1. Introduction

The LaFe$_{13-x}$Si$_x$ system has been widely investigated in recent years because of the giant magnetocaloric effect (GME) that was observed for compositions $1.0 \leq x \leq 1.8$ [1, 2]. A potential application of materials exhibiting a GME is magnetic refrigeration—a technology that has the prospect of replacing conventional gas compressor refrigerators due to higher efficiency and environmental sustainability [3–6].

In our contribution, we present details on the mechanism of the phase transition in the LaFe$_{13-x}$Si$_x$ system by investigating the magnetostructural coupling and internal pressure related to spin fluctuations in LaFe$_{11.6}$Si$_{1.4}$. LaFe$_{13-x}$Si$_x$ is a ferromagnetic material with a composition-dependent Curie temperature $T_C$. It is described as an itinerant-electron system in which the magnetic transition from the paramagnetic (PM) to a ferromagnetic (FM) state can be induced by either temperature or magnetic field, if applied just above $T_C$ [1, 7, 8]. The magnetic transition is of first-order for compositions $x \leq 1.6$ [9] and, if induced by a magnetic field, is referred to as an itinerant-electron metamagnetic (IEM) transition [1, 8]. The change in internal energy during the IEM transition results in the GME and is dominated by changes in the electronic structure [10].

LaFe$_{11.6}$Si$_{1.4}$ exhibits a first-order PM–FM transition at $T_C \approx 190$ K that is accompanied by a giant spontaneous...
magnetostriction of ~1.2% due to magnetostriction coupling [9, 11, 12]. It crystallizes in the cubic NaZn13-type structure (Fm3c #226) in which the Fe atoms are located on two inequivalent Wyckoff sites (8b and 96i) with the La and Si atoms occupying the 8a and 96i sites, respectively [13]. The PM and FM phases are isomorphous, apart from the difference in unit cell volume. The magnetically ordered phase has a simple FM spin structure and therefore no magnetic superlattice reflections appear in neutron diffraction patterns of the FM phase [12].

The paramagnetic state of LaFe13−xSi_x was recently described with a disordered local moment (DLM) model with a fluctuating Fe moment of ~1.9 \( \mu_B \) [10, 14]. In accordance with this model, the temperature dependence of the reciprocal paramagnetic susceptibility \( \chi^{-1} \) was found to obey the Curie–Weiss (CW) law for compositions \( x > 1.6 \) [9, 15–17]. Compositions of LaFe13−xSi_x with \( x \leq 1.6 \) also show CW behavior in the high-temperature regime, but in addition they exhibit a paramagnetic Curie temperature \( \theta_p \), as defined by the Bean–Rodbell model of magnetoelastic coupling [9, 18]. \( \theta_p < T_C \) is characteristic for first-order transitions, and the larger \( T_C - \theta_p \) the stronger the first-order character of the transition. Spin fluctuations terminate the FM state for first-order transitions before magnetic excitations realize the disappearance of collective magnetization and PM and FM phase therefore have different Curie temperatures [17]. For second-order transitions \( \theta_p = T_C \) and no deviation from CW behavior is observed [9].

The two main contributions to spin fluctuations are thermally excited spin fluctuations and zero-point fluctuations. On cooling, one would expect the amplitude of thermal spin fluctuations \( \xi_{th} \) to decrease, whereas the amplitude of zero-point fluctuations \( \xi_{zp} \) increases, resulting in a net conservation of the squared sum \( \xi_{th}^2 + \xi_{zp}^2 = const \) [19, 20].

Spin fluctuations as a driving force for magnetoelastic phase transitions have recently been discussed for other GME materials, such as rare earth manganites [21, 22], the (Mn,Fe)2(P,Si) system [23, 24] and Heusler compounds [25–27].

First-order phase transitions are generally described in terms of their Landau free energy. Spin fluctuations in itinerant-electron systems require a renormalization of the expansion coefficients in a Landau theory [7, 28] and had been introduced in the self-consistent renormalization (SCR) spin fluctuation theory by Moriya and Kawabata [20, 29]. The effect of spin fluctuations can be interpreted as an internal magnetic pressure that modifies the thermal expansion behavior [28, 30]. A deviation from linear thermal expansion can therefore be seen as evidence for the presence of strong spin fluctuations as Wada et al have demonstrated for Y(Mn1−xAl_x)2 and Y1−xSc_xMn_x2 [31].

Takahashi proposed a theory describing the effect of spin fluctuations on the magnetic properties of itinerant-electron systems [20, 32]. According to Takahashi’s theory, the magnetic pressure of spin fluctuations expresses itself in a fourth power of magnetization \( M^4 \) dependence of the forced magnetostriction \( \Delta L/L \) at \( T_C \) as seen in the following equation:

\[
\frac{\Delta L}{L} = \frac{v_0 (M, T_C)}{v_0} = C \cdot \xi_{th} (0, T_C) \cdot \frac{M^4}{M_0^4(0)}
\]  

where \( v_0 \) and \( v_0 (M, T_C) \) represent the spontaneous and magnetic field-dependent volume contribution to magnetostriction, respectively. \( C \) is constant under isothermal conditions and \( \xi_{th}(0, T_C) \) is the amplitude of thermal spin fluctuations in zero field. \( M(0) \) represents the spontaneous magnetization and \( M \) the magnetization in a magnetic field.

The effect of spin fluctuations can not only be inferred indirectly in macroscopic variables like the volume or the magnetization: the direct proof for the occurrence of fluctuations is their detection by scattering methods.

Neutron scattering is a universal tool to investigate spin structures, both localized in a magnetically ordered system and disordered in the form of spin waves. The intensity of magnetically scattered neutrons in general is defined by the following equation (2): [33]

\[
I_{mag}(Q) \sim |f_{mag}(Q)|^2 \sum_{\alpha,\beta} S^\alpha(\omega)(\delta_{\alpha,\beta} - Q_\alpha Q_\beta)
\]

where \( f_{mag}(Q) \) is the magnetic form factor, \( Q \) is the momentum transfer, \( \omega \) is the energy transfer and the summation runs over the Cartesian directions. \( S^\alpha(\omega) \) is the magnetic scattering function which is proportional to the space and time Fourier transform of the spin–spin correlation function. The term \( \delta_{\alpha,\beta} - Q_\alpha Q_\beta \) describes that neutron only probe the components of spin perpendicular to \( Q \). If the energy of scattered neutrons is not analyzed, \( I_{mag}(Q) \) is a snapshot of the spin correlations in reciprocal space.

In this contribution, we provide direct experimental evidence for the important role of itinerant spin fluctuations in the magnetic phase transition of LaFe11.6Si1.4. We report on evidence for paramagnetic spin fluctuations in LaFe11.6Si1.4 observed in the form of magnetic diffuse scattering in our temperature-dependent neutron powder diffraction data. Furthermore, we use x-ray diffraction to detect the effect of spin fluctuations on the lattice parameter, thereby verifying the ‘internal pressure’ effect of the fluctuations. Lastly, we confirm that the short-range magnetic correlations in LaFe11.6Si1.4 follow Takahashi’s theory of spin fluctuations in itinerant-electron systems by using the example of the magnetization dependence of critical forced magnetostriction.

2. Experimental

2.1. Sample preparation

LaFe11.6Si1.4 was synthesized from elemental materials with commercial purity in a Al2O3 crucible using an induction furnace, as described in [34]. To prevent oxygen contamination, the elemental lanthanum was arc melted prior to induction melting with the other elements. To compensate for lanthanum losses due to evaporation, an excess of 7% La was added. For better homogeneity, the sample was re-melted twice in an induction oven under Ar pressure of 1 bar. Afterwards the
The magnetic moment measurements were performed using the vibrating sample magnetometer option of a QD PPMS 14 (Quantum Design Physical Property Measurement System, LOT-QuantumDesign GmbH). A needle shaped sample, cut from the bulk specimen, was used in order to minimize the demagnetization factor due to shape anisotropy. The PM–FM transition temperature $T_C$ was determined from the temperature dependent magnetization measured in a magnetic field of 500 Oe from 300 to 100 K with a temperature sweep rate of 1 K min$^{-1}$.

The ac magnetic susceptibility was measured by means of an ACMS option of a QD PPMS 14 in an applied magnetic field of 10 kOe. The temperature dependent measurement was performed on cooling from 350 to 100 K in an alternating excitation field with frequency of 1 kHz and amplitude of 10 Oe.

2.3. Neutron diffraction

Unpolarized neutron diffraction experiments were performed at the high-resolution diffractometer SPODI at research reactor FRM II, Garching [35]. The wavelength was set to $\lambda = 1.5482$ Å by a stack of Ge(5 5 1) monochromator crystals. A cylinder ($11 \times 18$ mm, $\sim$11 g) cut from polycrystalline bulk LaFe$_{11.6}$Si$_{1.4}$ was placed in a cylindrical vanadium container and into a closed-cycle helium cryostat. Diffraction patterns were collected on cooling between 295 K and 5 K. Phase purity is confirmed aside from $\sim$1 wt.$\%$ fraction of $\alpha$-Fe side phase. The small goodness-of-fit ($\chi^2$) and residual (R) values from Rietveld refinement of profile ($R_\text{p}$), peak positions ($R_\text{Bragg}$) and below ($T_C$) magnetic structure ($R_\text{mag}$) suggest an excellent data quality and a well-fitting structure model. The temperature of the onset of magnetic transition $T_C$ = 200 K is identified by a splitting of all Bragg reflections, see figure 1(c). PM and FM phase coexist in the temperature range from 200 to 191 K and the magnetic transition is complete at the Curie temperature $T_C$ = 190 K.

First traces of the FM phase appear in the neutron diffraction pattern at $T_C$, identified by a splitting and shift of all reflections to lower $Q$ due to giant spontaneous magnetostriction, see figure 1(c). The observed coexistence of PM and FM phase is characteristic for a nucleation and growth process of first-order phase transitions, as it is expected for LaFe$_{11.6}$Si$_{1.4}$ [9, 12]. The lattice parameter of the FM phase $a$ (FM) is significantly larger than that of the PM phase $a$ (PM), see figure 2 and it further expands on cooling in the two-phase region, due to the increasing spontaneous Fe magnetic moment [12].

Apart from $\sim$1% volume expansion, the crystal structure of the PM phase of LaFe$_{11.6}$Si$_{1.4}$ is retained in the FM phase. Figure 3(a) shows a contour plot of the temperature dependence of the low $Q$ region from 295 to 5 K. The neutron diffraction patterns of the PM and FM phases display the same number of reflections, however, several Bragg reflections such as 2 0 0 and 2 2 0 gain intensity during the magnetic transition.

Strong diffuse intensity $I_{\text{diff}}$ is present above $T_C$ in the low $Q$ range up to $Q \approx 0.8$ Å$^{-1}$, see figure 3(b). On cooling from 295 K, $I_{\text{diff}}$ first increases and peaks at $T = 200 K = T_C$ but sharply drops thereafter. Simultaneously to the decrease in $I_{\text{diff}}$, the 200 and 220 Bragg reflections gain intensity due to FM contributions. The integrated intensity of Bragg reflections $I_{\text{Bragg}}(hkl)$ in neutron diffraction consists of two contributions: $I_{\text{Bragg}}(hkl) = I_{\text{mag}}(hkl) + I_{\text{nuc}}(hkl)$, where $I_{\text{mag}}(hkl)$ is the nuclear and $I_{\text{mag}}(hkl)$ the magnetic contribution due to long-range spin ordering. The close relation between $I_{\text{diff}}$ and $I_{\text{mag}}(hkl)$ is a hint towards a magnetic origin of $I_{\text{diff}}$, such as short-range FM correlations. Neutron diffraction studies published previously on the LaFe$_{13-\delta}$Si$_{1}$ system, however, do not
Figure 1. Rietveld refinement of the neutron diffraction pattern of LaFe$_{11.6}$Si$_{1.4}$ collected at (a) $T = 295$ K and (b) $T = 5$ K. Observed (red dots), calculated (black line) and difference (grey line) patterns are given, as well as reflection positions for LaFe$_{11.6}$Si$_{1.4}$ (blue), magnetic LaFe$_{11.6}$Si$_{1.4}$ (dark blue) and a side phase of $\sim$1 wt.% $\alpha$-Fe (wine). Structural parameters from the Rietveld refinement are listed in supplementary table S1 (available online at stacks.iop.org/JPhysCM/32/115802/mmedia). (c) Splitting and shift of high $Q$ nuclear reflection 10 8 6 of LaFe$_{11.6}$Si$_{1.4}$ on cooling over the magnetic ordering temperature. The high-temperature paramagnetic HT-PM and low-temperature ferromagnetic LT-FM phase coexist in the temperature range from 200 to 191 K.

Figure 2. Phase fraction (left) and lattice parameter (right) of the paramagnetic PM (filled circles) and ferromagnetic FM phase (open circles) obtained from Rietveld refinements of neutron diffraction data of bulk LaFe$_{11.6}$Si$_{1.4}$. The dashed line marks the onset of magnetic transition at temperature $T_{tr} = 200$ K. The dotted line shows the Curie temperature $T_C = 190$ K. Symbols are connected by spline curves serving as guides to the eye. The inset shows the crystal structure of LaFe$_{11.6}$Si$_{1.4}$ with two distinct Fe atoms (wine), Si (orange) partially occupying the Fe position on polyhedral corners and La (purple) occupying large voids in between Fe/Si polyhedra [39].
Figure 3. (a) Contour plot of neutron diffraction data measured for bulk LaFe$_{11.6}$Si$_{1.4}$ on cooling in the temperature range 295 to 5 K. Small angle diffuse scattering (top-left) and Bragg reflections 200 and 220 are marked. (b) Raw neutron diffraction patterns collected at 295, 200 and 5 K.

Figure 4. Evaluation of the temperature dependence of magnetic scattering intensity from neutron diffraction data of bulk LaFe$_{11.6}$Si$_{1.4}$. Integrated magnetic diffuse scattering $I_{\text{diff}}$ (left) and normalized 200 magnetic Bragg reflection intensity $I_{\text{mag}}(200)$ (right) are shown. The dashed line marks the onset of magnetic transition at temperature $T_u = 200$ K. The dotted line shows the Curie temperature $T_C = 190$ K. Symbols are connected by spline curves serving as guides to the eye.
show the low $Q$ region and thus do not allow for comparison with our work [12, 13, 40].

We integrate the area under the curve like $I_{\text{diff}} = \int_{0.8}^{1.0} I(Q) dQ$ in order to quantify the diffuse scattering contribution, see figure 4. Sizable $I_{\text{diff}}$ is already present at 295 K (1.55 $T_C$). Upon cooling, $I_{\text{diff}}$ increases approaching $T_C$ and has a maximum at $T_{tr} = 200$ K (1.05 $T_C$). $I_{\text{diff}}$ sharply drops with the appearance of FM phase in the temperature range between $T_u$ and $T_C$ and is converted into $I_{\text{mag}}(hkl)$. At $T = 186$ K (0.98 $T_C$), however, despite the magnetic transition being complete, $I_{\text{diff}}$ is still ~33 % of the value at $T_{tr}$, as can be seen in figure 4. A further conversion of the remaining $I_{\text{diff}}$ to $I_{\text{mag}}(hkl)$ continues down to 5 K. An explanation for significant $I_{\text{diff}}$ being present below $T_C$ is that the magnetization is not yet saturated. Consequentially, the conversion of remaining $I_{\text{diff}}$ to $I_{\text{mag}}(hkl)$ continues in the FM phase down to 5 K. The further increase in $I_{\text{diff}}$ far below $T_C$ indicates that the localized Fe magnetic moment increases up to its saturation value of 2.16 $\mu_B$ at 0 K [12].

3.2. Magnetization and ac magnetic susceptibility

As shown in figure 5, the LaFe$_{11.6}$Si$_{1.4}$ compound shows a sharp PM–FM transition in the dc magnetization measurements with a small thermal hysteresis of ~1 K. We can conclude from the sharp transition that the sample is chemically homogeneous. The small remnant magnetization above $T_C$ proves that only low amounts of FM impurities such as $\alpha$-Fe are present. $T_C = 191$ K is in perfect agreement with $T_C$ obtained from neutron diffraction.

The spontaneous magnetization amounts to 22.62 $\mu_B$/f.u. at 5 K; consequently one can extract an average value for the mean Fe magnetic moment. The average moment per Fe atom ($\mu_{Fe}$) is estimated to be 1.95 $\mu_B$/Fe. Magnetic measurements have been recorded at high temperatures, well above the magnetic transition temperature, in order to investigate the magnetic behavior of Fe in the paramagnetic state. At high temperatures, the thermal variation of the reciprocal magnetic susceptibility shows a CW behavior. A linear fit of the experimental data according to a CW law leads to a Curie constant $C = 67.9$ $\mu_B$/K/f.u.T. The effective paramagnetic moment $\mu_{\text{eff}}$ deduced from the Curie constant is found to be 5.10 $\mu_B$/Fe.

The investigation of the magnetic properties in both paramagnetic and ferromagnetic states has led to significantly different values of Fe magnetic moments in magnetically ordered state ($\mu_{Fe} = 1.95$ $\mu_B$/Fe) and in the disordered state ($\mu_{\text{eff}} = 5.10$ $\mu_B$/Fe). This leads to a number of spin $S_0 = 2.10$ and $S_0 = 0.975$ in the paramagnetic and the ferromagnetic state respectively and a corresponding Rhodes–Wohlfarth ratio $r = S_0/S_0$ of 2.15. This points to the itinerant character of magnetism in the present compound.

Figure 5. Temperature dependence of the magnetization $M$ (left) of bulk LaFe$_{11.6}$Si$_{1.4}$ under an applied field $\mu_0H = 0.05$ T and the inverse of the real part of the ac susceptibility $\chi^{-1}$ (right) measured at 1 T. The dashed line shows a linear fit of $\chi^{-1}$ in the paramagnetic regime according to Curie–Weiss law.
Since the obtained value of $r$ is larger than 1, we expect that the amplitude of local spin fluctuations varies significantly with temperature in this system, according to the SCR theory of spin fluctuations [41–43]. In this model, two extreme regimes characterized by different values of the Rhodes–Wohlfarth ratio $r$, are described: (i) local moment limit and (ii) weakly ferromagnetic limit. In the local moment limit we have $r = 1$ and in the opposite weakly ferromagnetic limit the theoretical model predicts a divergence of this ratio.

The inverse of the real part of the ac susceptibility $\chi'^{-1}$ shows CW behavior in the PM regime as predicted for itinerant ferromagnets by Moriya and Kawabata [29] and for the LaFe$_{13-x}$Si$_x$ system in particular by Fujita [14], but deviates from linearity at $T_{CW} = 225$ K. This change of slope in $\chi'^{-1}$ close to $T_C$ is expected for first-order transitions according to the Bean–Rodbell model [18]. We found the thermal expansion to be linear for $T > 300$ K with an expansion coefficient $\alpha = 1.1 \times 10^{-5}$ K$^{-1}$, independent of magnetic field. Jia et al reported a value of $\alpha = 8.2 \times 10^{-6}$ K$^{-1}$ [9] for the temperature range 300–250 K. Our data show, however, that the thermal expansion is not linear in the temperature range used for determination of the reported $\alpha$. Deviation from linearity starts at $T \approx 300$ K, most likely due to the appearance of short-range magnetic correlations.

3.3. Magnetic field and temperature-dependent x-ray diffraction

Figure 6 shows the thermal evolution of the lattice parameter of LaFe$_{11.6}$Si$_{1.4}$ determined from x-ray powder diffraction on cooling for different magnetic fields. Extracting separate lattice parameters $a(\text{PM})$ and $a(\text{FM})$ in the transition region was not possible because of the lower angular resolution of our x-ray compared to our neutron diffraction experiments. The PM–FM transition of LaFe$_{11.6}$Si$_{1.4}$ powder is not as sharp as for bulk material, which is a known effect for a powder due to decoupling of the particles [44]. We extracted a paramagnetic Curie temperature $\theta_p = 170$ K that is close to the value reported for LaFe$_{11.8}$Si$_{1.2}$ [9]. The large difference $T_C - \theta_p$ confirms the strong first-order nature of the phase transition.

Figure 6. Lattice parameter of LaFe$_{11.6}$Si$_{1.4}$ powder determined by x-ray diffraction for ZFC and FCC protocol in magnetic fields of 1, 3 and 5 T. The dotted line shows the extrapolation of linear thermal contraction above $T_C$. 

Applying a magnetic field during cooling shifts $T_C$ towards higher temperatures at a rate of $\sim 4$ K T$^{-1}$, which is consistent with previous studies [1, 9]. The magnetic transition is broader for larger magnetic fields due to an increasing second-order character of the IEM [1]. Simultaneously with $T_C$, $T_{comp}$ also increases with increasing magnetic field, whereas the high-temperature behavior above $T > 300$ K is identical for all
magnetic fields. We assume that the magnetic field enhances the short-range magnetic correlations in the PM regime at $T < 300$ K of LaFe$_{11.6}$Si$_{1.4}$. Therefore, $T_{\text{comp}}$ increases with increasing field—eventually inducing the IEM transition at $T > T_C(0 \text{ T})$. Below $T_C$ we observe a small increase in $a(\text{FM})$ with increasing magnetic field due to forced magnetostriction and saturation of magnetization far above 0 K.

4. Discussion

4.1. Diffuse Scattering

We integrate the diffuse signal shown in figure 3(b) up to $Q = 0.8$ Å$^{-1}$, which is justified by the fact that the maximum momentum transfer of spin fluctuations in Fe is $Q_{\text{max}} \approx 0.75$ Å$^{-1}$ [45]. Spin fluctuations transition into the Stoner continuum at larger $Q$ values and our $I_{\text{diff}}$, therefore, covers all essential magnetic fluctuations. We consider the fluctuation-dissipation theorem [46] to show that $I_{\text{diff}}$ in our neutron diffraction data is indeed a good proxy for $\xi$. However, the maximum of the as-observed $I_{\text{diff}}$ at $T_u$ does not necessarily translate to a maximum of $\xi$. The reason being that the PM–FM transformation begins at $T_u$ and spans over a 10 K temperature window. $I_{\text{diff}}$ decreases proportionally to the PM fraction ($W_{\text{PM}}$) between $T_u$ and $T_C$ whereas $I_{\text{mag}}(hkl)$ increases proportionally to the FM fraction ($W_{\text{FM}}$). If we assume that $I_{\text{diff}}$ originates only from the PM phase, we can correct $I_{\text{diff}}$ above $T_C$ by dividing through $W_{\text{PM}}$.

Figure 7 shows the comparison of $I_{\text{diff,corr}} = I_{\text{diff}}/W_{\text{PM}}$ with the real part of the ac susceptibility $\chi'$; the fact that they both show the same temperature dependence is evidence that $I_{\text{diff,corr}}$ measures $\xi$. We can now use $I_{\text{diff,corr}}$ instead of equation (2), while the error that we make by final integration in space and time is small, since $f_{\text{mag}}(Q)$ is a function that decays fast. $\xi$, consequently, increases continuously down to $T_C$.

4.2. Lattice parameter—internal magnetic pressure

We observe linear thermal contraction for LaFe$_{11.6}$Si$_{1.4}$ at temperatures above $T > 300$ K. Deviation from linear behavior starts below $T \approx 300$ K and is present in both, our neutron and x-ray diffraction data, see figures 2 and 6, respectively. Simultaneously, our neutron diffraction data reveals that strong magnetic fluctuations are present in the form of $I_{\text{diff}}$ at $T = 295$ K, see figure 3. $I_{\text{diff}}$, as shown in section 4.1, is a measure for $\xi$ and increases on cooling, until it diverges at $T_C$. At the same time, the lattice parameter deviates further from high-temperature linear behavior the closer the temperature gets to $T_C$. Below $T_{\text{comp}} = 225$ K, the lattice parameter even increases upon further cooling. A feature that is likely related is present in our ac susceptibility data, where we observe a deviation from CW behavior at the same temperature $T_{\text{CW}} = 225$ K, see figure 5.

In the two-phase region we observe a sharp increase in lattice parameter not only for the FM phase, but also for the PM phase. $a(\text{FM})$ is expected to increase due to spontaneous magnetostriction as the Fe magnetic moments assume long-range order [12]. The trend of $a(\text{PM})$, however, cannot be explained by magnetostriction. Instead, we use the model of internal magnetic pressure by Wagner and Wohlfarth. According to their theory, spin fluctuations exert a magnetic pressure proportional to $\xi$ and thus influence a system’s lattice parameter.

Figure 7. Comparison of the real part of the ac magnetic susceptibility $\chi'$ under $\mu_0H = 1$ T (black) and $I_{\text{diff,corr}}$ (green) normalized to 1. The dashed line highlights the temperature of onset of magnetic transition $T_u = 200$ K.
Since $\xi$ increases, we expect an increasing magnetic pressure on cooling and a larger effect on $\alpha$(PM) closer to $T_C$. From our data we find that both $\xi$ and $\alpha$(PM) increase upon cooling and sharply close to $T_C$, verifying the theory of internal magnetic pressure. The deviation from linear thermal contraction of LaFe$_{11.6}$Si$_{1.4}$ can therefore be seen as another measure of $\xi$.

### 4.3. Correlation between magnetization and critical forced magnetostriction

In order to probe the magnetization dependence of critical forced magnetostriction according to equation (1), we extracted lattice parameters of LaFe$_{11.6}$Si$_{1.4}$ powder from Rietveld refinements of our magnetic field and temperature-dependent x-ray diffraction measurements. We derived the forced magnetostriction $\Delta L/L$ from the relative change in lattice parameter in magnetic field compared to zero-field data. The resulting $\Delta L/L$ is shown as a function of $M^2$ and $M^4$ in figures 8(a) and (b), respectively. Magnetization measurements performed separately for LaFe$_{11.6}$Si$_{1.4}$ powder confirm a broadening of the transition and a slightly larger $T_C = 194$ K, which is still very close to the bulk value.

Classical SEW theory of magnetovolume coupling (neglecting fluctuations) suggests that $\Delta L/L$ versus $M^2$ follows a straight line through the origin at $T_C$ [47]. The forced magnetostriction of LaFe$_{11.6}$Si$_{1.4}$ indeed follows a straight line versus $M^2$ for all temperatures close to $T_C$, see figure 8(a), however, for none through the origin. This observation is in accordance with Takahashi’s SCR spin fluctuation theory. He suggests a change from $M^2$-linearity behavior to $M^4$-linearity behavior at $T_C$, as shown in equation (1) [20]. The plot of $\Delta L/L$ versus $M^4$, see figure 8(b), indeed indicates linearity through the origin at $T_C$.

### 5. Conclusions

Neutron and x-ray diffraction studies on LaFe$_{11.6}$Si$_{1.4}$ reveal that short-range magnetic correlations in the paramagnetic regime drive the first-order PM–FM transition. These spin fluctuations are observable as neutron diffuse scattering and exist as far as 100 K above the Curie temperature $T_C = 190$ K. On cooling, the magnetic diffuse intensity $I_{\text{diff}}$ and ac magnetic susceptibility $\chi$ show the same temperature dependence. $I_{\text{diff}}$ is, therefore, directly related to the amplitude of short-range magnetic correlations $\xi$. $\xi$ increases as $T_C$ is approached, creating an internal magnetic pressure that leads to a deviation from linear thermal contraction and an overcompensation close to $T_C$. Both the PM and FM phase coexist in the temperature range between $T_{\text{tr}} = 200$ K and $T_C$, in which $I_{\text{diff}}$ in the neutron diffraction patterns is transferred to magnetic Bragg intensity $I_{\text{mag}}$. The lattice parameter of PM phase increases sharply in the two-phase region, simultaneously to a large increase in $\xi$—verifying the pressure effect created by the spin fluctuations. The critical forced magnetostriction at $T_C$ is proportional to the fourth power of magnetization, which is in accordance with Takahashi’s SCR spin fluctuation theory.

Paramagnetic spin fluctuations might in general play an important role in driving the magnetocaloric effect in LaFe$_{13-x}$Si$_x$. We expect that the fluctuations are increasingly suppressed with larger Si concentrations $x$, especially for compositions $x > 1.6$, which have a second-order transition. However, the evolution of spin fluctuations in the LaFe$_{13-x}$Si$_x$...
system as function of $x$ and, consequentially, the extension to the commercially applied La(Fe,Mn)$_{13-x}$Si$_x$H system would be a topic for further investigation. It could provide a generalized view on the occurrence of the GME.

Acknowledgments

We would like to thank LVB Diop for performing the magnetic calculations and helpful discussions regarding the manuscript and DFG instrument grant INST 163/442-1 FUGG and LOEWE RESPONSE for funding.

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