SUPPORTING INFORMATION FOR:

Effect of antifluorite layer on the magnetic order in Eu-based 1111 compounds, Eu*T*AsF (*T* = Zn, Mn, and Fe)

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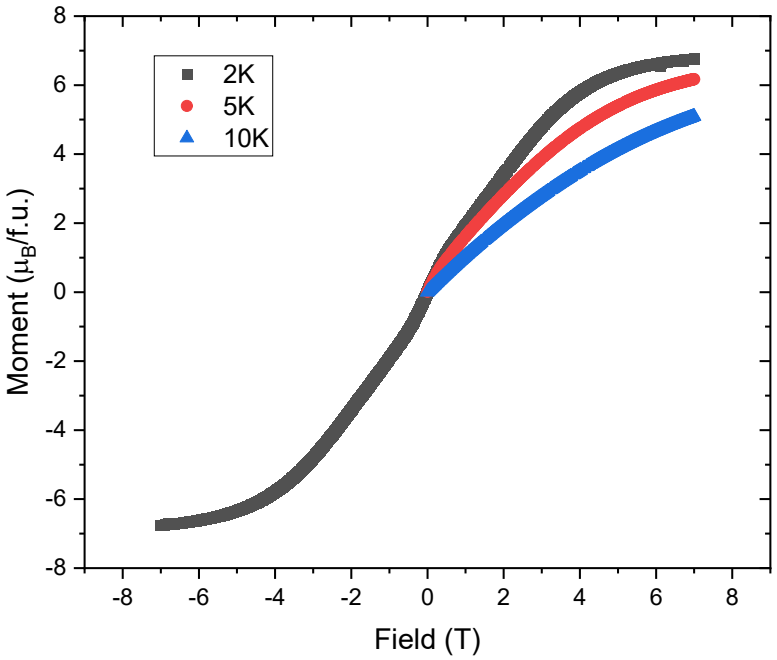

Figure 1. Magnetization vs Field curves for EuFeAsF measured at different temperatures.

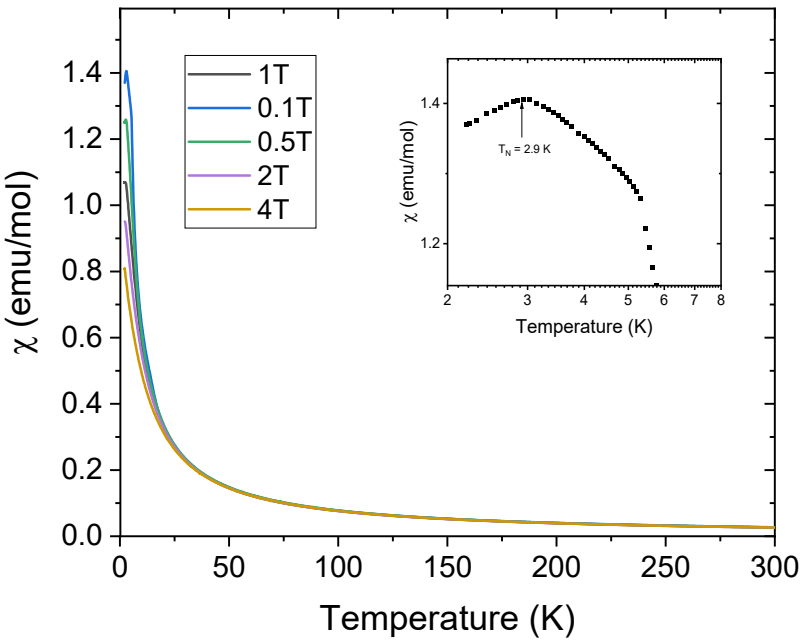

Figure 2. Magnetic susceptibility χ = M/H for EuFeAsF measured in different fields. Low temperature part measured in 0.1 T with the position of phase transition outlined.

Determination of the incommensurate Eu^{2+} magnetic moments structure

The diffraction patterns of EuFeAsF measured above (at 10 K) and below (at 1.5 K) the 3 K phase transition are shown in ***Figure 4***. Several very strong magnetic reflections are observed, some of them are even stronger than the nuclear ones. The triplet of magnetic reflections at 5.19 Å, 5.33 Å, and 5.57 Å, at the *d*-spacings comparable though not coinciding with the *a* and *b* cell parameters, is indicative of an incommensurate magnetic order. Indeed, these reflections, as well as those at lower *d*-spacings, can be indexed with the single incommensurate vector *k* = (0 0.961(1) ½) (assuming the following setting of the unit cell: *a* = 5.5917(2) Å, *b* = 5.6344(2) Å and *c* = 8.8821(3) Å). Representation analysis yields four maximal magnetic superspace groups of the initial nuclear group *Cmme*, which allow non-zero magnetic moments on Eu atoms: *Cmme*.1'(0$b^{1}/_{2}$)000*s*, *Cmme*.1'(0$b^{1}/_{2}$)*s*0*ss*, *Cmme*.1'(0$b^{1}/_{2}$)*s*00*s*, and *Cmme*.1'(0$b^{1}/_{2}$)00*ss*. Attempts to refine the magnetic structures using symmetry restrictions imposed by these superspace groups result in the third one, *Cmme*.1'(0$b^{1}/_{2}$)*s*00*s*, to give an acceptable description of all magnetic intensities. The resulting structure is an incommensurate spin-density wave. Magnetic moments are aligned along the *b*-direction, which is the propagation direction; their amplitude changes as a sin-wave from 7.36 μ_B to zero. The maximum magnetic moment is higher than theoretically expected (7 μ_B), which indicates that the proposed model is not physically realistic. Additionally, an incommensurate spin density wave is unlikely to be the ground-state structure because some of the localized moments of Eu^{2+} are effectively suppressed. Therefore, we searched for a more plausible model with the constant Eu^{2+} moment among subgroups of the initial superspace group.

Removing the 1' symmetry element results in a set of magnetic space groups that differ from each other by a permutation of the space-group generators (*m* and *m'*, *m* and *m'*, *b* and *b'* along *a*, *b* and *c* axes respectively), but allows one to stay in the same crystallographic space group *Cmme*, *i.e.* without splitting any of the atomic positions. Unfortunately, none of these eight groups led to a model that would be significantly different from the one provided by *Cmme*.1'(0$b^{1}/_{2}$)*s*00*s*. Therefore, we attempted to reduce the symmetry by removing one of the mirror planes and transforming the *mmm* crystallographic point symmetry into *mm*2. The group *Cmm*2.1'(0$b^{1}/_{2}$)*s*00*s* splits the position of Eu atom into two and allows magnetic moment to have a refinable component along the *c*-direction. By forcing the same amplitude of the magnetic moments in both atomic positions, as well as the same amplitude throughout the whole structure, we obtain the final solution provided in ***Table 1.*** The refinement returned the magnetic moment of 5.64(3) μ_B, which is only slightly lower than the expected value of 7 μ_B. The deviation may be explained by the proximity of our measurement temperature to the Néel temperature, such that magnetic moments are not fully saturated. Similar to the aforementioned commensurate case, the *b*-components of the magnetic moments form two stripes per unit cell that are coupled to each other in antiferromagnetic manner. The magnetic moments gradually rotate in the *bc* plane when passing from one Eu^{2+} ion to another along the *b*-direction. The overall spin arrangement in the *ab* plane resembles that in EuZnAsF (nearest-neighbor spins along *b* are antiparallel, ***Figure 5***), albeit with an additional incommensurate modulation.