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Piezomagnetic switching and complex phase equilibria in uranium dioxide: X-ray diffraction in pulsed magnetic fields.

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Actinide materials that exhibit both strong spin–lattice coupling and electronic correlations are predicted to host various new emerging ground states. One prominent example is the existence of piezomagnetism and magneto-elastic memory effect in antiferromagnetic Mott-Hubbard insulator, uranium dioxide (UO₂). Piezomagnetism is a rare phenomenon observed in special classes of antiferromagnetic crystals and its microscopic nature is still

under debate. Here, we report X-ray diffraction studies in a high-precision back-reflection geometry under strong pulsed magnetic fields on oriented high quality UO_2 crystals. In the antiferromagnetic state a [888] Bragg diffraction peak (cube's body diagonal) follows the bulk magnetostriction that expands under magnetic fields. Upon reversal of the field, the expansion turns to contraction-making the magnetostriction odd under time reversal, before the [888] peak follows the switching effect and piezomagnetic “*butterfly*” behaviour, characteristic of two structures connected by time reversal symmetry. An unexpected splitting of the [888] peak in the magnetic fields is observed, indicating the presence of time-reversed (TR) domains of the $3\text{-}k$ structure and a complex magnetic-field-induced evolution of the microstructure. These studies open the door for a microscopic understanding of the piezomagnetism and magnetic coupling across strong magneto-elastic interactions.

Strong coupling between magnetism and lattice vibrations can lead to many emerging phenomena, as have been shown in unconventional superconductors, heavy-fermions, multiferroics and other new functional materials [1] [2] [3] [4] [5]. Due to strong spin-orbit coupling, correlated $5f$ -electron spin systems represent a perfect platform to scrutinize the phenomena related to spin-phonon interactions, especially when interacting with other degrees of freedom such as multipolar ordering or Jahn-Teller interactions [6,7]. An excellent example is uranium dioxide. This antiferromagnetic Mott-Hubbard insulator [8] is the main nuclear fuel and the most studied actinide material to date [9]. Its correlated ground state is characterized by a competition among non-collinear magnetic dipoles, electric quadrupoles, and dynamic Jahn-Teller distortions [10] [11] [12]. Recently, it has been shown that, due to the magnetic symmetry of the non-collinear $3\text{-}k$ antiferromagnetic order (shown in the inset of Figure 1a) and strong magneto-elastic coupling, UO_2 undergoes a trigonal distortion under magnetic field and becomes a piezomagnet with exceptionally large coercive characteristics [13]. In

piezomagnetic crystals, a magnetic moment can be induced by the application of physical stress [14] [15]. This phenomenon has captured attention in recent years as a mechanism that could be used, in combination with multiferroics and piezoelectrics (especially at the nanoscale), to achieve control of magnetism by electric fields [16]. Piezomagnetism is also utilized in geology where the so-called volcano-magnetic effect is used for monitoring volcanic activities [17] [18] [19]. Despite intensive work, the microscopic nature and crystallographic evidence of piezomagnetism are still elusive [20] [21].

Here we show, for the first time, a direct micro-structural probe of piezomagnetism using single-crystal X-ray diffraction. By using a high-resolution back-reflection geometry setup [see the supplementary material for details], we were able to resolve small changes in the [888] Bragg peak of UO_2 when the magnetic field is applied along the [111] direction, which corresponds to the longitudinal lattice response along the parallel cube diagonal of its crystal lattice. In the paramagnetic state (above 30.5 K), the [888] Bragg peak moves towards larger 2θ values (see methods) with the applied magnetic field, exhibiting negative magnetostriction. When the UO_2 crystal is cooled below the magnetic transition temperature (below 30.5 K), the application of a magnetic field causes positive magnetostriction in agreement with the measurements of the macroscopic variation of the sample length using a fiber Bragg grating (FBG) technique [13]. When the magnetic field direction is reversed, the sample initially compresses until a critical field is reached and then rapidly expands. The overall behaviour resembles the magneto-elastic “*butterfly*” previously seen with the FBG magnetostriction measurements [13] and represents piezomagnetic switching between two magnetic structures connected by the time reversal symmetry. X-rays offer additional insight not available via bulk FBG methods – specifically, we observe a splitting of the [888] peak under the magnetic field. This arises from time-reversed domains of $3\text{-}k$ magnetic structures that have different responses to the applied magnetic fields. This study represents the first

crystallographic observation of piezomagnetism and the switching effect in general, and in a $5f$ -electron spin system in particular.

Results:

The presence of a sudden volume collapse in the unit cell of UO_2 at low temperatures has been known for some time [22] [23]. The small discontinuity corresponds to the rapid and simultaneous magnetic, electrical, and structural transition at $T_N = 30.5$ K. Comparing the relative change in the d -spacing corresponding to the [888] peak with temperature, which was taken using back-reflection geometry X-ray diffraction, to previously taken dilatometry on single crystal UO_2 in the $\langle 111 \rangle$ direction [13] in Figure 1a, one can see that the diffraction reproduces the expected behaviour. The conversion from the angle of the Bragg peak to micro-strain (or ppm) is made using: $\Delta L/L = [\sin \theta_0 / \sin \theta'] - 1$, where θ' is the Bragg angle of the [888] peak at a given temperature or applied magnetic field; θ_0 is its zero-field value. The precision of both techniques is comparable, showing approximately a -30 ppm strain change in the length along the $\langle 111 \rangle$ direction at 30.5 K. This shows that the volume collapse at T_N can be observed as the material goes from a paramagnetic state to the ordered $3\text{-}k$ antiferromagnetic state. When subjected to a pulsed magnetic field in the paramagnetic state ($T = 40$ K), the diffraction shows a small contraction of the unit cell along the $\langle 111 \rangle$ direction of about 50 ppm at the maximum field of 21.2 T, as seen in Figure 1b, which also matches the FBG magnetostriction measurements [13]. The structure then reversibly changes back after returning to zero field.

When repeating the measurements in the magnetically ordered state, a different and unexpected response occurs. Represented schematically in Figure 2a (not to scale), FBG magnetostriction measurements in a pulsed magnetic field below T_N revealed that the bulk crystal *expanded* in the $\langle 111 \rangle$ direction, the reverse of the non-magnetic state. While measuring diffraction in the back-reflection geometry, the sample was cooled to 15 K, then subjected to a pulsed field in the $\langle 111 \rangle$ direction.

Under the application of the magnetic field, the [888] peak splits and re-converges as the applied field rises and falls, reversibly returning to its original state after the pulse [see supplemental materials for more details]. This surprising result contrasts with the single peak seen in the non-magnetic state. Moreover, the split peaks shift in opposite directions, i.e. higher and lower scattering angles (2θ), indicating simultaneous contraction and expansion of the same crystal lattice. This can only occur if X-ray diffraction originates in two types of physically distinct, three dimensionally ordered regions of the sample with a robust local symmetry producing fully coherent Bragg peaks. Remarkably, the two regions respond with opposite sign just as one would expect from two magnetic domains related by time-reversal in a magnetic material. This is a very unusual observation, hard to explain in the context of the *fcc* crystal structure of UO_2 yet a natural consequence in a piezomagnetic system. We, hence, believe that the splitting arises from time-reversed domains of the 3-k magnetic structure that have opposite responses to the applied magnetic fields. The integrated intensities of the peaks along 2θ shown in Figure 2b reveal that there is a larger volume fraction of the sample that corresponds to expansion (blue peak), and a smaller component that corresponds to contraction (red peak). The peak positions of these two separate components plotted in Figure 2c show that, though the contracting component has a lower intensity, the absolute value of its corresponding strain is larger. The peak positions in the rising and falling field overlap symmetrically as well, showing no hysteresis. Repeatedly applying the pulsed field in the same field direction produces the same result. As can be seen from the figure, the magnetostriction obtained here, governed by the blue peak, agrees well with the FBG pulsed field magnetostriction measurements (black line) [13].

When the sample is subsequently exposed to a pulsed magnetic field applied in the direction opposite that of the previously applied field, startlingly different behaviour is seen. Represented schematically in Figure 3a, the FBG magnetostriction measurements show that in this reversed field direction in the magnetic state, the response along the $\langle 111 \rangle$ direction is a linear contraction,

displaying broken time-reversal symmetry, a characteristic of the piezomagnetic effect [13]. After reaching a certain temperature-dependent critical applied field strength [24], the sample rapidly returns to the previous expansion response seen in the single direction applied field used in the first part of the experiment, then follows that path back to the same initial state at zero field. Looking at the integrated intensities of the diffracted peaks in Figure 3b, the behaviour of the peaks is very different from the single field direction. Furthermore, one of the domains (red peak) is much more sensitive to the applied magnetic field, i.e. reaching 700 ppm strain in 20 T, than all the other peaks measured. As seen there, the overall response is compressive at approximately -10 T (see point *b* in Figure 3). But then, at increasingly negative field values, the data return abruptly to behavior similar to that seen in Fig. 2b, albeit with different relative intensities. The key point is that upon returning from -20 T to zero, the overall compressive strain is not recovered near -10 T (point *f* in Fig.3). Immediately switching the field direction again shows this same new behavior, resulting in a *butterfly*-like loop [13]. The peak is clearly seen to split again (first as a broadening of the peak and then as a separate peak), this time in a positive field following immediately after negative pulses at 25 K [a positive magnetic field part is shown in the supplemental materials]. The FBG experiments have shown that critical field of about 11 T is expected at this temperature, as marked by the solid line in Figure 3b [see the supplemental materials for more details and Ref.24]. This behaviour provides direct evidence that the “butterfly” hysteresis observed in the FBG experiment can therefore be related to piezomagnetic domain evolution revealed by X-rays. Figure 3c shows the relative positions of the peaks (blue squares and red circles). The lines are visual guides. The higher angle and intensity “blue peak” (point *b* in Figure 3) undergoes a switching from contraction to expansion at the critical field of \sim -11 T. This demonstrates that the sign of the strain in the dominating peak is rapidly reversed in this region so that the blue peak matches the butterfly pattern observed in the magnetostriction taken at the same temperature. If so, then only magnetic domains corresponding to the blue peaks have sufficiently low critical fields to exhibit the rapid reversal of strain behaviour

expected from the piezomagnetic effect. It has been proposed that, in UO_2 crystals subject to pulsed magnetic fields of reversed polarity, the magnetic subsystem switches between two states connected by time reversal [13]. In Figure 3c, this switching phenomenon between the two states is directly observed. In general, the piezomagnetic response, related to UO_2 's $fcc, Pa\bar{3}$ structure, can be explained using a model Hamiltonian that includes a strong magnetic anisotropy, elastic, Zeeman, Heisenberg exchange, and magnetoelastic contributions to the total energy. This simple model, where the degrees of freedom are the orientation of the magnetic moments of the four U atoms in the $Pa\bar{3}$ unit cell and the shear components of the strain tensor, successfully reproduces the intriguing experimental observations (see supplementary materials in Ref.13 for more details regarding the model used). Minimizing the total energy with respect to the elastic shear components allows obtaining their dependence on the applied magnetic field:

$$\varepsilon_{xy} = \frac{E}{c_{44}a^3} M_{st} H_z$$

with similar expressions for the other components. Here $E = 0.280 \text{ meV } T^{-1}$ is the strength of the magnetoelastic interaction [13], $c_{44} = 60 \text{ GPa}$ and $a = 5.47 \text{ \AA}$ are the experimental shear elastic and lattice constants respectively, and M_{st} is the staggered magnetization that is different from zero below the Néel temperature. The change of sign of the staggered magnetization, which can be positive or negative depending on which AFM structure connected by time reversal symmetry is stabilized, allows understanding the observed "butterfly" behaviour of the magnetostriction (Figure 3c).

The phenomenology of peaks splitting under applied magnetic field in UO_2 is contrary to the more common case of magnetic detwinning. More typically, the application of magnetic fields to materials containing symmetry-related twin domains will result in a free energy difference between the domains, favouring one over the other and driving the material into a single-domain state. Conversely in the case of

UO_2 , at least in the low-field regime, it is clear that the applied magnetic field drives opposing magnetostrictive responses in different time-reversed domains without converting one domain entirely into the other. As seen in Fig. 1, only a single peak, shifted by magnetostriction, appears above the Neel temperature. Also, a single peak is observed above T_N under high pressure [see Suppl. Mat.], and below T_N in the absence of magnetic fields. Because the two-peak structure seen in Figs. 2 and 3 is only observed in the magnetic-field-polarized antiferromagnetic state, it is likely that the two magnetic domains consist of distinct time-reversed versions of the $3\text{-}\mathbf{k}$ structure. The piezomagnetism of UO_2 [13] occurs as the $3\text{-}\mathbf{k}$ structure of the ordered phase switches between TR versions under the action of applied magnetic fields. The red circles represent the negative magnetostriction of the $-\text{TR}$ state in positive fields. That this state has not fully converted to the $+\text{TR}$ state by a field of +20 T indicates that either this TR state is pinned, or that the switching field in this domain is larger than 20 T. The robustness of minority domains to field-conversion may be closely related to the exceptionally hard piezomagnetic response of UO_2 . Interestingly, the presence of two 180 deg antiferromagnetic domains has been observed before in another piezomagnet, MnF_2 , via polarized neutron tomography [25,26]. Furthermore, the domain configuration was determined by and sensitive to the strain condition of the specimen. It was suggested that for given local stress, the domain configuration (type A and/or B) depends on the direction of the applied magnetic field, such as the domain type being reversed when the magnetic field points in the opposite direction [25]. A single domain state is possible if the stress is uniform over the entire crystal. This is a condition difficult to achieve, however, since an increase in the number of domains to attain small-scale variations of stress is prevented by the large wall anisotropy energy. In our experimental configurations, the strain effects in UO_2 might play an even bigger role. A potential domain pinning and strain effect imposed by the stycast epoxy (see figure 3 in the supplemental materials), could increase the piezomagnetic switching field so that the preparation of a single TR state becomes increasingly difficult. It has been shown that in other antiferromagnets, such as NiO crystal [27], the domain structure has been very

sensitive to mechanical stress and magnetic fields, where the twin wall dynamics appear to be limited by a spin-rotation energy loss. We speculate that the sensitivity of the switching field to mechanical stress might be related to the proximity of the mixed antiferromagnetic/antiferroquadrupolar state of UO_2 to the non-magnetic, purely quadrupolar phase [28]. A virtual transition into the quadrupolar state allows for the conversion from one TR state to the other without the need to overcome an anisotropy barrier. In this scenario, internal constraints modify the local distortions that give rise to electric-field gradients and hence reduce the presence of the purely quadrupolar phase. Experiments are underway to explore the sensitivity of the switching field to uniaxial strain in this material. We would like to point out that the 888 peak splitting effect (the presence of two magnetic domains) observed in UO_2 is real and does not originate in any way from the superposition of many fields- pulse combinations.

Summary and Conclusions:

We report the first low-temperature single crystal X-ray diffraction studies of UO_2 in pulsed magnetic fields. The high-resolution single crystal diffraction allows us to study details of subtle unit-cell distortions below and above the structural and magnetic phase transition. We unveil direct microstructural observations of the piezomagnetic and switching effects being a direct consequence of the non-collinear $3\text{-}k$ magnetic order that breaks time-reversal symmetry in a non-trivial way. These results will help in better understanding the strong coupling between magnetic and structural degrees of freedom in this important nuclear material. We also observe the presence of magnetic domains with distinct magnetic-field evolution in the magnetically ordered state of UO_2 , both when the field is applied repeatedly in a single direction and when the field direction is alternated. We argue that the origin of this behaviour is related to the presence of the distinct time-reversed versions of the

$3-k$ structure. This behaviour, especially the role of quadrupoles on the piezomagnetic properties, should be investigated in the future to further explore the details of the field-induced broken symmetries.

METHODS:

It is worth noting that the quality of the data obtained in the experiment was only possible by bringing together a combination of unique factors. Through use of an exceptional quality single crystal sample (evidenced in very narrow diffraction peaks), high intensity synchrotron radiation, high, pulsed magnetic fields, and a fast prototype compound area detector, we were able to achieve high precision in our measurements during the short time frame of a pulsed fields (using one of the two choices of ~ 7 and 10 ms in total duration [start-to-finish] currently available). The single crystal of UO_2 that was used in these studies is from the same batch as used in previous measurements (Refs. [5] and [13]). The crystal was aligned and cut into a plate approximately 500 x 500 x 200 microns, with the [111] crystal face normal to the plane. This allows us to study the longitudinal magnetostriction of the UO_2 crystal along the desired $\langle 111 \rangle$ crystallographic direction. The experiment was carried out in a dual cryostat single solenoid pulsed-magnet system described in [29], in beamline 6 ID-C of the Advanced Photon Source at Argonne National Laboratory.

The detector used was a compound type Mixed-Mode Pixel Array Detector (MM-PAD) with a silicon sensor [30] [31]. This allowed for high dynamic range (up to 2×10^7 X-rays/pixel/frame at 15.85 keV), low background single photon counting, and a fast frame rate so that multiple images at different rising and falling field strengths during pulses could be obtained. The angular 2Θ range of the beam, where 2Θ is the angle between the incident and scattered rays, was calibrated on the area detector using the [888] peak of a silicon reference crystal, mounted at the center of sample theta

rotation of the cryostat. The collection window for each frame was about 140 microseconds at a frame rate of 1 kHz, and the detector array was ca. 4 cm x 6 cm. For temperature scans in zero field, the sample was stabilized at various temperatures and then rotated through the Bragg condition at several hundred discrete sample theta positions as frames were taken at each position, then the frame intensities were added to obtain the full [888] peak profile. The center of mass in sample θ and Bragg peak position 2θ of the profile were found and converted to the change in d-spacing to find the change in unit cell length along the $\langle 111 \rangle$ direction. A similar process was used for measurements in the applied field, with fewer discrete sample θ rotation positions, limited by the long duty cycle of the magnet for each individual pulse, as elaborated on in the supplementary materials.

The capacitor bank used in these experiments provided a maximum field of about 21 Tesla and a pulse width of approximately 7 milliseconds, as shown in Supplementary Figure 5, with the ability to internally reverse the direction of the supplied current to the magnet coil. The reversed field part of the experiment was carried out in a similar manner, but after a first pulse in the positive direction (which was then discarded), successive pulses were taken in alternating directions along the $\langle 111 \rangle$ direction of the sample crystal. The resulting reversed-field direction frames were then analysed the same way as the single direction pulses (figures 3b and 3c).

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Contributions: K.G. proposed the experimental studies and D.A., Z.I., and K.G. designed the research. D.A., Z.I., J.W., K.S., J.R., M.J., B.L., K.S. and K.G. performed the experiments. T.D., D.A., C.S., provided the UO₂ single crystal samples. D.K. helped in the sample preparation. A.S., T.S., M.J., K.G., and M.S. discussed and developed the theoretical description. All authors helped prepare various pieces of the experimental apparatus, discussed and interpreted the results, and contributed to the preparation and writing of the manuscript.

Competing interests statement: The authors declare that they have no competing financial interests.

Data availability: The data that support the findings of this study are available from the corresponding author upon request.

Correspondence and requests for materials should be addressed to K.G. (e-mail: gofryk@inl.gov).

Figure 1 (a) The relative change of the lattice constant in the $\langle 111 \rangle$ direction of the UO_2 single crystal as a function of temperature, measured by dilatometry of the bulk crystal (dashed line) and X-ray diffraction of the [888] peak (red squares). The X-ray data were obtained by single crystal diffraction in high-angle back-reflection geometry using 15.85 keV X-rays. An abrupt collapse in unit cell volume can be seen at $T_N = 30.8$ K, marked by an arrow. Insets show the structure of the UO_2 cubic unit cell above T_N (paramagnetic phase) and below T_N (ordered phase). **(b)** The magnetic response of the [888] peak in the paramagnetic phase ($T = 40$ K) to an applied pulsed magnetic field in the $\langle 111 \rangle$ direction. The initial state at 0 T (black line) shifts to higher 2θ at maximum applied field of 21.2 T (red line), then back to the same initial value after the pulse (blue line). The shift in 2θ of the peak corresponds to a small ~ 50 ppm strain contraction in the $\langle 111 \rangle$ direction, consistent with bulk single crystal magnetostriction measurements [2].

Figure 2 (a) A schematic diagram showing the bulk sample magnetostriction along the $\langle 111 \rangle$ direction to a magnetic field applied repeatedly to it in the same direction (bold blue line), showing the sample reversibly expanding. A pale blue pattern shows field dependence of the magnetostriction for positive and negative fields directions **(b)** A waterfall plot of fits to the integrated intensity along 2θ of selected fields and at 15 K. The peak at lower 2θ (blue) corresponding to expansion is much larger than the peak at higher 2θ (red). **(c)** The peak positions of the two peaks at 15 K, with filled symbols corresponding to rising field and open symbols to falling field. Red circles represent the smaller peak and blue squares the larger one. The blue peak and its positive magnetostriction can be seen to match previous magnetostriction measurements using the fiber Bragg grating technique (black line), with about a 150 ppm expansion at the maximum 21.1 T applied field. The red dash line is guide to the eye.

Figure 3 (a) A schematic diagram showing the bulk sample magnetostriction along the $\langle 111 \rangle$ direction in a magnetic field applied immediately in the direction opposite to that of the previously applied field (bold blue line). As opposed to figure 2a, the sample shows irreversible behaviour by instead initially contracting in the $\langle 111 \rangle$ direction, but then rapidly switching back to the previous expanding behaviour once a threshold field has been surpassed. **(b)** The integrated intensity in 2θ for selected fields and at temperature, $T = 25$ K. As opposed to 2c, the higher 2θ peak is initially larger, but then a change in relative intensities and peak position can be seen. **(c)** A plot of the relative strain calculated from the peak positions in the reversed field state at 25 K. The peak corresponding to the dominant strain behaviour as determined by the bulk magnetostriction is the blue squares and the secondary peak corresponding to negative strain is the red circles. Note that the curves for positive fields are similar to the data in Fig. 3b. Open symbols are used for rising fields and closed for falling fields. The blue solid line is guide to the eye. The solid black line is the reversed field measurements using the FBG technique taken in positive and negative fields at 25 K.

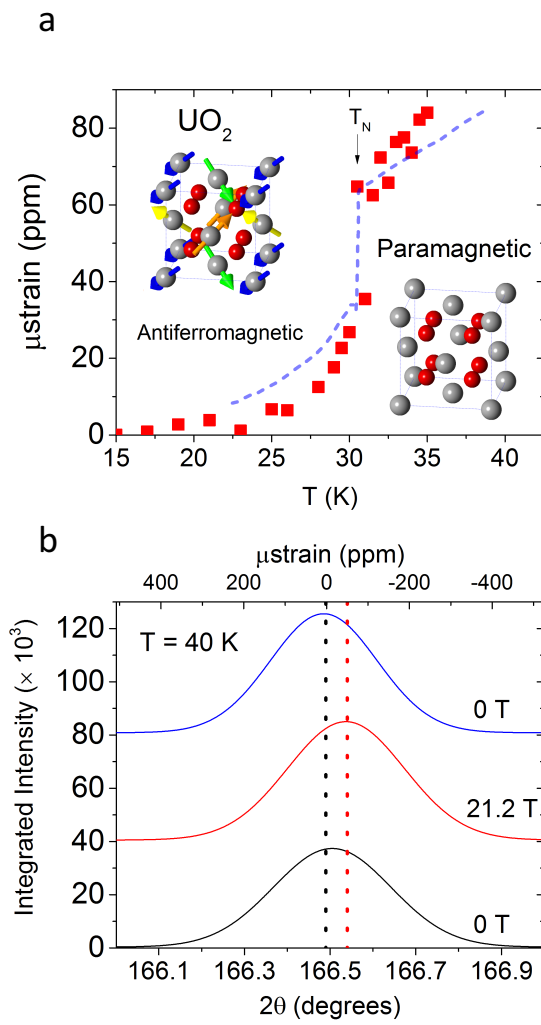


Figure 1

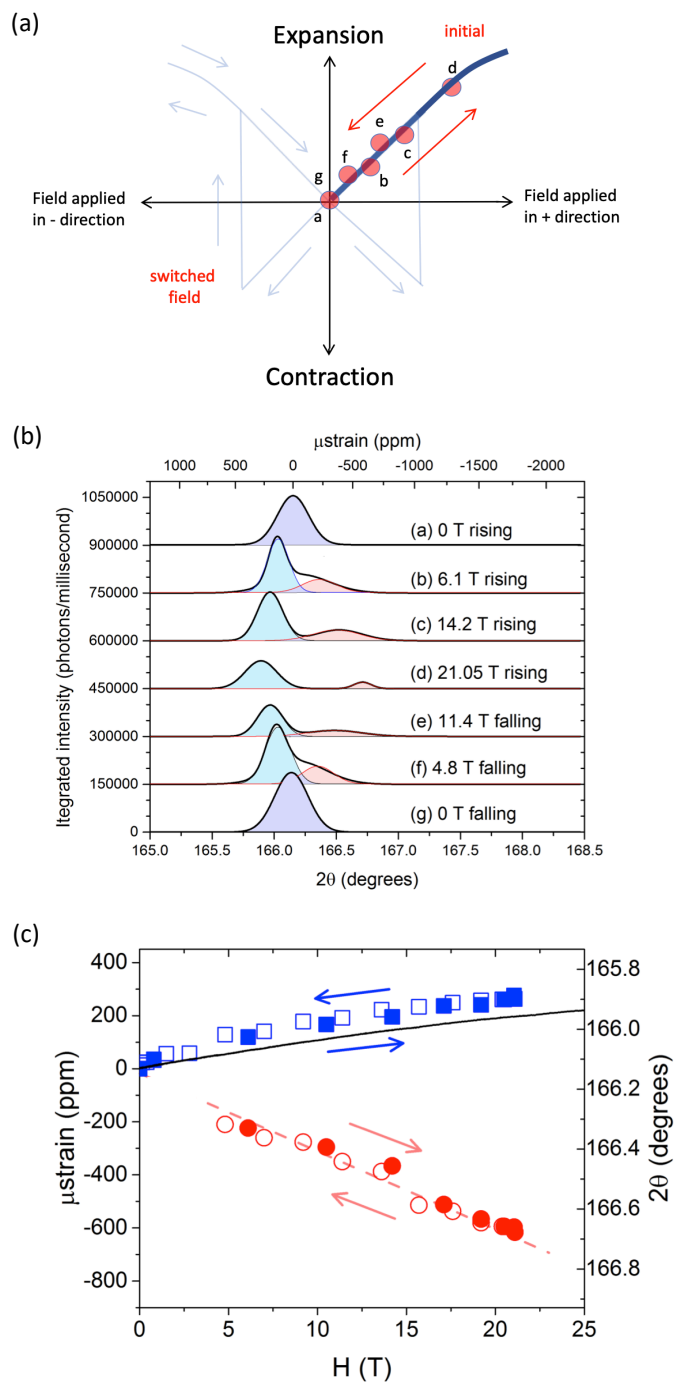


Figure 2

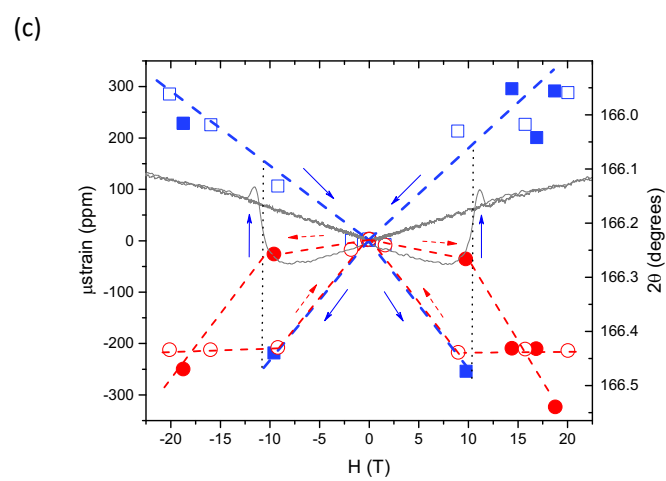
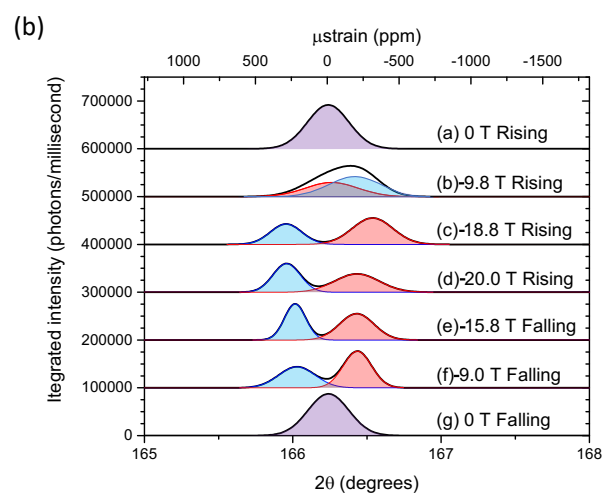
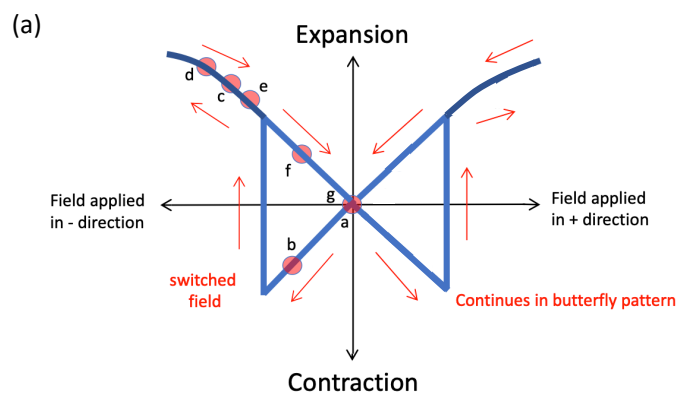


Figure 3