Magnetic ordering in non-centrosymmetric CePdAl$_3$ and CePtAl$_3$

M. Stekiel,$^1$ P. Čermák,$^{2,3}$ M. Meven,$^{2,4}$ C. Franz,$^{2,5}$ S. Weber,$^1$ R. Schönmann,$^1$
V. Kumar,$^1$ K. Nemkovskiy,$^2$ H. Deng,$^{2,4}$ A. Bauer,$^1$ C. Pfleiderer,$^1$ and A. Schneidewind$^2$

$^1$Physik-Department, Technische Universität München, D-85748 Garching, Germany
$^2$JCMS at MLZ, FZ Jülich GmbH, Lichtenbergstr. 1, D-85747 Garching
$^3$Charles University, Faculty of Mathematics and Physics, Department of Condensed Matter Physics, Ke Karlovu 5, 121 16, Praha, Czech Republic
$^4$Institut für Kristallographie at MLZ, RWTH Aachen, Lichtenbergstrasse 1, D-85747 Garching
$^5$TUM at MLZ, Technische Universität München, D-85748 Garching, Germany

(Dated: June 16, 2021)

Owing to the hybridization of cerium’s localised 4$f$ electron and conduction band composed of $d$-electrons, cerium based intermetallics exhibit various kinds of magnetic interactions. In crystals, these can result in exotic types of magnetic ordering. In this study, we report a detailed single-crystal neutron diffraction study on CePdAl$_3$ and CePtAl$_3$. We have synthesized a large crystal of CePdAl$_3$, which crystallizes in a non-centrosymmetric, orthorhombic structure with space group $Cmc2_1$, a new, distorted variant of the tetragonal BaNiSn$_3$ structure observed in other CeTaAl$_3$ compounds, such as CePtAl$_3$. Low-temperature diffraction measurements showed that CePdAl$_3$ orders in a collinear antiferromagnetic structure below $T_N = 5.3(1)$ K, with magnetic moments pointing along the $a$-axis direction and an ordered magnetic moment $\mu = 1.64(3) \mu_B$/Ce$^{3+}$. Tetragonal CePtAl$_3$ shows a modulated, cycloidal type of ordering with $k = (\frac{1}{2} 0 0)$, and a transition temperature $T_N = 3.2$ K. Symmetry analysis allows two types of ordering, which show modulation of both amplitude and direction of magnetic moments. These results allow to conclude that in CeTaAl$_3$ system the orthorhombic distortion ($T$ = Pd, Ag) releases some underlying magnetic frustration that results in modulated types of magnetic ordering in tetragonal compounds ($T$ = Cu, Au, Pt).

I. INTRODUCTION

Cerium based intermetallics comprise of wide range of systems exhibiting variety of interesting physical phenomena owing to the competition or coupling between the electronic, magnetic and structural degrees of freedom. One of the most renowned is CePt$_3$Si$_3$ [1], the only non-centrosymmetric compound in which heavy-fermion superconductivity is discovered at ambient conditions. Superconductivity occurs in several related systems applying pressure: CeRhSi$_3$ [2], CeIrSi$_3$ [3], and CeCoGe$_3$ [4, 5]. They all exhibit antiferromagnetic ordering before entering the superconducting state upon decreasing temperature, with varying levels of complexity which arise from the competition between Ruderman-Kittel-Katsuya-Yoshida (RKKY) interaction, Kondo screening and crystal electric field (CEF) effects [6, 7]. These discoveries provide a motivation for a systematic characterisation of electronic and magnetic properties and their connection to the crystal structure.

In this study we focus on the CeTaAl$_3$ family of compounds, which up to date comprises of $T$ = Cu, Au, Ag, Pt, Pd. Although none of them was found to be superconducting so far, a CEF-phonon hybridization was reported in CeCuAl$_3$ [8] and CeAuAl$_3$ [9], whose nature and detailed properties remain unclear. The so-called vibronic bound state (VBS) arises from interactions between phonons and transitions of the cerium’s unpaired 4$f$ electron within its orbital, which 6-fold degeneracy ($j = \frac{5}{2}$, $2j + 1 = 6$) is lifted due to the crystal electric field. It is not well understood yet whether a whole band of optical phonons couples to the transition, as originally proposed for CeAl$_2$ [10, 11] and CeCuAl$_3$ [8], or a transverse phonon within a particular $k$-vector range, as suggested for CeAuAl$_3$ [9]. It is also unknown whether the VBS has some connection to superconductivity, or affects the long-range magnetic order.

Neutron diffraction studies in the magnetically ordered state in CeTaAl$_3$ are so far reported for $T$ = Cu and Au only. CeCuAl$_3$ exhibits an amplitude modulated, collinear, antiferromagnetic (AFM) structure, with $k = (0.40.60)$ below $T_N = 2.7$ K [12], while CeAuAl$_3$ exhibits a helical AFM structure with $k = (0.00.52)$ below $T_N = 1.2$ K [13]. Bulk properties measurements, such as heat capacity, electric resistivity, magnetization and magnetic susceptibility show that CeAgAl$_3$ is a ferromagnet at $T_C < 3.8$ K [14]. There are conflicting findings on CePtAl$_3$ and CePdAl$_3$. No evidence of a long-range magnetic order in CePtAl$_3$ and CePdAl$_3$ down to $\approx 0.1$ K was reported by Franz et al. (2016) [15], while previous works report spin-glass behaviour in CePtAl$_3$ below $T_c = 0.8$ K [16], and AFM ordering in CePdAl$_3$ below $T_N = 6$ K [17]. Although Schank et al. [17] ascribed the lack of long-range magnetic order in some CeTaAl$_3$ compounds due to strong hybridization between 4$f$ and conduction band electrons, studies of their crystal structure and differences originating from different synthesis method, provide alternative explanation.

CeTaAl$_3$ compounds crystallise in a variance of the tetragonal BaAl$_4$ structure [18], a representative of the disordered ThCr$_2$Si$_2$ structure type [19], space group $I4/mmm$, which is shown in Fig. 1A. CeCuAl$_3$, CePtAl$_3$, and CeAuAl$_3$ crystallize in the BaNiSn$_3$ structure type [15], with space group $I4/mmm$, a non-centrosymmetric
variant of the BaAl\(_4\) structure. In BaAl\(_4\), aluminium atoms occupy 4e Wyckoff position [20], while in BaNiSn\(_3\) the 4e position splits into two independent 2a positions (highlighted with double headed arrows throughout Fig.1). In CeTAl\(_3\) the inversion symmetry is broken, since \(T\) atoms tend to occupy one 2a site, while Al atoms the other one. Additionally, their positions, (0,0,\(z_T\)) and (0,0,\(z_{AI}\)) are no longer bound to obey the relation \(z_T + z_{AI} = 1\) imposed by inversion symmetry. There can exist a certain, sample dependent level of occupational disorder between \(T\) and Al 2a sites in stoichiometric CeTAl\(_3\) compounds, which is obviously higher in solid solutions such as CeCu\(_x\)Al\(_{4-x}\) [21]. On the other hand, CeAgAl\(_3\) was reported to crystallise in a centrosymmetric, orthorhombic PbSbO\(_3\)Cl structure type \([15]\) with space group \(Cmcm\) (Fig.1C). It can be viewed as an orthorhombic distortion of the tetragonal unit cell, which introduces various changes in interatomic distances. Interestingly, the inversion symmetry is restored, but it doesn’t fall on a particular atomic site. In BaAl\(_4\) the Ba atom lies on a centrosymmetric site, while in CeAgAl\(_3\) the inversion symmetry lies in the corner of the unit cell not occupied by any atom, see Fig.1C. As such, the local environment of the cerium atom does not define the inversion symmetry in CeAgAl\(_3\), in opposite to compounds with BaNiSn\(_3\) and BaAl\(_4\) structure. Finally, recent results on the crystal structure of CePdAl\(_3\) from Franz et al. (2016) \([15]\) have shown some inconsistencies between the single crystal sample obtained from the CePdAl\(_3\) ingot and the ground polycrystalline sample, measured by single-crystal and powder X-ray diffraction, respectively. For the polycrystalline sample, the reported structure was that of the BaNiSn\(_3\) type with the \(I4mm\) space group and \(a = b = 4.3\,\text{Å}\) and \(c = 10.3\,\text{Å}\), while the single crystal shows the BaNiSn\(_3\) superstructure with a \(3 \times 3 \times 1\) unit cell \((a \times b \times c)\) and splitting of all atomic positions, indicating a correlated disorder. More generally, the crystal structure of CeTAl\(_3\) compounds can be described as a three dimensional \([TAI]_3\) network, forming large cages occupied by Ce atoms. This network is highlighted in Fig. 1 by connecting \(T\) and Al atoms, and has different degrees of distortion in BaAl\(_4\), CeCuAl\(_3\), and CeAgAl\(_3\). But, the exact origin of the distortion is unknown.

In summary, there are conflicting reports on the crystal structure of CePdAl\(_3\) and magnetic ordering in CePdAl\(_3\) and CePtAl\(_3\). It seems that they originate from differences in sample synthesis and lack of direct measurements of magnetic ordering, which motivated us to perform single crystal neutron diffraction measurements on carefully grown, large single crystals of CePdAl\(_3\) and CePtAl\(_3\). In addition, complementary X-ray powder diffraction was performed on CePdAl\(_3\) to verify the structural differences in its polycrystalline and single-crystal form. Various neutron diffraction techniques and instruments at MLZ (Germany) were employed in order to provide proper characterization of magnetic ordering in CePdAl\(_3\) and CePtAl\(_3\).

II. EXPERIMENTAL METHODS

A. Sample synthesis and characterisation

Centimetre size single crystals were grown from polycrystalline rods in the optical floating zone furnace, following the method of \([15]\), described in more details in \([22, 23]\). The CePtAl\(_3\) crystal measured in the current study comes from the same synthesis batch as in \([15]\). Our CePdAl\(_3\) single crystal was grown in an optical furnace with a reduced growth rate of 1 \(\text{mm/h}\), compared to 5 \(\text{mm/h}\) in the original synthesis project \([15]\). This change allowed to grow a centimetre long crystalline rod, out of which single crystals of millimetre size were extracted for neutron diffraction measurements. An end part of the crystallized CePdAl\(_3\) rod was grinded, in order to obtain a polycrystalline sample for complementary X-ray powder diffraction measurements.

CePtAl\(_3\) was intensively characterised by magneti-
tion, susceptibility, Hall effect and specific heat measurements, as reported in [22, 24], while the characterization of our CePdAl₃ crystals are published in parallel by Kumar et al. [25]. The specific heat data presented in the following were measured in a Quantum Design physical properties measurement system at temperatures down to 2 K using a quasi-adiabatic large heat pulse method.

### B. Neutron and X-ray diffraction

Modulation vectors of magnetic ordering in CePtAl₃ and CePdAl₃ were determined from measurements at DNS [26] equipped with a 2D detector bank allowing to measure large volumes of reciprocal space, and with xypolarisation analysis, that allows to distinguish between magnetic and nuclear signal [27]. The incoming wavelength of 4.2 Å allowed to measure a momentum transfer range Q of 0.3-2.7 Å⁻¹. The samples were oriented in the (HK0) plane for CePdAl₃ and (H0L) for CePtAl₃. They were cooled below their magnetic ordering temperatures, 3 K for CePdAl₃ and 0.45 K for CePtAl₃, respectively. The data were analysed and reduced with the Mantid package [28] allowing to project the measured intensities on selected crystal planes.

X-ray diffraction measurements were performed with polycrystalline CePdAl₃ on a Stoe Stadi P diffractometer, with wavelength 1.54 Å, in 2θ range of 5-100 deg. and step size of 0.015 deg., resulting in Q range of 0.7-8 Å⁻¹. Possible lattice types were cross-checked with the EXPO2014 program suite [29] and refined with Jana2006 [30].

The neutron single crystal diffraction measurements on CePdAl₃ were performed on HEiDi [31] with a neutron wavelength 0.793 Å at temperatures between 300 K and 2.4 K employing a closed-cycle cryostat. The measurements show that the crystal was twinned, which obscures the data analysis. Attempts to detwin the crystal by extracting smaller sample pieces did not prove successful. We have manually discarded the reflections with poor profile or background and performed the refinement on a non-averaged set of reflections. We excluded the reflections which measured intensity differs highly from the calculated ones, specifically by \( I_m - I_c > 10 \sigma(I_m) \), where \( I_m \) is the measured intensity, \( I_c \) the calculated intensity and \( \sigma(I_m) \) the estimated error of measured intensity.

Single crystal neutron diffraction measurements on CePtAl₃ were performed on POLI [32]. The sample was measured at ambient temperature, as well as at 0.45 K in a {parent} cryostat. We have attempted to measure magnetic reflections at 0.45 K, but their intensities were too low to be determined reliably. We performed additional measurements on PANDA [33], a cold three-axis spectrometer (TAS) with low background, and obtained the intensities of 28 magnetic reflections in the (H0L) plane.

### III. RESULTS

#### A. CePdAl₃ crystal structure

The analysis of diffractograms from X-ray powder diffraction on CePdAl₃ provides the best indexation with a C-centred orthorhombic lattice and lattice parameters \( a=6.379 \text{ Å}, \ b=10.407 \text{ Å}, \ c=5.975 \text{ Å} \) at ambient conditions. This is quite different from the result of the first synthesis attempt in [15], where CePdAl₃ was reported to be of tetragonal \( I4mm \) structure, with \( a=4.344 \text{ Å} \) and \( c=10.592 \text{ Å} \) for the polycrystalline sample, and \( a=12.988 \text{ Å} \) and \( c=10.580 \text{ Å} \) for the single crystal. We interpret this inconsistency as a structural instability that originates from subtle changes to the synthesis process and will be discussed later.

There are two candidates for the CePdAl₃ space group that follow the extinction rules and give low R-factors: the non-centrosymmetric \( Cmc2_1 \) and the centrosymmetric \( Cmcm \) space group. A model with \( Cmc2_1 \) gives a slightly lower R-factor, \( R(Cmc2_1)=2.75 \) compared to \( R(Cmcm)=2.9 \), but it allows a larger number of free parameters, 32 for \( Cmc2_1 \) model and 27 for \( Cmcm \) model, respectively. The main difference between them is that the \( Cmc2_1 \) space group allows to free the \( z \) component of the atomic positions, which is fixed in the \( Cmcm \) model. These refined values differ slightly from the fixed ones within a range of 0.1-0.2 Å.

Consecutive single-crystal neutron diffraction measurements on CePdAl₃ at ambient conditions give similar results. The best candidate is the model with \( Cmc2_1 \) symmetry with \( R(Cmc2_1)=6.32 \) and 24 parameters refined, while the \( Cmcm \) model is second best with \( R(Cmcm)=7.21 \) and 19 parameters refined. The refined atomic positions are the same as obtained from powder diffraction within the experimental error. The resulting crystal structure is shown in Fig. 2A.

Systematic overlap and splitting of selected reflections show a twinning of the measured crystal (see Fig. 4A). The twins have a common \( b \) axis and interchanged \( a \) and \( c \) axes, i.e. they are rotated by 90 deg around the \( b \) axis with respect to each other. Note, that in the orthorhombic setting the axes are changed, and the common \( b \) axis for the twins is actually the four-fold axis \( c_t \) in the tetragonal setting, as shown in Fig. 1C. Thus, the twinning is of pseudo-tetragonal type, where the four-fold axis is lost, while the diagonal mirror planes are preserved.

Considering the change of the crystal axes, the orthorhombic lattice parameters are \( a \approx \sqrt{2}a_t, \ b = c_t, \ c \approx \sqrt{2}a_t \), and the difference \( a-c \) can be used to quantify the degree of distortion, as discussed later. The partial overlap of the reflection obscures the analysis, but it was carefully taken care of, as mentioned in section IIIB. In both models the refinement of atomic displacement parameters (ADP) yields displacement ellipsoids flattened
along the $b$-direction. This might occur due to twinning, since the $b$-axis is common for both twin domains, and insufficient number of high-2$\theta$ reflections. On the other hand, the refinement was not improved by considering positional or substitutional disorder, which in some cases led to negative ADP or divergence. In conclusion, the single-crystal diffraction data allows to conclude with high degree of confidence that the symmetry of CePdAl$_3$ crystal synthesised in this study is described with non-centrosymmetric Cmc$_2$$_1$ space group, and the crystal structure shown in Fig. 2A.

Thus, CePdAl$_3$ is closely related to CeAgAl$_3$, which crystallizes in the Cmcm space group with lattice parameters $a = 6.205\,\text{Å}$, $b = 10.837\,\text{Å}$ and $c = 6.118\,\text{Å}$ and similar crystal structure at ambient conditions [15]. They both exhibit an orthorhombic distortion from the tetragonal structure, as illustrated in Fig.1. The magnitude of that distortion is higher in CePdAl$_3$, for which $a-c \approx 0.4\,\text{Å}$ while for CeAgAl$_3$ $a-c \approx 0.1\,\text{Å}$. Just as for the tetragonal CeTAl$_3$, the structure of CePdAl$_3$ consists of three-dimensional $T$-Al network forming large cages occupied by Ce atoms. Interestingly, the orthorhombic distortion does not split the nearest-neighbour Ce-Ce distances within the $ac$-plane, as the four nearest-neighbours are all $4.356\,\text{Å}$ apart, see Fig. 2B. On the other hand, the next-nearest-neighbours are of different distances, given by $a$ and $c$ axes lengths, as well as the four Ce atoms from neighbouring $ac$ planes, that form the pseudo-tetragonal unit cell. In the undistorted scenario the distances between the central atom and eight neighbours forming the corners of pseudo-tetragonal cell would be the same, however, in CePdAl$_3$ these distances vary between $5.67\,\text{Å}$ and $6.22\,\text{Å}$, see Fig. 2B and Supplementary Materials [34].

B. Low-temperature specific heat of CePdAl$_3$

The specific heat of the orthorhombic CePdAl$_3$ is shown in Fig. 3. It is characterized by a sharp, lambda-like anomaly with a maximum at $T_1 = 5.4\,\text{K}$ and a shoulder with a cut-off at $T_2 = 5.7\,\text{K}$. No further phase transitions were observed at temperatures up to $300\,\text{K}$. For comparison, the specific heat of the tetragonal CePdAl$_3$ from Ref. [22] is also shown in Fig. 3, lacking phase transitions down to $2\,\text{K}$, consistent with the resistivity measurements in Ref. [15]. Note that a double anomaly in specific heat was also observed by Schank et al. [17] and interpreted in terms of a transition to an antiferromagnetic state, but no analysis of the crystal structure was presented.

C. CePdAl$_3$ magnetic structure

The mapping of the reciprocal space of CePdAl$_3$ by neutron diffraction at $3\,\text{K}$, i.e. below the transition marked by heat capacity measurements, is shown in Fig. 4. The reflections are separated in purely nuclear and magnetic contributions owing to polarization analysis, Fig. 4A and Fig. 4B, respectively. Some nuclear reflections are split, which is consistent with the twinning described in Sec. III A. In reciprocal space the twinning translates simply to common $k$ and interchanged $h$ and $l$ indices. The reciprocal space was mapped to crystal coordinates by indexing the majority twin, indexed with subscript 1 in Fig. 4. In result, the reflection of the minority twin, subscript 2 in Fig. 4 are not aligned with the grid. Taking twinning into account, all magnetic

![Figure 2](https://example.com/figure2.png)

Figure 2. (A) Crystal structure of the orthorhombic CePdAl$_3$. (B) Ce-Ce atomic distances around the atom indicated with blue arrow in panel (A). (C) Antiferromagnetic arrangement of Ce magnetic moments in CePdAl$_3$, the same atoms as in (B) are shown. (D) View along the $b$-axis, showing the ferromagnetic sheets stacked along the $c$-axis direction. In panels (B),(C),(D) Pd and Al atoms are omitted for clarity.

![Figure 3](https://example.com/figure3.png)

Figure 3. Specific heat as a function of temperature. Data of orthorhombic CePdAl$_3$ (red squares) synthesized in this study is compared with data of tetragonal CePdAl$_3$ (black circles) synthesized by Franz et al. [15].
reflections were observed at integer positions, indicating a commensurate magnetic structure, with magnetic unit cell the same size as the chemical unit cell. Some magnetic reflections are observed in the positions forbidden by the extinction rules established by $Cmc_2_1$ space group, indicating an antiferromagnetic type of ordering.

Figure 4. Neutron diffraction intensity in the (HK0) plane of CePdAl$_3$ at 3 K, measured at DNS. (A) Intensity of the non-spin-flip (NSF) channel, where nuclear Bragg reflections are observed, and (B) in the spin-flip channel (SF), where magnetic reflections are visible. The crystal was twinned, which results in splitting and misalignment of some reflections. Indices subscripts indicate two twins.

The transition temperature of CePdAl$_3$ was determined upon cooling by monitoring the intensities of selected reflections in two temperature regions. Between ambient temperature and 10 K, no significant changes in intensity or positions of the nuclear reflections were observed, which together with heat capacity data, allows to conclude that there is no structural phase transition in this temperature region. While cooling from 10 K to 2.4 K the same nuclear reflections do not show any further changes, while some magnetic reflections emerge around 5 K. The profile and temperature behaviour of the representative (001) is shown in Fig. 5. The intensity of the reflection as a function of temperature follows the power-law behaviour $I \propto (1-T/T_N)^{2\beta}$. The fit yields the transition temperature $T_N=5.3 (1)$ K and critical exponent $\beta=0.24 (2)$. Although the heat capacity data shows two characteristic temperatures, sharp peak at $T_1=5.4$ K and a shoulder at $T_2=5.7$ K, the neutron diffraction data were recorded with temperature step of 0.2 K and do not indicate the existence of a double transition.

Full data collection was performed at 10 K and 2.4 K. Crystal structure refinement to the dataset at 10 K and to nuclear reflections at 2.4 K gives the same structure as at ambient conditions (Fig. 2A). The symmetry analysis shows, that the $Cmc_2_1$ space group allows four types of magnetic ordering following the decomposition into irreducible representations: $\Gamma_{Mag} = 1\Gamma_1^1 + 2\Gamma_2^2 + 1\Gamma_3^3 + 2\Gamma_4^4$.

The best fit to the data at 2.4 K is obtained with the $\Gamma_1^1$ representation, which corresponds to magnetic space group $Cmc_2_1$ (no. 36.172). This describes a collinear, antiferromagnetic ordering of Ce atoms, with moments along the $a$-axis direction, as shown in Fig. 2C. Such ordering forms ferromagnetic sheets within the $ab$-plane, stacked along the $c$-axis direction (see Fig. 2D), with refined value of the ordered magnetic moment of $\mu_{Ce}=1.672 (72) \mu_B$.

Figure 5. (A) Profile of the (001) magnetic reflection of CePdAl$_3$ measured at HEiDi showing the development of magnetic ordering. Points indicate measured data, lines are guides to the eye. (B) Integrated intensity of (100) reflection as a function of temperature shown with circles and errorbars, together with a fit of the order parameter, shown with solid line.

D. CePtAl$_3$ crystal structure

Single-crystal neutron diffraction at ambient conditions confirms that CePtAl$_3$ crystallises in the BaNiSn$_3$ structure with non-centrosymmetric $I4mm$ space group, as previously reported by [15], and shown in Fig. 1B. A large number of measured reflections, 343 independent reflections, and the completeness of the dataset allow for a reliable refinement of the displacement parameters. It results in a significant elongation of the displacement ellipsoid on the 2a Al site along the $c$ axis, while other sites show fairly isotropic displacement. Such behaviour suggests a positional disorder of the Al site. In addition, the refinement improves significantly by considering occupational disorder, i.e. mixing between the 2a site of Pt and Al, marked with double headed arrow in Fig. 1B. Direct evidence of atomic disorder in the sample is also observed in form of diffuse Huang scattering [35], as shown in Fig 8B. Profile of the (002) nuclear reflection is composed of a narrow, resolution limited, diffraction signal, with full-width at half-maximum (FWHM) 0.91 deg., and a broader diffusive component with FWHM of 3.88 deg.

The best refinement is obtained with simultaneous modelling of the positional and occupational disorder, in
a way that the Pt site is occupied in 76% by Pt atoms and in 24% by Al atoms, while the Al site in 76% by Al atoms and 24% by Pt atoms. The Al and Pt atoms at the $2a$ Al site are separated by a distance of 0.4 Å along the $c$ axis direction, which is 4% of the length of the $c$ axis. The occupancies are constrained to retain the stoichiometric composition CePtAl$_3$, refinements without such constraint do not improve the quality of the fit.

Measurements at 1.5 K show that the refined crystal structure of CePtAl$_3$ is the same as at ambient conditions, with small differences in atomic positions, which proves that CePtAl$_3$ does not undergo a structural phase transition in this temperature range.

### E. Low-temperature specific heat of CePtAl$_3$

The specific heat of CePtAl$_3$, shown in Fig. 6, exhibits a maximum centered at $T_c = 2.8$ K that is considerably broader than the one in orthorhombic CePdAl$_3$, see Fig. 3. The height of the peak is also relatively small, 4 J mol$^{-1}$ K$^{-1}$ in CePtAl$_3$ compared to 18 J mol$^{-1}$ K$^{-1}$ in CePdAl$_3$. Taken together, the specific heat of CePtAl$_3$ suggests that onset of magnetic order takes place across a rather wide temperature range. Note that this intrinsic broadening may also be responsible for the lack of signatures of the magnetic phase transition in electrical resistivity data reported by Franz et al. [15].

![Figure 6](image6.png)

**Figure 6.** Specific heat as a function of temperature for CePtAl$_3$. A broad maximum is observed around $T_c = 2.8$ K.

### F. CePtAl$_3$ magnetic structure

In search for signs of long-range magnetic order in CePtAl$_3$, its reciprocal space was mapped by neutron diffraction at 0.45 K. In addition to the nuclear reflections shown in Fig. 7A, which can be indexed according to the tetragonal $I4mm$ lattice, ten weak magnetic reflections were observed, as shown in Fig. 7B. Their intensities are roughly two orders of magnitude lower than of nuclear reflections, and they can be indexed by considering the modulation vector $\mathbf{k} = (\frac{2}{3} 0 0)$. According to the tetragonal symmetry, magnetic reflections with modulation vectors from the full star of the $\mathbf{k}$ vector should be observed, which is $k_1 = (\pm \frac{2}{3} 0 0)$ and $k_2 = (0 \pm \frac{2}{3} 0)$, generated by the four-fold symmetry axis $c$. Due to the experimental limitations only reflections in the (H0L) plane could be measured. This implication on the determination of the magnetic structure symmetry will be discussed later.

![Figure 7](image7.png)

**Figure 7.** Neutron diffraction intensity in the (H0L) plane of CePtAl$_3$ at 0.45 K, measured at DNS. (A) Non-spin-flip channel (NSF), where nuclear reflections are observed, (B) spin-flip channel (SF), where magnetic reflections with modulation vector $\mathbf{k} = (\frac{2}{3} 0 0)$ are observed.

Upon cooling, the intensities of the magnetic (1 1 0) and nuclear (0 0 2) reflections were recorded in more detail (Fig. 8). The intensity of the (1 1 0) magnetic reflection allows to determine the Néel temperature. Close to the transition temperature, around 3.5 K, the power-law behavior $I \propto (1 - T/T_N)^{2\beta}$ does not fit the intensity well (see Fig. 8C and inset). A similar effect was reported for MnO in [36], where the order parameter, i.e. intensity of the reflection, does not fall sharply to zero at $T_N$, but is smeared around the transition, due to compositional inhomogeneities. The same model can be applied here. The average crystal structure of CePtAl$_3$ is described with occupational and positional disorder (Sec. IIID), which relates to local structural inhomogeneities in the sample. In result, different regions of the sample will exhibit slightly different magnetic transition temperature $T_N$, and the distribution of $T_N$ can be described by a Gaussian function with an average Néel temperature $T_N$ and its standard deviation $\sigma_{T_N}$. This distribution is then convoluted with the power-law dependence of the order parameter and fit to observed magnetic intensities, which results in $T_N = 3.2$ K and $\sigma_{T_N} = 0.4$ K. This model also explains the broad signature in the heat capacity data.

In order to establish a proper model of the magnetic structure, the allowed symmetries of the magnetic ordering are evaluated with the ISODISTORT package [37] su-
perspace group approach. Considering the I4mm space group and a magnetic modulation vector \( \mathbf{k} = (\frac{2}{3} 0 0) \), the resulting possibilities are either a tetragonal structure with multiple modulation vectors (multi-k) from the full star of \( \mathbf{k} \), \( \mathbf{k}_1 = (\pm \frac{2}{3} 0 0) \) and \( \mathbf{k}_2 = (0 \pm \frac{2}{3} 0) \), or an orthorhombic structure with only one modulation vector \( \mathbf{k} = (\pm \frac{2}{3} 0 0) \) (single-k). Since all measurements were performed within the (H0L) plane, it is not possible to verify the existence of the reflections with \( k_2 = (0 \frac{2}{3} 0) \), and therefore, to distinguish between the tetragonal and orthorhombic model. However, there are indications favouring the tetragonal model: (1) lack of reflection splitting in the DNS mapping data (Fig. 7), and (2) very good refinement of the structure below \( T_N \) at 1.5 K with the tetragonal model, where the equivalent twinned, orthorhombic I4mm2 model does not improve the fit. Nevertheless, both multi-k tetragonal and single-k orthorhombic structures need to be considered.

For both models the best fit was obtained with the \(-\sigma_2\) representation of either the multi-k, tetragonal \( I4mm1'(a, 0, 0)00s(0, a, 0)s0s \) superspace group or the single-k, orthorhombic \( I2mm1'(0, 0, g)s0s \) superspace group. In both cases there are two free parameters describing the magnetic ordering, a component \( m_{\parallel} \) along the \( c \)-axis, and a component \( m_{\perp} \) perpendicular to it. The orthorhombic model assumes the existence of magnetic domains and involves an additional parameter, the domain population factor, set to 0.5. Despite the similarities in these models so far, the ordering of the moments in the real space is quite different. The magnetic moment associated with the Ce atom at position \( r = [x, y, z] \) in the tetragonal model (superscript t) is defined as:

\[
m_t(r) = [m_{\parallel t}(kx), m_{\perp t}(ky), m_{\perp t}(\cos(kx)+\cos(ky))],
\]

while in the orthorhombic model (superscript o) as

\[
m_o^r(r) = [m_{o}^r \sin(kx), 0, m_{o}^r \cos(kx)],
\]

assuming a single propagation vector along the \( a \) axis \( k = (\frac{2}{3} 0 0) \).

Fig. 9 shows the resulting arrangements of magnetic moments considering both models. In the multi-k model (Fig.9A and Fig.9B), the magnetic unit cell is a 3x3x1 supercell of the chemical unit cell. Within a single layer with \( z=0.0 \) or \( z=0.5 \), there is a central Ce atom with magnetic moment \( m \) along the \( c \) axis, and its eight neighbours with magnetic moments pointing towards this central Ce atom, slightly canted out of the \( ab \)-plane, thus resembling a mini-vortex. These vortices are arranged in a two-dimensional square lattice with periodicity of 3\( a \). The magnetic moment amplitudes are refined to values of \( m_{\parallel} = 0.836(83) \mu_B \) and \( m_{\perp} = 0.339(78) \mu_B \). There is a slight amplitude modulation of the magnetic moment magnitude, such that the central spins with \( m \parallel c \) have a total moment of \( |m| = 0.678 \mu_B \), its four closer neighbours (atoms lying along \( a \) or \( b \) crystal axis) \( |m| = 0.744 \mu_B \) and are canted 18 deg. upwards, while the four diagonal neighbours \( |m| = 1.08 \mu_B \) and are canted 18 deg. downwards from the \( ab \)-plane (Fig. 9B).

The single-k model has a unit cell of 3x1x1 of the chemical unit cell, and shows a cycloidal modulation of the magnetic moments, as shown in Fig. 9C. The amplitudes are refined to the values of \( m_{\parallel} = 0.48(11) \mu_B \) and \( m_{\perp} = 1.18(12) \mu_B \), which also gives rise to an amplitude modulation, but only with two values: 0.48\( \mu_B \) for moments pointing along \( c \)-axis and 1.05\( \mu_B \) for the ones pointing 13 deg. away from the \( a \)-axis direction, see Fig. 9D.

IV. DISCUSSION

The combined X-ray and neutron diffraction measurements show unambiguously, that the CePdAl\(_3\) sample in-
Figure 9. Magnetic unit cell of CePtAl$_3$ in the tetragonal (A,B) and orthorhombic (C,D) models. Only cerium atoms are shown for clarity, and their magnetic moments are marked with red arrows. Ce atoms are coloured according to the amplitude of their magnetic moment, with the values given in the legends below panels. (A) Magnetic unit cell in the multi-$k$ tetragonal model showing layers of cerium atoms with $z$=0, 0.5, 1. (B) View along the c-axis of cerium atoms with $z$=0.5, showing the mini-vortex structure of multi-$k$ model. (C) Three magnetic unit cells in the single-$k$ orthorhombic model, stacked along the b-axis direction to facilitate comparison with the multi-$k$ model. (D) Arrangement of magnetic moments shown along the b-axis, in the single-$k$ model.

vestigated in this study crystallises in the $Cmc2_1$ space group, a non-centrosymmetric variant of the PbSbO$_2$Cl structure type [38] with $Cmcm$ space group. The centrosymmetric $Cmcm$ structure was observed for CeAgAl$_3$ [14, 15] and some SrPdGa$_3$ intermetallics [39]. However, according to the International Crystal Structure Database [40], the $Cmc2_1$ structure variant of CePdAl$_3$ investigated in this study is a unique structure type, never reported before. Lack of inversion symmetry in CePdAl$_3$ allows to free the atomic $z$ coordinate, which can be interpreted as an accommodation of further distortion from the $Cmcm$ structure of CeAgAl$_3$, on top of the orthorhombic distortion exhibited by CePdAl$_3$ and CeAgAl$_3$ from the tetragonal compounds such as CePtAl$_3$, CeAuAl$_3$, and CeCuAl$_3$, illustrated in Fig. 1.

Comparing current results to [15] and [17], it is clear that the synthesis method greatly affects the structure and crystalline quality of the grown CePdAl$_3$ crystals. While the original method described in [15] results in tetragonal crystals of CePdAl$_3$ which do not order magnetically down to 0.1 K, a slower growth process in the optical furnace provides large crystals with orthorhombic $Cmc2_1$ structure and antiferromagnetic order below $T_N$=5.3 K. Important to say, the polycrystalline sample and millimeter size single-crystal reported in this study exhibit the same crystal structure.

Interestingly, [17] synthesized a polycrystalline sample of tetragonal CePdAl$_3$ with lattice parameters similar to [15], and obtained a sample with unknown structure after annealing at around 700-900°C for a week. Upon measuring the heat capacity of their sample, they observed the same signature as shown in Fig. 3, i.e. sharp lambda peak with a shoulder at the high temperature side. Possibly, after annealing they obtained the same orthorhombic CePdAl$_3$ as reported in the current study, while their initial synthesis attempt, as well as that of Franz et al. [15], involves quenching the high-temperature tetragonal phase, which is metastable at ambient conditions. This scenario suggests that the orthorhombic $Cmc2_1$ structure of CePdAl$_3$ reported here is thermodynamically stable at ambient conditions, and CePdAl$_3$ undergoes a structural phase transition from tetragonal to orthorhombic structure between ambient temperature and 1200 K, responsible for pseudo-tetragonal twinning.

The fact that tetragonal CePdAl$_3$ does not order magnetically at low temperatures, contrary to orthorhombic, points to the crucial role of the atomic disorder, or lack thereof, that can stabilise long range magnetic ordering in CePdAl$_3$. Such effect was observed prominently in another Ce-113 compound, CeCuGa$_3$ [41]. There, a sample with occupational disorder of Cu and Ga atoms, i.e. BaAl$_4$ structure, adapts ferromagnetic ordering, while the sample with an ordered BaNiSn$_3$ structure orders antiferromagnetically. Doping the sample with larger amount of Cu, with stoichiometries CeCu$_x$Ga$_{4-x}$ and $1 < x < 1.5$, suppresses long-range magnetic order in the investigated temperature range.

Such a strong correlation between disorder and magnetism provides an interesting platform for material engineering, where different physical properties, the magnetism in this case, can be tuned for a specific material by carefully choosing the synthesis parameters.
The orthorhombic CePdAl\(_3\) orders in a collinear antiferromagnetic structure with moments pointing along the \(a\)-axis direction, as shown in Fig. 2C, with a transition temperature \(T_N = 5.3\) K. The value of the ordered magnetic moment in CePdAl\(_3\) is 1.667\(\mu_B/\text{Ce}^{3+}\) at 2.4 K, which is the highest in CeTAI\(_3\) compounds investigated so far (see Table I).

This simple type of magnetic order is quite different from the modulated types reported for tetragonal CeCuAl\(_3\) [12] and CeAuAl\(_3\) [13] (see Tab. I), but it bears strong resemblance to the ferromagnetic ordering in the orthorhombic CeAgAl\(_3\). Based on magnetization and magnetic susceptibility measurements CeAgAl\(_3\) orders ferromagnetically below \(T_C = 3.8\) K with a magnetic moment of roughly 1\(\mu_B/\text{Ce}^{3+}\) [14]. The ordered moments are restricted to be in the \(ab\) plane [22].

Our results confirm that CePtAl\(_3\) crystallises in the BaNiSn\(_3\) structure with \(I4mm\) space group. The crystal exhibits a certain level of occupational and positional disorder between the Pt and Al sites (see red arrows in Fig. 1), that also manifests in Huang scattering around nuclear reflections, see Fig. 8D, and smearing of the order parameter, see Fig. 8B. In contrast to previous reports it exhibits a modulated, long-range magnetic order with a transition temperature \(T_N = 3.2\) K. The transition is not sharp, which might be the reason of the broad features in resistivity [15], which do not match the magnetic transition temperature, and suggests a distribution of \(T_N\) due to structural inhomogeneity.

The exact determination of magnetic structure is flawed by the fact that only \((h0l)\) reflections were measured, and cannot be distinguished between a tetragonal multi-\(k\) model and twinned, orthorhombic single-\(k\) model. Interestingly, the model of the magnetic structure of the tetragonal CeCuAl\(_3\) proposed by [12], is of orthorhombic symmetry, since the magnetic moments are arranged along the \(11\overline{1}0\) diagonal, clearly favoured over the model with moments pointing along the \(11\overline{0}\) diagonal, which is equivalent to \((11\overline{0})\) assuming tetragonal symmetry. In fact, just as in case of CePtAl\(_3\), constraining the model to tetragonal symmetry leads to a multi-\(k\) structure with a complex arrangement of magnetic moments, discussed in more detail in Supplementary Materials ??.

The determination of the magnetic order in CePdAl\(_3\) and CePtAl\(_3\) facilitates an overview of magnetism in CeTAI\(_3\) compounds, summarised in Table I. One can clearly conclude that the tetragonal CeTAI\(_3\) compounds exhibit modulated, non-collinear types of ordering, while the orthorhombic ones collinear structures, commensurate with the chemical unit cell. This points to the important role of the orthorhombic distortion, that releases some underlying frustrations in these systems, without which the tetragonal systems exhibit complex magnetic ordering. As pointed out in Fig. 2B, the orthorhombic distortion does not split the nearest-neighbour (NN) Ce-Ce distances, which defines the length of the \(a\)-axis of the tetragonal unit cell. In fact, this distance is very similar in CePdAl\(_3\), 4.356 \(\text{Å}\), and CeAgAl\(_3\), 4.358 \(\text{Å}\), even though the former is antiferromagnetic and latter ferromagnetic. More prominent differences are observed for the twelve next-nearest-neighbour (NNN) Ce-Ce distances, which vary between 5.67 \(\text{Å}\) and 6.37 \(\text{Å}\) in CePdAl\(_3\) and between 6.12 \(\text{Å}\) and 6.31 \(\text{Å}\) in CeAgAl\(_3\). For comparison, in tetragonal CePtAl\(_3\), the NN distances are 4.329 \(\text{Å}\), while twelve NNN are all roughly 6.14 \(\text{Å}\). This provides a clue that subtle changes in the NNN magnetic interaction might influence the ordering, since these compounds all exhibit different kind of magnetic ordering. In fact, Ce-based intermetallics are expected to possess complex magnetic behaviour, since the localised magnetic moment of Ce exhibit Kondo screening effect, providing a mixed localised-itinerant type of magnetism [8, 16, 42]. As such, the simplistic arguments based on interatomic distances don’t predict well the type of magnetism exhibited by these compounds.

Klicpera et al. [12] suggest a diagram that relates the magnetic modulation vector of CeTX\(_3\) compounds based on unit cell volume and NNN Ce distances. It predicts that both CePdAl\(_3\) and CePdAl\(_3\) should possess \(k = (00k_z)\) modulation vector, which is clearly not the case for CePdAl\(_3\), and it is disputable, whether orthorhombic CePdAl\(_3\) falls into the diagram at all or its commensurate type of order fits into the diagram with \(k_z = 0\). From our findings, one could assume that Ce NNN distances are more important for the magnetic properties than NNN distances.

In summary, the CeTAI\(_3\) series provides an interesting variety of magnetic and structural properties, and due to interplay between different degrees of freedom it resists systematic classification so far, while at the same time providing a substantial platform to study those effects.

V. CONCLUSIONS

We have synthesised large single crystals of CePdAl\(_3\) and CePtAl\(_3\) that allowed us to perform detailed single-crystal neutron diffraction measurements, providing magnetic order at low temperatures. Previous inconsistencies considering the crystal structure and magnetic ordering of these compounds are related to different crystal growth procedures, and hint to structural instabilities in the systems.

We have determined that CePdAl\(_3\) crystallises in a non-centrosymmetric, orthorhombic \(\text{Cmcm}\) structure, a distorted variant of the tetragonal \(I4mm\) \(\text{BaAl}_4\) structure. CePdAl\(_3\) orders antiferromagnetically below \(T_N = 5.3\) K with collinear, commensurate magnetic structure.

Precise measurements showed that CePtAl\(_3\) orders magnetically at \(T_N = 3.2\) K, in a modulated antiferromagnetic structure with commensurate modulation vector \(k = (\frac{3}{4}00)\). The ordering of the magnetic moments in real space can be described either by a complex three-
dimensional arrangement, or cycloidal modulation.

Systematic analysis of our data provides a significant role of disorder and frustration onto the magnetic properties of CeTAl$_3$ compounds.

Further studies giving more insight in the relationship of the crystal structure and superconductivity or hybridized elementary modes request either studies under external pressure and inelastic neutron scattering experiments, respectively.

VI. ACKNOWLEDGEMENTS

The work was funded within the DFG project WI33203-1 by providing the position of Michal Stekiel, and by TRR80.

We very much appreciate Anatoliy Senyshyn’s support for the x-ray measurement and data analysis, and kindly acknowledge Thomas Müller for helping to improve the analysis of the DNS data. We also thank for support at the instruments by MLZ staff.

Parts of the neutron measurements were performed on HEiDi and POLI both jointly operated by the RWTH Aachen University and JCNS within the JARA-FIT collaboration.

VII. AUTHOR CONTRIBUTIONS

MS, AS, CP designed research; MS, AS, CF, PC, SW, RS performed research; RS, CF provided samples; MS, SW; MM, KN, HD supported experiments; MS, AS, CF wrote the paper.

[1] E. Bauer, G. Hilscher, H. Michor, Ch. Paul, E. W. Scheidt, A. Gribanov, Yu. Seropegin, H. Noël, M. Sigrist, and P. Rogl. Heavy Fermion Superconductivity and Magnetic Order in Noncentrosymmetric CePt$_3$Si. Phys. Rev. Lett., 92:027003, Jan 2004.

[2] N. Kimura, K. Ito, K. Saitoh, Y. Umeda, H. Aoki, and T. Terashima. Pressure-induced superconductivity in noncentrosymmetric heavy-fermion CeRhSi$_3$. Phys. Rev. Lett., 95:247004, Dec 2005.

[3] Naoyuki Tateiwa, Yoshinori Haga, Tatsuma D. Matsuda, Shugo Ikeda, Etsuji Yamamoto, Yusuke Okuda, Yuichiro Miyauchi, Rikio Settai, and Yoshichika Onuki. Strong-Coupling Superconductivity of CeIrSi$_3$ with the Noncentrosymmetric Crystal Structure. Journal of the Physical Society of Japan, 76(8):083706, 2007.

[4] R. Settai, I. Sugitani, Y. Okuda, A. Thamizhavel, M. Nakashima, Y. Onuki, and H. Harima. Pressure-induced superconductivity in CeCoGe$_3$ without inversion symmetry. J. Magn. Magn. Materials, 310:844, 2007.

[5] Tomoya Kawai, Hiroshi Muranaka, Marie-Aude Measson, Tetsuya Shimoda, Yusuke Doi, Tatsuma D. Matsuda, Yoshinori Haga, Georg Knebel, Gerard Lapertot, Dai Aoki, Jacques Flouquet, Tetsuya Takeuchi, Rikio Settai, and Yoshichika Onuki. Magnetic and Superconducting Properties of CeTX$_3$ (T: Transition Metal and X: Si and Ge) with Non-centrosymmetric Crystal Structure. Journal of the Physical Society of Japan, 77(6):064716–064716, 2008.

[6] Christian Pfeifferer. Superconducting phases of f-electron compounds. Rev. Mod. Phys., 81:1551–1624, Nov 2009.

[7] Ernst Bauer, Heinich Kaldarar, Andry Prokofiev, Esmael Royanian, Alex Amato, Julian Sereni, Werner Brämer-Escamilla, and Ismardo Bonalde. Heavy Fermion Superconductivity and Antiferromagnetic Ordering in CePt$_3$Si without Inversion Symmetry. Journal of the Physical Society of Japan, 76(5):051009, 2007.

[8] D. T. Adroja, A. del Moral, C. de la Fuente, A. Fraile, E. A. Goremychkin, J. W. Taylor, A. D. Hillier, and F. Fernandez-Alonso. Vibron Quasibound State in the Noncentrosymmetric Tetragonal Heavy-Fermion Compound cecu$_3$. Phys. Rev. Lett., 108:216402, May 2012.

[9] Petr Čermák, Astrid Schneiderwind, Benqiong Liu, Michael Marek Koza, Christian Franz, Rudolf Schömann, Oleg Sobolev, and Christian Pfeifferer. Magnetoelastic hybrid excitations in CeAuAl$_3$. Proceedings of the National Academy of Sciences, 116(14):6695–6700, 2019.

[10] Peter Thalmeier and Peter Fulde. Bound State between a Crystal-Field Excitation and a Phonon in CeAl$_2$. Phys. Rev. Lett., 49:1588–1591, Nov 1982.

[11] P Thalmeier. Theory of the bound state between phonons and a CEF excitation in CeAl$_2$. Journal of Physics C: Solid State Physics, 17(23):4153–4177, Aug 1984.

[12] M. Klüpcera, P. Javorský, P. Čermák, A. Schneiderwind, B. Ouladdiaf, and M. Diviš. Neutron scattering study of magnetic order in single-crystalline CeCuAl$_3$. Phys. Rev.
B - Condens. Matter Mater. Phys., 91(22):2–7, 2015.

[13] D. T. Adroja, C. De La Fuente, A. Fraile, A. D. Hillier, A. Daoud-Aladine, W. Kockelmann, J. W. Taylor, M. M. Koza, E. Burzuri, F. Luis, J. I. Arnaudas, and A. Del Moral. Muon spin rotation and neutron scattering study of the noncentrosymmetric tetragonal compound CeAuAl$_3$. Phys. Rev. B - Condens. Matter Mater. Phys., 91(13):1–12, 2015.

[14] S. Nallamuthu, Andrea Dzubinska, Marian Reiffers, Jesus Rodriguez Fernandez, and R. Nagalakshmi. Ferromagnetism in orthorhombic RAgAl$_3$ (R = Ce and Pr) compounds. Physica B: Condensed Matter, 521:128–133, 2017.

[15] C. Franz, A. Senyshyn, A. Regnat, C. Duvinage, R. Schönmann, A. Bauer, Y. Prots, L. Akselrud, V. Hlukhyy, V. Baran, and C. Pfleiderer. Single crystal growth of CeTAl$_3$ (T = Cu, Ag, Au, Pd and Pt). Journal of Alloys and Compounds, 688:978–986, 2016.

[16] S. Mock, C. Pfleiderer, and H. V. Löhnøysen. Low-Temperature Properties of CeTAl$_3$ (T = Au, Cu, Pt) and CeCuGa$_3$. J. Low Temp. Phys., 115(1-2):1–14, 1999.

[17] C Schank, F Jähring, L Luo, A Gruau, C Wissel, R Borth, G Olesch, C.D. Bredl, C Geibel, and F Steglich. 4f-conduction electron hybridization in ternary CeTMAI compounds. Journal of Alloys and Compounds, 207–208(0):329 – 332, 1994.

[18] E. Parthé, B. Chabot, H. F. Braun, and N. Engel. Ternary BaAl$_3$-type derivative structures. Acta Crystallographica Section B, 39(5):588–595, Oct 1983.

[19] Michael Shatruk. ThCr$_2$Si$_2$ structure type: The “perovskite” of intermetallics. Journal of Solid State Chemistry, 272:198–209, 2019.

[20] K.R. Andress and E. Alberti. Roentgenographische Untersuchung der Legierungsreihe Al-Ba. Zeitschrift fuer Metallkunde, 27(6):126–128, 1935.

[21] M. Klicpera, D.T. Adroja, K. Vlăsková, M. Boehm, H. Mutka, B. Ouladdiaf, T. Guidi, and P. Javorský. Magnetic Structure and Excitations in CeCu$_3$Al$_{1-x}$ System. Inorg. Chem., 56:12839–12847, 2017.

[22] Christian Franz. Untersuchung von Quantenphasenübergängen bei fehlender Inversionssymmetrie. Dissertation, Technische Universität München, München, 2014.

[23] A. Bauer, G. Benka, A. Regnat, C. Franz, and C. Pfleiderer. Ultra-high vacuum compatible preparation chain for intermetallic compounds. Review of Scientific Instruments, 87(11):113902, 2016.

[24] Rudolf Schömann. Single crystal growth and low temperature properties of non-centrosymmetric cerium systems, 2015.

[25] V. Kumar and C. Pfleiderer. let’s see. To be published.

[26] Heinz Maier-Leibnitz Zentrum. DNS: Diffuse scattering neutron time-of-flight spectrometer. Journal of large-scale research facilities, 1:A27, 2015.

[27] W. Schweika. XYZ-polarisation analysis of diffuse magnetic neutron scattering from single crystal. J. Phys.: Conf. Ser., 211:012026, 2010.

[28] O. Arnold, J.C. Bilheux, J.M. Borreguero, A. Buts, S.I. Campbell, L. Chapon, M. Doucet, N. Draper, R. Ferrez Leal, M.A. Gig, V.E. Lynch, A. Markvardsen, D.J. Mikkelson, R.L. Mikkelson, R. Miller, K. Palm, P. Parker, G. Passos, T.G. Perring, P.F. Peterson, S. Ren, M.A. Reuter, A.T. Savici, J.W. Taylor, R.J. Taylor, R. Tolchenov, W. Zhou, and J. Zikovsky. Mantid—data analysis and visualization package for neutron scattering and µsr experiments. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 764:156 – 166, 2014.

[29] Angela Altimore, Corrado Cuocci, Carmelo Giacovazzo, Anna Moliterni, Rosanna Rizzi, Nicola Corriero, and Aurelia Falcicchio. EXPO2013: a kit of tools for phaseing crystal structures from powder data. Journal of Applied Crystallography, 46(4):1231–1235, Aug 2013.

[30] Václav Petříček, Michal Dušek, and Lukáš Palatinus. Crystallographic Computing System JANA2006: General features. Zeitschrift für Kristallographie - Crystaline Materials, 229(5):345–352, 2014.

[31] Heinz Maier-Leibnitz Zentrum. HEiDi: Single crystal diffractometer at hot source. Journal of large-scale research facilities, 1:A7, 2015.

[32] Heinz Maier-Leibnitz Zentrum. POLI: Polarised hot neutron diffractometer. Journal of large-scale research facilities, 1:A16, 2015.

[33] Heinz Maier-Leibnitz Zentrum. PANDA: Cold three axes spectrometer. Journal of large-scale research facilities, 1:A12, 2015.

[34] Michal Stekiel. Supplementary Information, 2020.

[35] H. Peisl. Diffuse X-ray scattering from the displacement field of point defects and defect clusters. Journal of Applied Crystallography, 8(2):143–149, Apr 1975.

[36] Marcin Regulski, Radosław Przeniosło, Izabela Sosnowska, and Jens-Uwe Hoffmann. Short and Long Range Magnetic Ordering in β-MnO$_2$ –A Temperature Study-. Journal of the Physical Society of Japan, 73(12):3444–3447, 2004.

[37] Harold T. Stokes, Dorian M. Hatch, Branton J. Campell, and David E. Tanner. Isodispase: a web-based tool for exploring structural distortions. Journal of Applied Crystallography, 39(4):607–614, 2006.

[38] G. Giuseppetti and C. Tadini. Riesame della struttura cristallina della nadorite: PbSbO$_2$ structure type. Zeitschrift für Kristallographie - Crystalline Materials, 207(208):329 – 332, 1994.

[39] Stefan Seidel, Rolf-Dieter Hoffmann, and Rainer Pöttgen. SrPdGa$_2$ – an orthorhombic superstructure of the ThCr$_2$Si$_2$ type. Zeitschrift für Kristallographie - Crystalline Materials, 229(6):421–426, 2014.

[40] G Bergerhoff, ID Brown, F Allen, et al. Crystallographic databases. International Union of Crystallography, Chester, 360:77–95, 1987.

[41] Devang A. Joshi, P. Burger, P. Adelmann, D. Ernst, T. Wolf, K. Sparta, G. Roth, K. Grube, C. Meingast, and H. v. Löhnøysen. Magnetic properties of single-crystalline CeCuGa$_3$. Phys. Rev. B, 86:035144, Jul 2012.

[42] Rainer Pöttgen and Bernard Chevalier. Cerium intermetallic with ZrNiAl-type structure – a review. Zeitschrift für Naturforschung B, 70(5):289–304, 2015.