Appl. Crystallogr.* **44**, 1272–1276 (2011).