Pressure effects on the electronic properties of the undoped superconductor ThFeAsN

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The recently synthesized ThFeAsN iron-pnictide superconductor exhibits a $T_c$ of 30 K, the highest of the 1111-type series in absence of chemical doping. To understand how pressure affects its electronic properties, we carried out microscopic investigations up to 3 GPa via magnetization, nuclear magnetic resonance, and muon-spin rotation experiments. The temperature dependence of the $^{75}$As Knight shift, the spin-lattice relaxation rates, and the magnetic penetration depth suggest a multi-band $s^\pm$-wave gap symmetry in the dirty limit, while the gap-to-$T_c$ ratio $\Delta/k_BT_c$ hints at a strong-coupling scenario. Pressure modulates the geometrical parameters, thus reducing $T_c$, as well as $\tau_m$, the temperature where magnetic-relaxation rates are maximized, both at the same rate of approximately $-1.1$ K/GPa. This decrease of $T_c$ with pressure is consistent with band-structure calculations, which relate it to the deformation of the Fe 3d$_{xz,yz}$ orbitals.

Introduction. The doping-induced superconductivity below $T_c = 26$ K in LaFeAsO$_{1-x}$F$_x$ triggered long-term research interests towards iron-based superconductors (FeSC), further boosted by the $T_c = 55$ K of SmFeAsO$_{1-x}$As$_x$. Recently, we reported on superconducting properties of ThFeAsN, an undoped FeSC with a remarkable $T_c$ of 30 K. Our data indicate that Fermi-surface modifications due to structural distortions and correlation effects may be as effective as doping in suppressing the antiferromagnetic order in favor of the formation of a superconducting phase. This is in contrast with most other REFeAsO-type compounds (RE = rare earth), where the quaternary parent compounds usually order magnetically and superconductivity is established via P-doping or H-doping. Due to strong electron correlations (compared with kinetic energy), iron pnictides are immediately coupled systems. For this reason, the experimental values of $T_c$ are distinctly higher than those calculated by assuming the electron-phonon coupling mechanism, which claims $T_c$ values below 1 K. Among the strong-correlation effects, antiferromagnetic (AFM) spin fluctuations are widely accepted to mediate the SC pairing but the detailed interaction model and an unequivocal identification of the gap symmetry are still being debated.

In an attempt to establish (i) what causes the suppression of AFM order in nominally undoped FeSC compounds, (ii) why they become superconductors, and (iii) what determines their $T_c$ values, we investigated ThFeAsN under applied hydrostatic pressure using different local probes. Hydrostatic and/or chemical pressure modify the structure and thus tune the $T_c$ of iron-based superconductors, such as FeSe, whose original $T_c = 8.5$ K increases to 36.7 K at 8.9 GPa. In particular, hydrostatic pressure is regarded as a clean tuning parameter for studying the effects of structural distortions on the electronic properties. A dependence of $T_c$ on the crystallographic As-Fe-As bond angle or on the anion height above the iron layers implies an $h_{pn}$ (see Fig. 1 in Mizuguchi et al.) has previously been noted. With $a = 4.037$ Å and $c = 8.526$ Å, the tetragonal (P4/nmm) structure of ThFeAsN implies an $h_{pn} = 1.305(4)$ Å, lower than the optimum anion height $h_{pn}^{\text{opt}} = 1.38$ Å. Hence, in the case of ThFeAsN, structural deformations induced by hydrostatic pressure would invariably lower $T_c$, in contrast to the above mentioned FeSe case. To test this hypothesis and understand how pressure affects the electronic properties of an undoped 1111 superconducting compound, we performed magnetization-, nuclear magnetic resonance (NMR) and muon-spin rotation ($\mu$SR) measurements on ThFeAsN under applied pressures up to 3 GPa.

First, we confirm experimentally the expected reduction of $T_c$ with pressure. Then, on account of the $T_c$-dependence of the NMR Knight-shifts and spin-lattice relaxation rates, as well as $\mu$SR relaxation rates, we argue that the energy gap $\Delta$ of superconducting ThFeAsN adopts the $s^\pm$ symmetry, which persists up to at least 1.47 GPa. In the same pressure region, the ratio $\Delta/k_BT_c$ is reduced continuously from 2.16(3) at ambient pressure to 1.82(3) at 2.48(2) GPa, thus exceeding the BCS weak-coupling value of 1.76. The moderate variation of $T_c$ with pressure is corroborated by results of band-structure calculations which imply only tiny changes in the electronic excitation spectrum around $E_F$. The abrupt quenching of magnetic excitations, as indicated by a cusp in $1/T_c(T)$ at $T_m$ > $T_c$, persists upon increasing pressure and $T_m$ is reduced at the same rate as $T_c$.

Synthesis and preliminary characterization. The polycrystalline ThFeAsN sample was synthesized via high-temperature solid-state reaction, as reported in Ref. 16. X-ray diffraction and energy-dispersive x-ray measurements confirmed the absence of spurious phases (within $\sim 1\%$).

Magnetization measurements under applied pressure. The magnetization measurements were performed with a superconducting quantum interference device (SQUID) MPMS XL magnetometer. Preliminary measurements at ambient pressure revealed the presence of a tiny quantity of impurities ($\sim 0.18\%$, assuming that they are of ferromagnetic nature). This, along with a broad drop-down in $M(T)$ data below $T_c$, related to defect-induced disorder, suggest that ThFeAsN in the SC phase should be described by models in the dirty limit. Hydrostatic pressures up to 3.1 GPa were achieved by means of a home-made diamond-anvil cell with a beryllium-copper (BeCu) body. We chose Daphne Oil 7575 as the pressure-transmitting medium and a piece of lead to monitor the pressure. For the magnetometry mea-
The magnetic response was of the order of 1.5–3 $10^{-6}$ emu. The line represents a $T^5$ behavior of relaxation, with the exponent decreasing down to 3.6 at 1.47 GPa. To improve the readability of the plot, we are not indicating the location of $T_m$ and $T_c$ values, but we report them in Fig. 2. (c) Temperature dependence of $75$As NMR Knight-shift at three selected pressures. Uncertainties are of the order of the marker size. Inset: the $75$As NMR signal measured at 1.47 GPa, 7.06 T, and 25 K.

Fig. 1. (a) Temperature-dependence of magnetization for selected applied pressures, measured at $\mu_0 H = 2$ mT. (b) $75$As NMR $1/T_1(T)$ data at ambient- and at selected hydrostatic pressures. The inset shows the temperature dependence of $1/T_1(T)$ at ambient pressure, highlighting the maximum at $T_m$ and a kink at $T_c$. Recently, a similar but broader feature in $1/T_1(T)$ of ReSe was attributed to a pseudogap behavior.\textsuperscript{18} Note that for ThFeAsN $T_m \sim 1.2 T_c$, at least up to 1.47 GPa.

Fig. 2. Pressure dependence of $T_m$ and $T_c$ as determined via magnetometry and NMR measurements. $T_m$ (green diamonds) refers to the temperature where the electronic relaxation rates are maximized. The inset shows the temperature dependence of $1/T_1(T)$ at ambient pressure, highlighting the maximum at $T_m$ and a kink at $T_c$. Recently, a similar but broader feature in $1/T_1(T)$ of ReSe was attributed to a pseudogap behavior.\textsuperscript{18} Note that for ThFeAsN $T_m \sim 1.2 T_c$, at least up to 1.47 GPa.

measurements we used a tiny piece of ThFeAsN ($m \sim 40 \mu g$), whose magnetic response was of the order of 1.5–3 $\mu$emu. Because of the tiny signal, each measurement was performed with a background-subtraction procedure. Typical magnetization data at different applied pressures are shown in Fig. 5(a). The linearly decreasing trend of $T_c$, as determined from magnetization data, is shown in Fig. 2 (red squares) and agrees well with the prediction of a reduced $T_c$ at lower anion heights. A linear fit within the explored pressure range gives a slope of $\partial T_c/\partial p = -1.12 \pm 0.02$ K/GPa, similar to $-1.5$ K/GPa found in LiFeAs,\textsuperscript{19} another iron-based superconductor without doping.

NMR measurements under applied pressure. NMR measurements up to 1.47 GPa were performed using a BeCu piston-clamped high-pressure cell. The $75$As NMR investigations included line- and spin-lattice relaxation time ($T_1$) measurements in a magnetic field of 7.06 T.\textsuperscript{20} $T_1$ values measured at both peaks of the central-transition line via inversion recovery resulted identical. Pressure was monitored \textit{in situ} by using the nuclear quadrupolar resonance signal of $^{65}$Cu in Cu$_2$O.\textsuperscript{21}

A typical $75$As NMR line at 7.06 T is shown in the inset of Fig. 5(c). Due to the large quadrupole moment of $^{75}$As ($Q = 31.4 \text{ fm}^2$), we considered only the central component of the NMR spectrum, which exhibits a typical second-order powder pattern with dipolar broadening. For temperatures from 4 to 295 K and hydrostatic pressures from zero up to 1.47 GPa, the central-line transition exhibits minor changes in shape and position. The spectra were fitted using the quadrupolar exact software (QUEST),\textsuperscript{22} assuming no planar anisotropy ($\eta = 0$, as from experimental observations) and obtaining typical quadrupolar frequencies $\nu_Q$ of $\sim 5.6$ MHz. The full width at half maximum (not shown) is negligibly affected by temperature or pressure, thus confirming the absence\textsuperscript{3,23} of AFM long-range order, which would otherwise result in a remarkable broadening of the spectral lines starting at the onset of the transition.

Figure 5(c) shows the Knight shift $K_n(T) = (\gamma - \nu_Q)/\nu_Q$ values as a function of temperature. At all the applied pressures, $K_n(T)$ exhibits a linearly decreasing trend below $T_c$, compatible with an $s^\pm$-wave scenario.\textsuperscript{12} In fact, $K_n(T) \sim \text{Re} \chi''(q = 0, \omega \rightarrow 0)$, i.e., in the uniform susceptibility limit ($q = 0$), the inter-band scattering is suppressed and the Knight-shift value includes only the independent contributions from the hole- and electron bands.\textsuperscript{12,24} In the clean limit, this implies an exponential temperature-dependence for $K_n(T)$ in the $s^\pm$-wave case. However, as confirmed by magnetization data, our sample is not free of impurities. As reported in the literature,\textsuperscript{24–27} impurity self-energies form resonance states inside the SC gap and, thereby, affect the functional form of $K_n(T)$. The results of these calculations are compatible with the linear trend we observe. From the Knight-shift perspective, a dirty $s^\pm$-wave superconductor exhibits the features of a clean $d$-wave SC, but the latter inter-
We observe that pressure reduces the distance between
the peaks. This implies a slight symmetry enhancement
upon increasing pressure, resulting in $eq = V_{zz} = 2I(2I - 1)h\nu_0/(3eQ)$. Here $V_{zz}$ is obtained from the qua-
rupole splitting frequency via

$$\Delta f = \frac{29}{16} \frac{I(I + 1) - \frac{3}{2}}{\nu_0}.$$

We find a power-law behavior of $1/T$ in monotonously by
$1.45 \pm 0.1 K / \text{GPa}$, i.e., virtually with the
same slope as $T_{c2}$. Although we
confirmed the earlier findings$^{28}$ and were used as reference
to analyze the high-pressure data. The muon fraction stop-
ing in the pressure cell ($f_{\text{cell}} = 60\%$) was determined by
fitting a zero-field (ZF) spectrum with the cell relaxation
rate fixed to the GPS value, hence leaving the muon
fraction fixed to the GPS value, $T_{c1}$, and
the muon stopping fraction as the only free parameter. The absence of
significant changes with temperature in the ZF relaxation
rate of the sample, even at the highest pressure, rule out
a possible pressure-induced magnetic order. Thus, we focused
on the TF measurements in the SC region, carried
out at 70 mT. The data were analyzed using:

$$A(t)/A_0 = (1 - f_{\text{cell}})\cos(\gamma_B B_{\text{cell}} t + \phi) \exp(-\lambda_{\text{sc}} t - \alpha_{\text{cell}} t^2/2) + f_{\text{cell}}\cos(\gamma_B B_{\text{cell}} t + \phi) \exp(-\lambda_{\text{cell}} t - \sigma_{\text{cell}} t^2/2),$$

where $A_0$ is the initial asymmetry, $\gamma_B$ the muon gyromagnetic
ratio, $B$ the local field at the muon stopping site, $\phi$ the initial phase, and $\lambda$ and $\sigma$ the exponential and Gaussian
relaxation rates, whose subscript labels denote the parameters
for muons stopping in the sample and the cell, respecti-
vely. To ensure a robust fit, the change of $B_{\text{cell}}$ and $\sigma_{\text{cell}}$
was related to the field shift in the sample relative to $B_{\text{ext}}$:$^{28}$

$$B_{\text{cell}}(T) = B_{\text{ext}} + c_1 [B_{\text{ext}} - B_{\text{sc}}(T)],$$

$$\sigma_{\text{cell}}(T) = \sigma_{\text{cell}}^0 (T > T_c) + c_2 [B_{\text{ext}} - B_{\text{sc}}(T)].$$

$\lambda_{\text{sc}}$ and $\sigma_{\text{cell}}$ are proportionality constants. Since $\lambda_{\text{cell}}$
varies with temperature, its intrinsic $T$-dependence was
determined by requiring that the zero-pressure GPS measure-
ments reproduce the GPS results, from which we evalu-
ated an average penetration depth $\lambda_{\text{ab}}(0 \text{K}) = 255(1) \text{ nm}$. As can be seen from the temperature dependence of the

Transverse-field (TF) $\mu$SR measurements under high pres-
sure. The $\mu$SR investigations were performed at the GPS
(ambient pressure) and the GPD (high-pressure) spectrom-
eters of the Paul Scherrer Institute, Villigen. Since the high-
pressure measurements require a relatively large sample mass ($\sim 2 \text{ g}$), a new polycrystalline sample with $T_c = 27 \text{ K}$ was prepared. The lower $T_c$ is due to a different prepara-
tion protocol. The GPS measurements on the new batch
confirmed the earlier findings$^{28}$ and were used as reference
to analyze the high-pressure data. The muon fraction stop-
ing in the pressure cell ($f_{\text{cell}} = 60\%$) was determined by
fitting a zero-field (ZF) spectrum with the cell relaxation
rates fixed to their literature values$^{27}$ and the sample relaxation rate fixed to the GPS value, hence leaving the muon
stopping fraction as the only free parameter. The absence of
significant changes with temperature in the ZF relaxation
rate of the sample, even at the highest pressure, rule out
a possible pressure-induced magnetic order. Thus, we focused
on the TF measurements in the SC region, carried
out at 70 mT. The data were analyzed using:

$$A(t)/A_0 = (1 - f_{\text{cell}})\cos(\gamma_B B_{\text{cell}} t + \phi) \exp(-\lambda_{\text{sc}} t - \alpha_{\text{cell}} t^2/2) + f_{\text{cell}}\cos(\gamma_B B_{\text{cell}} t + \phi) \exp(-\lambda_{\text{cell}} t - \sigma_{\text{cell}} t^2/2),$$

where $A_0$ is the initial asymmetry, $\gamma_B$ the muon gyromagnetic
ratio, $B$ the local field at the muon stopping site, $\phi$ the initial phase, and $\lambda$ and $\sigma$ the exponential and Gaussian
relaxation rates, whose subscript labels denote the parameters
for muons stopping in the sample and the cell, respecti-
vely. To ensure a robust fit, the change of $B_{\text{cell}}$ and $\sigma_{\text{cell}}$
was related to the field shift in the sample relative to $B_{\text{ext}}$:

$$B_{\text{cell}}(T) = B_{\text{ext}} + c_1 [B_{\text{ext}} - B_{\text{sc}}(T)],$$

$$\sigma_{\text{cell}}(T) = \sigma_{\text{cell}}^0 (T > T_c) + c_2 [B_{\text{ext}} - B_{\text{sc}}(T)].$$

where $c_1$ and $c_2$ are proportionality constants. Since $\lambda_{\text{cell}}$
varies with temperature, its intrinsic $T$-dependence was
determined by requiring that the zero-pressure GPS measure-
ments reproduce the GPS results, from which we evalu-
ated an average penetration depth $\lambda_{\text{ab}}(0 \text{K}) = 255(1) \text{ nm}$. As can be seen from the temperature dependence of the
Upon increasing pressure, the latter being closer to ab initio we resorted to the Vienna explanation. As the key reason to prevent AFM order is confirmed by both NMR and zero-field even at the highest pressures suggests that the reduction of 

\[ \text{superconducting } T_c \] 

at 0.06(1) GPa to 3.8(1) meV at 2.48(2) GPa 

implies a reduction of the gap value is suppressed faster than \( \frac{\partial T_c}{\partial p} = -1.12(2) \text{ K/GPa} \) and weakens the pairing interaction, as measured by the ratio \( \frac{\Delta(0)}{k_B T_c} \). Interestingly, \( T_m(p) \) too is reduced by pressure at the same rate of \( T_c \), confirming that magnetic excitations which reflect AFM spin fluctuations, while competing with superconductivity, play an essential role in the pairing process. Finally, our experimental data and DFT calculations indicate an \( s^\pm \) SC order parameter independent of pressure and suggest that intrinsic disorder plays a key role in suppressing antiferromagnetism in ThFeAsN.

**Conclusion.** In ThFeAsN, the Knight shift \( K(T) \), the spin-lattice relaxation times \( T_1(T) \), and the London penetration depth \( \lambda(T) \) indicate that pressure reduces \( T_c \) \( \frac{\partial T_c}{\partial p} = -1.12(2) \text{ K/GPa} \) and weakens the pairing interaction, as measured by the ratio \( \frac{\Delta(0)}{k_B T_c} \). Interestingly, \( T_m(p) \) too is reduced by pressure at the same rate of \( T_c \), confirming that magnetic excitations which reflect AFM spin fluctuations, while competing with superconductivity, play an essential role in the pairing process. Finally, our experimental data and DFT calculations indicate an \( s^\pm \) SC order parameter independent of pressure and suggest that intrinsic disorder plays a key role in suppressing antiferromagnetism in ThFeAsN.

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$^{77}$As, with spin $I = 3/2$ and Larmor frequency $v_L = 51.523$ MHz at 7.06 T, was chosen because it occupies a single site and is sensitive to the structural- and electronic variation in the FeAs layers under pressure. The $^{77}$As NMR spectra were obtained via fast Fourier transformation of spin-echo signals generated by $\pi/2-\pi$ rf pulses of 2 and 4 $\mu$s, respectively, with recycle delays ranging from 0.1 s at room temperature up to 6 s at 5 K and echo times of 50 $\mu$s. Given the long rf-pulse length, frequency sweeps in 40-kHz steps were used to cover the spectrum central transition (~1 MHz wide).

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We recall that the transport and magnetic properties of ThFeAsN are similar to those of LaFeAsO$_{1-\delta}$, which indicates that the absence of a long-range magnetic order in the nominally undoped ThFeAsN can be due to intrinsic disorder. It is known that the long-range antiferromagnetic order in the iron-based superconductors can be even destroyed by nonmagnetic impurities, despite the electron-band structure remaining unchanged with respect to the undoped case.

The resulting phase diagram is similar to that obtained by introducing extra holes or electrons in the FeAs layers. Theoretically, this can be understood as a result of the stronger effect of nonmagnetic impurities on the AFM order than on the multiband $s^\pm$-wave superconductivity. While both the intra- and the interband impurity scattering are destructive for the long-range AFM order, only the interband scattering is pair-breaking for an $s^\pm$ superconducting state.

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I. SUPPLEMENTAL MATERIAL

We include the accurate description of the methods used to determine the values of $T_s$ and $T_m$ from the magnetization (see Fig. 1a) and the NMR data (see Fig. 1b). The resulting metadata are plotted as a function of pressure in Fig. 2 of the manuscript.

$T_c$ was evaluated from the magnetization data at the onset value, resulting from the crossing of two lines, i.e. the linear fit of the neighboring regions below ($0.8 < T/T_c < 1$) and above ($1 < T/T_c < 1.2$) the critical temperature, respectively at each applied pressure.

In the case of the NMR data, we plotted (see Fig. 1 of the Supplemental Material) the $T$-derivative of $1/T_1 T$ and identified $T_s$ as maximum [inflection point in $1/T_1 (T(T))$] and $T_m$ as zero of the function [maximum in $1/T_1 (T(T))$].

Fig. 5. From the $^{75}$As NMR $T_1$ values, we show the $T$-derivative of the $1/T_1 (T)$ curve for the four applied hydrostatic pressures. The decreasing trend for both $T_s$ and $T_m$ values upon increasing pressure is highlighted by two horizontal arrows. The dotted lines are a guide to the eye. Since the datasets are noisy, we empirically enhanced the accuracy in estimating the $T_s$ and $T_m$ values, by fitting these data with shape-preserving splines.