

## High-pressure neutron diffraction at ultra-low temperature

Y. Wu<sup>1,\*</sup>, T. E. Sherline<sup>1</sup>, J. J. Molaison<sup>1</sup>, A. M. dos Santos<sup>1</sup>, H. D. Zhou<sup>2</sup>, H. B. Cao<sup>1</sup>, J. J. Pierce<sup>1</sup>, and B. Haberl<sup>1,†</sup>

<sup>1</sup>Neutron Scattering Division, Neutron Sciences Directorate, *Oak Ridge National Laboratory*, Oak Ridge, Tennessee 37830, USA

<sup>2</sup>Department of Physics and Astronomy, *University of Tennessee*, Knoxville, Tennessee 37996, USA



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Neutron diffraction is a powerful tool in the study of quantum materials and associated emergent phenomena as it allows for direct detection of magnetic structure even at ultra-low temperatures. As pressure can drive systems across quantum critical points and tune emergent behaviors at the quantum scale, coupling neutron diffraction with extreme conditions of high pressure at (ultra-)low temperatures has been a key research target over decades. This has proven challenging as magnetic neutron diffraction typically requires large sample volumes due to flux limitations, whereas high-pressure studies require small sample volumes due to pressure limitations. Combining these two contradictory requirements poses many difficulties, but also brings opportunities in research and development. Here, we address these shortcomings through the use of a diamond anvil cell equipped with polycrystalline anvils with relatively large sample volumes coupled with the high neutron flux made available by the high-pressure diffractometer of the Spallation Neutron Source. This particular diamond anvil cell configuration has previously proven useful for single-crystal samples and is here coupled with the ultra-low temperatures of a dilution refrigerator for the first time. Our study on a single crystal of  $\text{Yb}_2\text{O}_3$  successfully collected magnetic neutron diffraction data at 5.3 GPa below 200 mK inside a dilution refrigerator. We have performed a detailed analysis of the magnetic diffraction data to investigate the magnetic structure under pressure and observed a subtle shift in the transition temperature. This development will open the door for future studies on magnetic materials with these much-desired coupled extremes of high pressure and ultra-low temperatures.

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### I. INTRODUCTION

Pressure is a widely applied clean “tuning” parameter for manipulating properties of materials and probing distinctive characteristics of quantum phenomena. Recent investigations have reported a number of pressure-induced exotic phases in quantum systems, including quantum spin liquid states in pyrochlore magnets, plaquette phases in Shastry-Sutherland lattice, pressure-induced superconductivity, and unconventional quantum criticality in 1D chains [1–5]. Neutron scattering under the extreme conditions of high pressure and (ultra-)low temperature is essential to illustrate the structure and magnetic interactions under pressure, as these significantly contribute to the underlying mechanisms in those quantum systems.

While high-pressure studies coupled with “standard” low temperatures to 5 K are routinely offered at neutron scattering facilities, matters become particularly challenging when coupling high-pressure techniques with ultra-low temperatures

(ULTs). High-pressure neutron scattering techniques have made much progress and achieved even Mega bar high pressures with diamond anvil cells (DACs), however, this usually requires high-precision alignment with respect to the beam and/or very large presses for the requested high forces. Both these scenarios are not conducive to combination with ULT environments. Consequently, across the entire suite of neutron facilities, many high-pressure neutron experiments at ULT continue to focus on piston-cylinder-type clamp cells, e.g., neutron diffraction studies at temperatures below 1 K with clamp cells (e.g., Refs. [2,6,7]) or even as low as 50 mK (e.g., Ref. [3]). The maximum pressures in such neutron piston-cylinder cells (mainly clamp cells) are, however, typically limited to 2–3 GPa.

Thus, concerted efforts are underway to maximize the high pressures possible at (ultra-)low temperatures for neutron diffraction. A key example is the demonstration of neutron diffraction at 22.6 GPa and 1.79 K in a Paris-Edinburgh cell achieved at the Institute Laue-Langevin, France [8]. The Paris-Edinburgh cell in an  $\sim 1.5$  K cryostat is available in their user program, and experimentation toward further cooling in a dilution refrigerator is underway. Another example is hybrid-anvil cells developed for low temperature and high magnetic fields [9], which are routinely used up to  $\sim 5$ –8 GPa at 1.5 K [10,11] at the WISH beamline of the ISIS Neutron and Muon Source, UK. Finally, a breakthrough experiment at the Laboratoire Léon Brillouin (LLB), France, observed the crystallization of a spin liquid at 1.4 K and pressures up to 9 GPa in a sapphire anvil “dilution” cell [12].

\*Contact author: wuy1@ornl.gov

†Present address: Department of Materials Physics, Research School of Physics, Australian National University, Canberra, ACT 0200, Australia.

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Much effort is spent on further increasing capabilities for high pressure coupled with ULTs, specifically for higher pressures up to  $\sim 10$  GPa at temperatures below 1 K. For example, efforts were underway at ISIS to combine their compact pressure cell capable of close to 10 GPa and pressure monitoring via ruby fluorescence [13] with ULT experimentation. Offline tests in an E18 Vericold dilution refrigerator on an adapted cell achieved 70 mK while confirming pressure performance using the ruby fluorescence method, although adaptation to a beamline has not been completed [14,15]. A remarkable breakout experiment led by I. Goncharenko on  $\text{Tb}_2\text{Sn}_2\text{O}_7$  performed neutron powder diffraction at 4.6 GPa and 60 mK by placing a sapphire anvil cell inside a dilution refrigerator [16,17]. Unfortunately, it was not possible to continue this work, and this capability is not available to the user community.

At Oak Ridge National Laboratory (ORNL), efforts have focused on the use of clamped DACs equipped with polycrystalline diamond anvils. These cells allow for relatively large single-crystal samples and are equipped with a spring specifically designed for pressure stability across temperature excursions [18,19]. Here, we adapt these cells to the ULT enabled through dilution refrigeration. Neutron diffraction data in DAC were collected on the dedicated high-pressure diffractometer of the Spallation Neutron Source (SNS), the Spallation Neutrons and Pressure (SNAP) beamline [20,21], and neutron diffraction at a maximum pressure of 5.3 GPa was achieved at 170 mK.

Under these conditions, the magnetic phase of the rare-earth compound  $\text{Yb}_2\text{O}_3$  was studied as test sample to demonstrate our capability. The ground magnetic state of the cubic C-type structure of  $\text{Yb}_2\text{O}_3$  has been reported as noncollinear antiferromagnetic with a Néel temperature of  $T_N = 2.3$  K at ambient pressure. An early neutron diffraction study [22] reported the best fit to the magnetic structure as a three-domain model, where the (111) and (110) directions exhibit different magnetic moments. In this system, the weak interactions between the anisotropic magnetic moments could act as a perturbation on top of its crystal field, while simultaneously also being strongly correlated with the crystal field. Thus, tuning the magnetic interaction is expected to lead to emergent phenomena and introduce frustrations. Further, the high-symmetry structure was also advantageous for this development.

Future work will expand on this capability, toward high-quality data with better signal-to-noise ratio at higher pressures (note that these DACs are capable of 10 GPa or more [18,19]) and enable *in situ* pressure controlling to further facilitate the study of emergent phenomena and dynamic interactions in quantum materials. Nonetheless, this current work opens the possibility of routine magnetic neutron diffraction at ULT in the 10 GPa range.

## II. EXPERIMENTAL METHODS

The  $\text{Yb}_2\text{O}_3$  single crystals were grown using the floating zone technique. The crystal structure of the sample was characterized with single-crystal X-ray diffraction on a Rigaku X-ray machine. The crystallinity of the samples was further confirmed using Laue diffraction measurements.

A  $\text{Yb}_2\text{O}_3$  sample was then measured at ambient pressure down to 1.2 K using the DEMAND instrument [23] of the High Flux Isotope Reactor (HFIR), ORNL, configured with a large-area 2D Anger camera and  $\langle 110 \rangle$  silicon multiwafer monochromator.

Samples suitable for the DAC were prepared on  $\langle 111 \rangle$  plane for high-pressure neutron diffraction. The crystal was oriented with Laue diffraction prior to cutting it with a diamond saw. It was then polished for maximum size fitting into the gasket chamber accounting for the latter's shrinkage upon compression. The final crystal was approximately square-shaped with dimensions of  $\sim 0.75$  mm  $\times$  0.75 mm and a height of 0.15 mm.

For the high-pressure experiment, a CuBe clamped neutron DAC [19] was prepared. The design of this DAC is optimized for pressure stability at low temperatures through the incorporation of a thick point-loaded disk spring [18,19]. The particular CuBe spring used here undergoes elastic deformation only while loads remain below 9 metric ton and thus performs as designed in this regime [19]. Testing of DACs with this spring design at various beamlines at the SNS and HFIR upon cooling to  $\sim 5$  K did not reveal any issues with drift and pressures remained constant. Given the pressure stability to  $\sim 5$  K, we surmise continued pressure stability into the millikelvin regime.

The DAC used here was equipped with 10 mm tall conical anvils made from Versimax, a polycrystalline diamond sintered with SiC (Hyperion Materials & Technology, Worthington, OH, USA). The anvils were cut into 3 mm culets. A premachined gasket made from 15-5 PH stainless steel [24] was further preindented to a final thickness of 490  $\mu\text{m}$  and a chamber diameter of 1.45 mm. The gasket was heat treated at 482  $^\circ\text{C}$  for  $\geq 4$  h to achieve full hardness.

The prepared DAC with the crystal in the gasket chamber was placed on SNS's SNAP beamline. The beamline was operated using its focusing guide and two detector banks centered at  $2\theta = -50^\circ$  and  $2\theta = 105^\circ$  for large  $Q$  coverage. It was initially configured with choppers set to 60 Hz and a center wavelength of  $\lambda = 2.1$   $\text{\AA}$  for a wavelength band of 0.5–3.7  $\text{\AA}$ . For final measurements, this was changed to 60 Hz and 3.9  $\text{\AA}$  for a wavelength band of 2.3–5.5  $\text{\AA}$  for increased access to low  $Q$ .

On the beamline, initial alignment of the cell and crystal was performed using a decoupled hBN collimator with a 1-mm-diameter aperture. The  $\text{Yb}_2\text{O}_3$  crystal in the DAC was oriented at a specific angle in the (*HHL*) plane so that the posts of the DAC did not obscure the detector banks. Deuterated glycerol (D8, 99%, Cambridge Isotope Laboratories, Inc., Andover, MA, USA) was added as pressure-transmitting medium (PTM) [19,25]. Glycerol was chosen here as it does not pose any issues with leakage and presents reasonable hydrostaticity. While it undergoes a glass transition at 5.5 GPa [25], it does not develop a pressure gradient across the large culet until  $\sim 9$  GPa [19]. Crystal alignment was confirmed after sealing with 1.5 metric ton of applied load.

A final load of 5 metric ton was applied for an estimated sample pressure of  $\sim 4.5$  GPa as per a previously measured pressure-load (PL) curve [18]. The sample was again checked on the beamline. Thereafter, the cell was equipped with a custom-designed incident beam collimator as shown in Fig. 1.

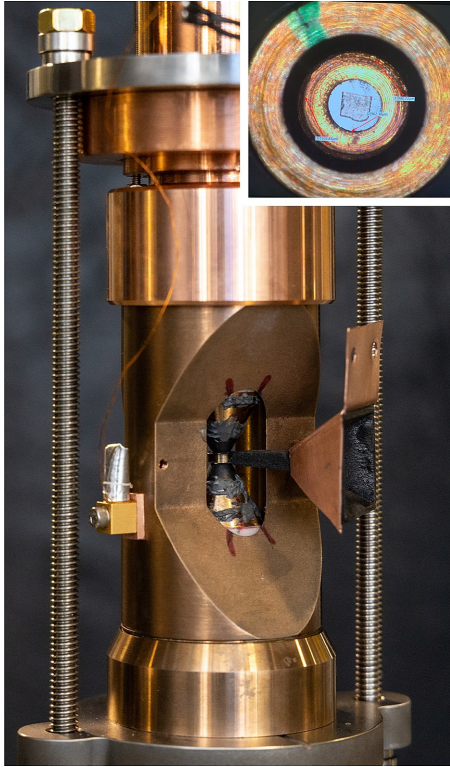


FIG. 1. Photograph of the diamond anvil cell mounted onto the dilution refrigerator. The 3D-printed collimator is directly attached to the cell through a small screw bolted into the right-hand cell post (not visible here) so that the end of the collimator funnel directly points at the gasket/sample assembly. The sensor used to track the temperature of the cell body is seen where it is screwed onto the left post. The inset shows a photograph of the single-crystal sample inside the gasket prior to closing the cell.

The collimator was 3D printed from  $B_4C$  and infiltrated with cyanoacrylate (superglue) [26,27]. It was designed with a final opening on the sample/gasket side of  $0.6 \times 1.54 \text{ mm}^2$ . It was confirmed that the diffraction peaks remained visible and that the orientation remained correct.

In the next step, the DAC was clamped to the mixing chamber of a custom Oxford Instruments TritonNANO dilution refrigerator with a force of roughly 1 metric ton to provide adequate thermal contact. The thermal performance was determined previously during offline testing. During these offline tests, the DAC was mounted to the TritonNANO dilution refrigerator in the same manner, with a thermometer mounted at the sample position of the DAC (i.e., between the two anvils) with an applied force of approximately half a metric ton. A larger force was precluded by the yielding of the copper lug to which the thermometer was mounted to ensure good and accurate temperature sensing. In these offline tests, monitored with a Lakeshore 370 resistance bridge, the base temperature of the mixing chamber was 58 mK, while the DAC body reached 60 mK and the sample thermometer between the anvils reached 83 mK, a difference of 23 mK.

The success of the present online experiment is derived from lessons learned from an initial experiment in which the background from the aluminum thermal heat shields and outer vacuum can had precluded observation of the crystal. In the

present experiment, windows were cut into the three heat shields before covering them with 0.016-mm-thick aluminum foil, thus greatly reducing the background. The modification of the thermal shields was shown in offline testing to produce a modest increase of 1 mK in the base temperature of the dilution refrigerator and 2 mK in that of the DAC.

An additional lesson from the initial experiment concerned beam heating. Whereas the base temperature of the DAC inside the dilution refrigerator in the absence of the beam was 58 mK, the DAC temperature increased to 191 mK when the cell/sample assembly was exposed to the beam. This initial experiment was performed with a larger 3D-printed  $B_4C$  collimator designed for single-crystal experiments in this DAC [26], which was directly attached to the DAC body. It was subsequently replaced with the smaller collimator used here that is attached via an insulating Vespel mount.

The DAC position was later fine-tuned by scanning the hexapod in the presence of the neutron beam. While the TritonNANO dilution refrigerator cooled on the beamline, data were continuously collected and alignment using the hexapod was refined iteratively. A base temperature of 170 mK was achieved in the presence of the beam, as determined from the temperature measurement on the DAC body. During the offline test, we had found a temperature difference of  $\sim 20$  mK between this position on the DAC body and the position between the anvils. As the real pressure experiment precluded renewed measurement between the anvils, we quote this DAC body temperature throughout the manuscript as our base temperature. Overall, the temperature of the sample would be no more than 20 mK above the quoted base temperature of 170 mK, thus below 200 mK.

### III. RESULTS AND DISCUSSION

Diffraction data were collected from the  $Yb_2O_3$  sample inside the DAC during different experimental conditions, and the data obtained after sealing the DAC (1.5 ton) collected with the movable hBN collimator are shown in Fig. 2. These single-crystal diffraction data were calibrated with silicon standard data and normalized with vanadium profile when analyzing with the MantidWorkbench [28]. At ambient pressure as well as at the sealing load of 1.5 ton, the nuclear peaks were straightforward to process using the single-crystal integration algorithms in Mantid. Next, a crystal structure refinement was performed with Fullprof [29]. The refinement quality of the structure is reasonable with  $R_{\text{factor}} = 7.754$  and  $\chi^2 = 35.32$  obtained from using 18 unique nuclear peaks within the accessible scattering planes limited by the narrow opening of the funnel collimator. The refinement quality is shown in Fig. 2(b).

At 5 ton of applied load, strain led to significant broadening of the peaks, making them more difficult to discern and process on the detector. This made the integrated intensities less reliable. We attribute this to possible bridging of the crystal between anvils and/or the sample chamber edges. Yet, several strong nuclear peaks can still be observed and the positions of these peaks in  $d$ -spacing are unambiguous. These were used to determine the pressure as detailed in Appendix A. This yields  $P = 5.34 \pm 0.80 \text{ GPa}$  as per the experimental equation of state by Meyer *et al.* [30]. The somewhat higher pressure

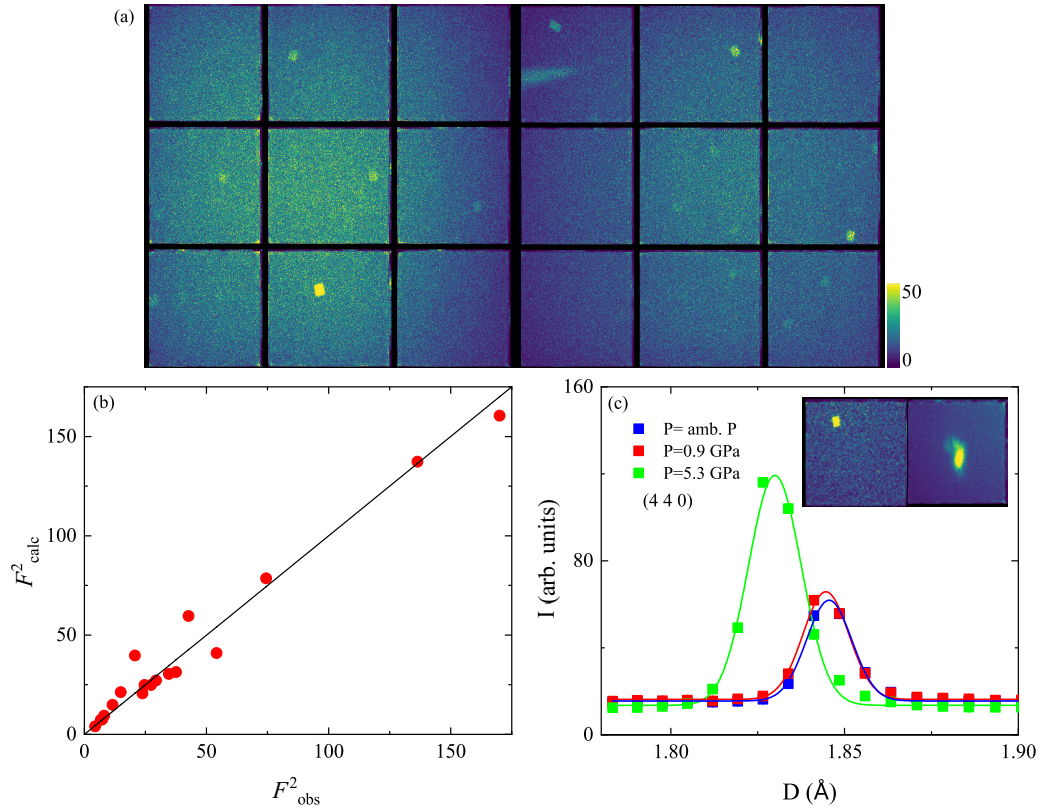


FIG. 2. (a) Nuclear peaks of the Yb<sub>2</sub>O<sub>3</sub> sample inside the DAC seen on the 2D detector at room temperature and after sealing at 1.5 ton. (b)  $F^2_{\text{calc}}$  vs  $F^2_{\text{obs}}$  from the nuclear refinement of the collected peaks. (c) Peak width of the (440) peak at ambient pressure, 1.5 ton load, and 5 ton load, with insets showing images of the (440) peaks on the 2D detector bank at the two loads. All data were collected with the center wavelength of 2.1 Å.

obtained here compared to the previous PL curve is attributed to better packing with the liquid PTM.

Details of the analysis of the (440) peak are shown in Fig. 2(c). The crystal appeared to have slightly rotated during pressurization, resulting in a change in diffraction conditions and an increase in peak intensities. We found that the full width at half maximum (FWHM) of the integrated 1D peaks versus  $d$ -spacing only changed slightly. Specifically, after fitting with a Gaussian, we obtained an FWHM of 0.0153(6), 0.0155(5), and 0.0179(7) Å at the three pressure points. This illustrates that sufficient hydrostaticity was maintained upon room temperature compression by our use of glycerol as PTM. Nonetheless, the peak shapes change on pressure increase, as illustrated in the insets shown in this figure.

Structure refinement of the nuclear structure was attempted at this maximum pressure. The refinement returns large  $\chi^2$  values even when the intensity consistency is acceptable ( $\chi^2 = 3.39$ ) within a selected integration area around the single-crystal peak. Thus, this high-pressure structure refinement is of insufficient quantitative merit. Yet, judging by the existence and relative intensities of a few visible peaks such as the (222), (440), and (226) peaks, the structure at 5.3 GPa appears to still be in the ambient pressure structure, consistent with previous reports in the literature [30], although a slight distortion toward the high-pressure A-type or B-type phase cannot be ruled out.

Once in the dilution refrigerator, the signal was further impacted by the change in collimator setup and the addition

of heat shielding. As a result, several background powder features obscured some of the nuclear peaks. This is illustrated in Fig. 3, where the 2D diffraction data of the cell at 5.3 GPa outside of the refrigerator and within the refrigerator and the low-temperature diffraction results of the right-side panel are shown. Adjusting the instrument collimation outside of the refrigerator only improved the background in part of the detector regions. Details on this procedure are given in Appendix B. Full single-crystal structure refinement of the nuclear peaks was difficult with the limited discernible nuclear peaks; we can only conclude that there should be no structure transition down to base temperature. Investigation of the nuclear peaks showed, however, that no discernible changes in peak width or shape occurred on cooling. This suggests that no significant changes in hydrostatic environment were caused by the freezing of the PTM upon cooling.

Once the temperature decreases below  $T_N$ , the antiferromagnetic (AFM) peaks appear with a wave vector (001), consistent with literature [22], observing the magnetic peaks at  $h + k + l = 2n + 1$ . Here, the (111) peak was the strongest magnetic peak observed in the accessible scattering plane. Figure 4(a) contrasts the presence of this magnetic peak at 170 mK, well below  $T_N$ , with its absence at 3.5 K, above  $T_N$ . The temperature-dependent peak intensity (order parameter) of this (111) peak from 170 mK to 4.5 K is shown in Fig. 4(b). The integrated intensity was obtained by fitting Gaussians to the integrated 1D peaks using algorithms available in MantidWorkbench, whereby the FWHM limit was restricted to

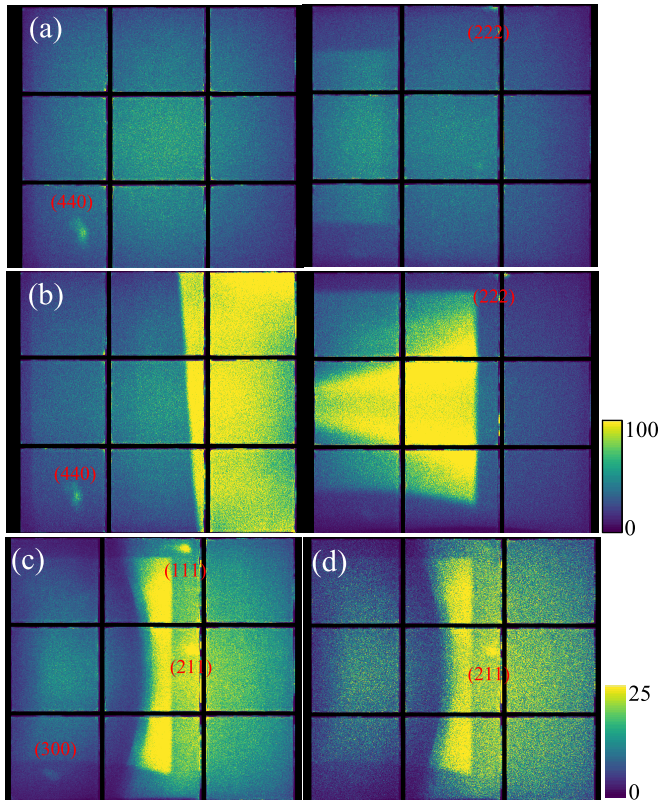


FIG. 3. 2D detector panel showing the diffraction data at 5.3 GPa at room temperature (a) outside of and (b) within the TritonNano. Both datasets were obtained with the 3D-printed funnel collimator in place. The collection was performed at the center wavelength of 2.1 Å. (c) The low-angle detector panel at  $T = 170$  mK, where magnetic peaks (111) and (300) are observed. (d) The low-angle detector panel at 3.5 K.

0.1 Å based on the magnetic peak profile. The temperature dependence of the (111) peak intensity yields  $T_N \sim 2.5$  K. We compare this to a previous ambient pressure measurement on DEMAND, which, in good agreement with literature [22], yielded a  $T_N$  of the (111) peak of 2 K. Thus, pressure leads to a slight increase in  $T_N$  by increasing the exchange coupling. No additional transitions were observed.

The previous magnetic structure study of  $\text{Yb}_2\text{O}_3$  [22] reports a three-domain AFM model, where the  $C_3$  moments are along  $\langle 111 \rangle$  directions and the  $C_2$  moments along  $\langle 111 \rangle$  directions. This same behavior is seen for our own sample material since magnetic structure refinement performed on the ambient pressure data collected at DEMAND also supports the three-domain AFM model (not shown). For the high-pressure data, following the background subtraction, several magnetic peaks were identified within the accessible scattering planes. We attempted to perform a magnetic structure refinement using these limited magnetic peaks, assuming that the magnetic structure remained detwinned under pressure.

Following the symmetry analysis using the Maxmagn module on the Bilbao crystallographic server [31], there are four maximum subgroups, belonging to  $P_1a - 3$  and  $P_1cca$  symmetry groups. Each symmetry group allows for two different distributions of the magnetic moments, one where both

the (111) and (110) magnetic moments exist and one where only the (110) moments dominate. Fitting for our pressure data can converge for both of the two  $P_1a - 3$  subgroups. The two solutions are shown in Fig. 4. The fitting quality of Model1 and Model2 is very close as shown in Fig. 4(c); Model2 fits only slightly better than Model1 but both with large error bars. According to the refinement, we could conclude that the magnetic structure under pressure agrees with the early results [22] as Model2 that contains both (111) and (110) contributions. However, to unambiguously distinguish between the two  $P_1a - 3$  models for the magnetic structure of  $\text{Yb}_2\text{O}_3$ , a more extensive coverage of magnetic scattering is needed. Nevertheless, the current analysis shows that the experimental condition can enable the determination of simple magnetic structures in quantum systems.

In all, there is subtle change in the magnetic phase of  $\text{Yb}_2\text{O}_3$  upon pressure increase at 5.3 GPa. The order temperature is shifted to a slightly higher temperature at high pressure. The under-pressure magnetic space group agrees with the previous reported ambient pressure structure. However, further  $P$ - $T$  points as well as the detection of additional magnetic peaks through collection in different orientations are needed to yield further insight into magnetic structure determination.

We have thus been able to successfully demonstrate the measurement of magnetic structure at pressures of  $\sim 5$  GPa and below 200 mK by employing our clamped neutron DAC inside a dilution refrigerator. It is useful to compare this experiment with the only past neutron diffraction experiment conducted in a DAC inside a dilution refrigerator, the groundbreaking experiment on  $\text{Tb}_2\text{Sn}_2\text{O}_7$  by [16,17]. There, the “dilution cell” designed by Goncharenko [32] was used, a CuBe DAC typically also equipped with a spring for temperature stability. It was smaller in size than the ORNL DAC used here, 30 mm diameter  $\times$  85 mm [32] versus our 50 mm diameter  $\times$  97 mm [18]; both allowed for a large scattering aperture. Like in our experiment, the cell body was shielded, but using Cd [32] in contrast to our  $\text{B}_4\text{C}$ . The sample in Goncharenko’s DAC was closely packed powder without pressure medium to ensure sufficient sample size. This introduces a strong nonhydrostatic component, which is bound to affect phase behaviors. The use of a powder sample over a single crystal simplifies experimental setup, but can occlude details only obtainable via single-crystal data analysis.

Further, it is interesting to note that the reported base temperature of 60 mK in Goncharenko’s experiment is the same as the base temperature on the DAC body in our offline test. While the larger size of our cell could lead to increased barriers in DAC-refrigerator connection and cooling performance, the same base temperature suggests that those were overcome with our careful treatments. The marked difference during the two online experiments indicates that the past experiments at LLB suffered less from beam heating effects. Clearly increasing neutron fluxes by orders of magnitude and the incidence of higher-energy neutrons as done at current and future spallation sources could give rise to additional effects that were previously unobserved, such as the beam heating we observe here. Future experiment planning relying on high neutron fluxes will thus have to take these effects into account for experimental planning.

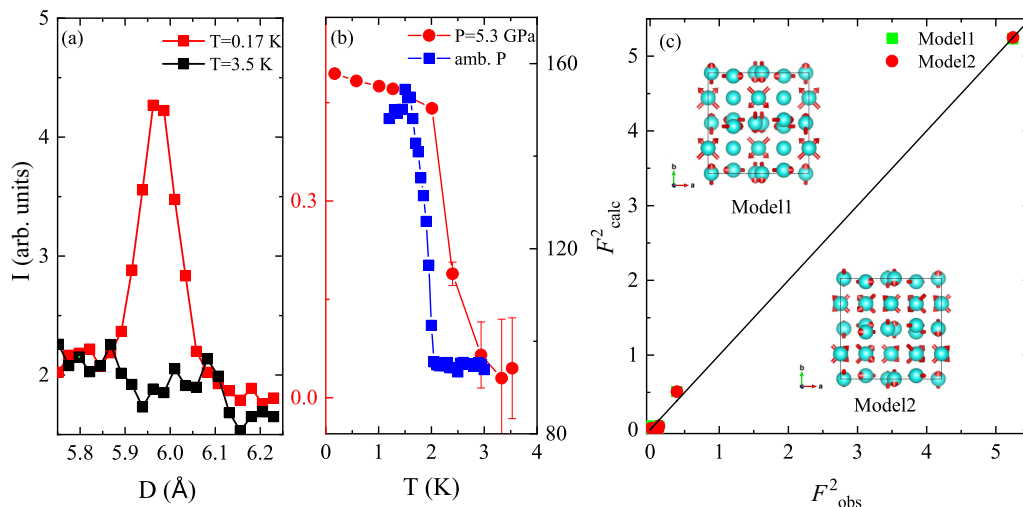


FIG. 4. (a) Magnetic (111) peak of  $\text{Yb}_2\text{O}_3$  at 170 mK and 3.5 K collected on warming. (b) Temperature dependence of the peak intensity of the  $\text{Yb}_2\text{O}_3$  (111) peak at 5.3 GPa upon warming from 170 mK to 4.5 K (red) compared to the same order parameter obtained at ambient pressure on DEMAND. (c)  $F^2_{\text{calc}}$  vs  $F^2_{\text{obs}}$  of the magnetic structure Model1 (with only  $\langle 110 \rangle$  moment contribution) and Model2 (with both  $\langle 111 \rangle$  and  $\langle 110 \rangle$  moment contributions) refinement with peaks collected at  $T = 170$  mK, 5.3 GPa. Insets are the refined magnetic structure Model1 and Model2 of  $\text{Yb}_2\text{O}_3$  at 5.3 GPa for data collected at 170 mK.

While posing challenges, the increasing neutron flux also offers many opportunities, such as the ability to conduct neutron spectroscopy on small samples, including samples contained within neutron DACs. In fact, the large-angle clamped DAC used here was designed for the SNS's indirect geometry VISION neutron spectrometer and a successful proof of principle on hydrogenated powder samples has been demonstrated [18]. Yet, experimentation with single-crystal samples in a DAC within a dilution refrigerator greatly benefits from precise instrument alignment capabilities as well as tightly focused neutron beams. While most current neutron spectrometers are not equipped with these capabilities, future instruments are designed with such applications in mind. Examples can be found at ORNL's Second Target Station, where much focus has been directed on enabling multi-extremes [33], including the use of DACs for spectroscopy through the specific addition of necessary alignment capabilities. While these instruments have yet to be built, the future ability to use neutron spectroscopy will clearly open many opportunities in the study of quantum materials.

#### IV. CONCLUSION

The combination of high pressure at ultra-low temperatures is challenging for neutron diffraction since comparatively low neutron flux conditions prefer large sample volumes, whereas high pressures require small sample volumes. Past works have thus predominantly focused on the relatively large sample volumes afforded by cylindrical pressure cells such as gas cells or piston-cylinder cells. These are, however, limited in pressure to 2–3 GPa. Breaking out of this pressure regime to higher pressures at ultra-low temperatures typically requires opposed anvil cells.

At ORNL, we have previously developed a large-volume DAC equipped with polycrystalline diamond Versimax anvils

for high-pressure single-crystal diffraction studies on magnetic materials [18,19]. Here, we leverage this particular DAC for ultra-low temperatures through coupling it with a dilution refrigerator. Taking advantage of the high flux of the SNS's SNAP diffractometer allows for magnetic diffraction at pressures beyond the typical limitations. Specifically, on a single-crystal sample of  $\text{Yb}_2\text{O}_3$  we observe magnetic diffraction at 5.3 GPa and below 200 mK. The magnetic structure data exhibit a reasonable signal-to-noise ratio and could be used for magnetic structure refinement. The analysis suggests a subtle shift in the order parameter. Further experiments at higher pressure are expected to yield more insight into possible magnetic phase transitions.

This experiment represents the first successful study in the ultra-low temperature regime using the ORNL DAC and clearly demonstrates its ability for neutron diffraction at ultra-low temperatures and pressures above 2–3 GPa. While the current experiment was performed at 5.3 GPa, the cell and anvil configuration is capable of higher pressures [18,19]. Going forward, we continue working on further background suppression for higher signal-to-noise ratio in high-pressure neutron scattering experiments and on concepts for *in situ* pressure control inside the dilution refrigerator for better probing of dynamic interactions. Overall, the current capability, which will be available to researchers through ORNL's user program, opens the regime for neutron single-crystal diffraction studies at ultra-low temperature and up to 10 GPa pressure. Such advances in experimental capabilities can clearly stimulate emerging directions in the investigation of quantum materials with high pressure.

#### ACKNOWLEDGMENTS

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TABLE I. Measured  $d$ -spacings of individual single-crystal reflections (in Å) at ambient pressure, after sealing (1.5 ton) and at maximum pressure (5 ton). The peak positions were determined by a fit in MantidWorkbench for an accuracy of four digits and by directly reading off the spectrum for an accuracy of three digits in cases where fitting was failed.

| $hkl$ | 0 ton           | 1.5 ton         | 5 ton           |
|-------|-----------------|-----------------|-----------------|
| 211   | 4.2762 ± 0.0061 |                 |                 |
| 222   |                 |                 | 2.9982 ± 0.0020 |
| 332   | 2.225 ± 0.002   | 2.222 ± 0.002   |                 |
| 440   | 1.8449 ± 0.0003 | 1.8439 ± 0.0003 | 1.8302 ± 0.0009 |
| 611   | 1.694 ± 0.002   | 1.693 ± 0.002   | 1.677 ± 0.002   |
| 541   | 1.6104 ± 0.0006 | 1.6085 ± 0.0002 | 1.593 ± 0.002   |
| 622   | 1.5713 ± 0.0004 | 1.5704 ± 0.0003 | 1.558 ± 0.002   |
| 631   | 1.5377 ± 0.0004 | 1.5347 ± 0.0007 |                 |

laboration on collimator development. We also acknowledge Victor Fanelli and Tyler White for assistance during the installation of the dilution refrigerator. This research used resources at the Spallation Neutron Source and High Flux Isotope Neutron Reactor, DOE Office of Science User Facilities operated by the Oak Ridge National Laboratory (ORNL). The beam time was allocated to SNAP on proposal numbers IPTS-27209.1 and IPTS-29052.1. The DEMAND data are from IPTS-25620.1. The crystal growth performed at the University of Tennessee (H.D.Z.) was supported by the U.S. Department of Energy under Grant No. DE-SC0020254. This work has been partially supported by U.S. Department of Energy. ORNL is managed by UT-Battelle, LLC, under Contract No. DE-AC05-00OR22725 for the U.S. Department of Energy.

#### DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

#### APPENDIX A: PRESSURE DETERMINATION

The pressure at room temperature was determined based on the single-crystal diffraction data obtained on SNAP using the equation of state by Meyer *et al.* [30]. First, the  $d$ -spacings of reflections larger than 1.5 Å were determined after reducing the data to 1D in MantidWorkbench [28]. For the more prominent reflections, these peak positions were determined using the built-in peak fitting algorithm. Not all reflections could be fitted due to imperfect peak shapes and insufficient signal-to-noise ratio. These were then directly determined by the reduced intensity spectrum. The observed reflections for each load are summarized in Table I. Note that the sample had

TABLE II. Pressure determined based on the measured  $d$ -spacings shown above, together with the average lattice parameter  $a$ , the volume ratio  $V/V_0$ , and the pressure  $P$  reduced. See text for further details.

| Load (ton) | $a$ (Å)        | $V/V_0$         | $P$ (GPa)   |
|------------|----------------|-----------------|-------------|
| 0          | 10.440 ± 0.006 | 1               | 0           |
| 1.5        | 10.423 ± 0.004 | 0.9953 ± 0.0021 | 0.87 ± 0.40 |
| 5          | 10.347 ± 0.011 | 0.9736 ± 0.0035 | 5.34 ± 0.80 |

rotated slightly upon load increase from 1.5 to 5 ton resulting in the observation of a slightly different set of peaks.

Considering a cubic unit cell, the corresponding unit cell parameter was calculated from the peak  $d$ -spacings. Averaging the resulting parameters from all peaks yields lattice parameter  $a$  for each load as presented in Table II. The error is thereby the standard error across the different reflections. Using this average  $a$ , an average ratio of  $V/V_0$  together with the propagating error was calculated. From these, the corresponding pressure  $P$  together with the propagating error was calculated with  $B_0 = 181$  GPa,  $B'_0 = 7.3$ , and the Murnaghan equation of state as per Meyer *et al.* [30].

#### APPENDIX B: BACKGROUND SUBTRACTIONS

For the data inside the dilution refrigerator, all data are first processed by applying a 2D mask using the SNAPreduce algorithm in MantidWorkbench to mask out the regions on the 2D detector where no single-crystal peaks are present. The total intensity data in each detector module are integrated to 1D data depicted as intensity versus  $d$ -spacing. The 1D spectrum data are also calibrated with Si standard spectrum and the intensities are normalized with a vanadium sample measured at the same settings in the process. Overall, 15 spectra (labeled 0–14) are rendered from this process for the Yb<sub>2</sub>O<sub>3</sub> sample inside the dilution refrigerator. Peaks could be identified with their corresponding  $d$ -spacing as shown in Figs. 5(a)–5(c). To effectively identify and integrate the peaks, an additional background subtraction is performed in order to remove excessive background from the extreme environments. For the magnetic peaks, diffraction data with the same diffraction condition taken above  $T_N$  ( $T = 5$  K) was used as the background. For the nuclear peaks, interpolated curves from the data after stripping off the nuclear peaks was used as the background. These backgrounds are subtracted from the diffraction data at base temperature to extract peak intensities. The magnetic peaks' processing is illustrated in Fig. 5(d).

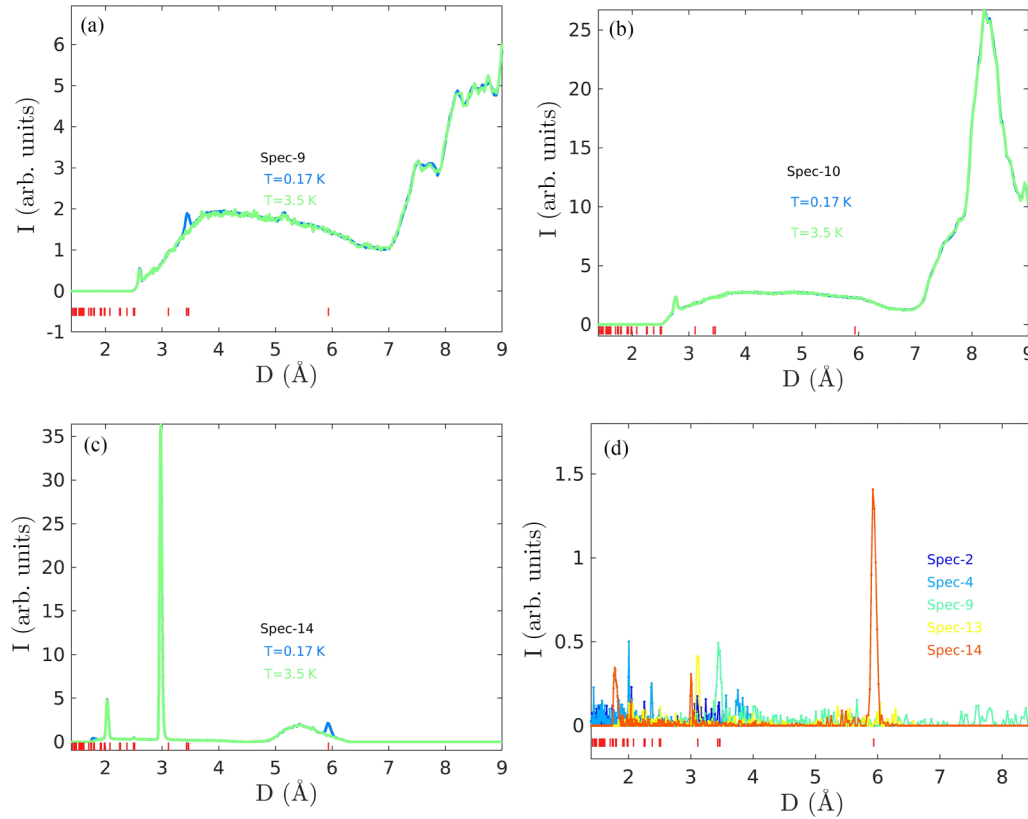


FIG. 5. Background subtraction used to detect the distinguishable magnetic peaks within the accessible scattering planes at  $T = 170$  mK,  $P = 5.3$  GPa: Panels (a)–(c) show example scans represented by spectra 9, 10, and 14 after reduction at  $T = 0.17$  K and  $T = 3.5$  K. Panel (d) shows the reduced 1D data at  $T = 0.17$  K following the background subtraction using the corresponding  $T = 3.5$  K scan. The red lines at the bottom of the figures indicate the expected magnetic peak positions of  $\text{Yb}_2\text{O}_3$ .

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