Unusual magnetic behaviour in the potential multiferroic, $Pb_3TeCo_3V_2O_{14}$

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The langasites have been a major focus of study in the last decade due to the variety of properties they exhibit including piezoelectricity in $La_3Ga_5SiO_{14}$ [1], highly degenerate ground states in $Pr_3Ga_5SiO_{14}$ [2], and multiferroicity in $Ba_3NbFe_3Si_2O_{14}$ [3]. The latter has been a particularly active area of research because of complex spin dynamics arising from chiral, helical antiferromagnetic ordering at T_N = 26 K. The chirality introduced by the magnetic structure breaks symmetry, which allows this material to undergo a ferroelectric polarization [4]. Efforts to increase the multiferroic transition temperature by chemical substitution have been unsuccessful; Fe^{3+} occupies a tetrahedral site that other magnetic ions find unfavourable compared to the octahedral Nb^{5+} site.

Drawing on previous work from B.V. Mill [5], we have optimized a method to substitute Fe^{3+} for other magnetic cations by taking advantage of Te^{6+}, which is only known to exist in octahedral coordination [6]. Te^{6+} has a high affinity for the Nb^{5+} site forcing magnetic ions like Co^{2+}, Mn^{2+}, and Cu^{+} into the tetrahedral site. We have synthesized isostructural $Pb_3TeCo_3V_2O_{14}$, a member of the dugganite family, and have performed neutron scattering experiments on the C2 diffractometer [7].

Although our efforts to increase the ordering temperature have failed, the underlying magnetic behaviour of this system is unlike any other langasite studied thus far; the dugganite has *two* magnetic transition temperatures at T_{N1} = 8.6 K and T_{N2} = 6.0 K (Figure 1) more reminiscent of other Co-V systems like $Co_3V_2O_8$ and monoclinic $Co_2V_2O_7$. A remarkable reordering of Co^{2+} moments occurs within a 2.6 K range. The first magnetic structure has a propagation vector k = (0.752, 0, 1/2) while the second has a propagation vector k = (5/6, 5/6, 1/2). While the second structure has been studied in detail, knowledge of the first structure remains rather limited, though recent work suggests this phase may contain a dynamic component (Figure 2).

The temperature dependence of the dielectric constant was measured to determine if this sample displays magnetoelectric coupling. An anomaly can be seen at each magnetic transition temperature (Figure 3). Thorough investigation of the structure yields two possible causes for these anomalies. Firstly, short V-O distance along the *c* axis seen across all members of the dugganite series may introduce antiferroelectric ordering. An alternative explanation may be the dynamical nature of the first magnetic phase. Perhaps a preferred moment orientation exists in a dynamic row of Co^{2+} moments, which coexist with static ordered moments. Further study is needed to confirm these findings.

References

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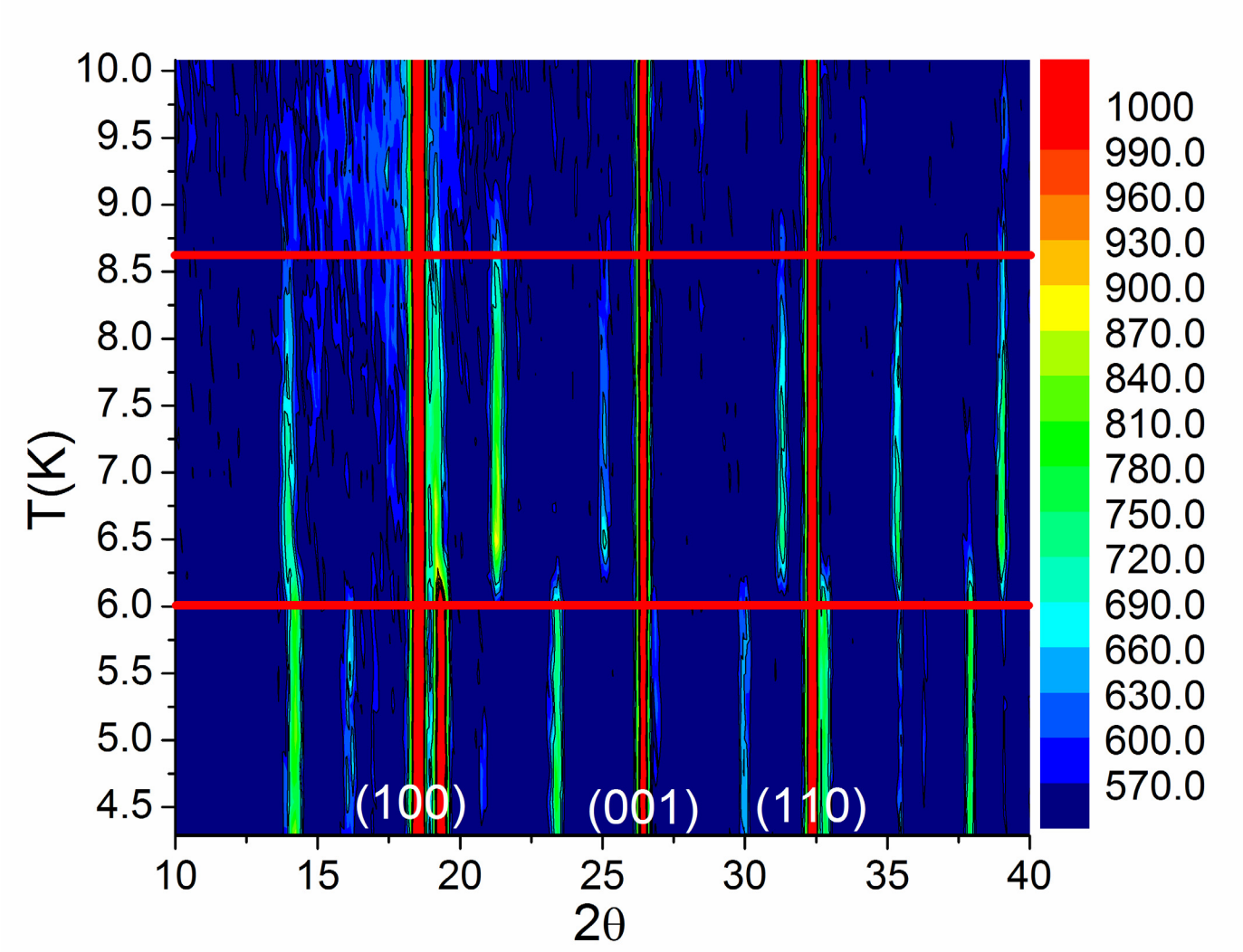

Fig. 1 Neutron diffractogram contour plot showing two magnetic transition temperatures at T_{N1}=8.6 K and T_{N2}=6.0 K (red lines). Diffuse scattering surrounding the (100) peak is seen at T > 8.0 K.

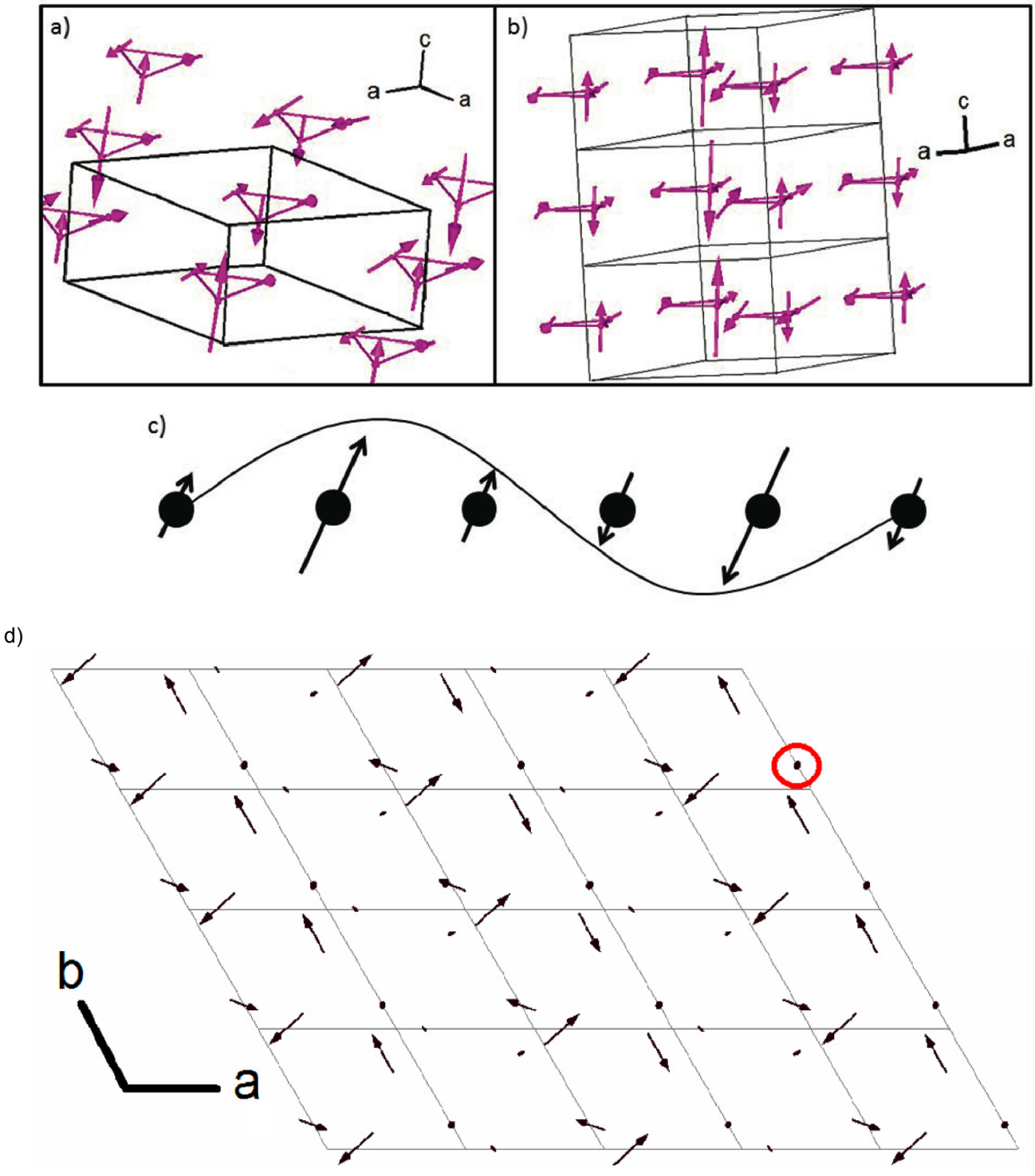

Fig. 2 The magnetic structure at T<6.0 K from the a) *ab* and b) *ac* planes; c) the magnitude of the moment changes over 6 unit cells; d) the magnetic structure for 6.0<T<8.6 K. The black dots (circled in red) represent dynamic spins.

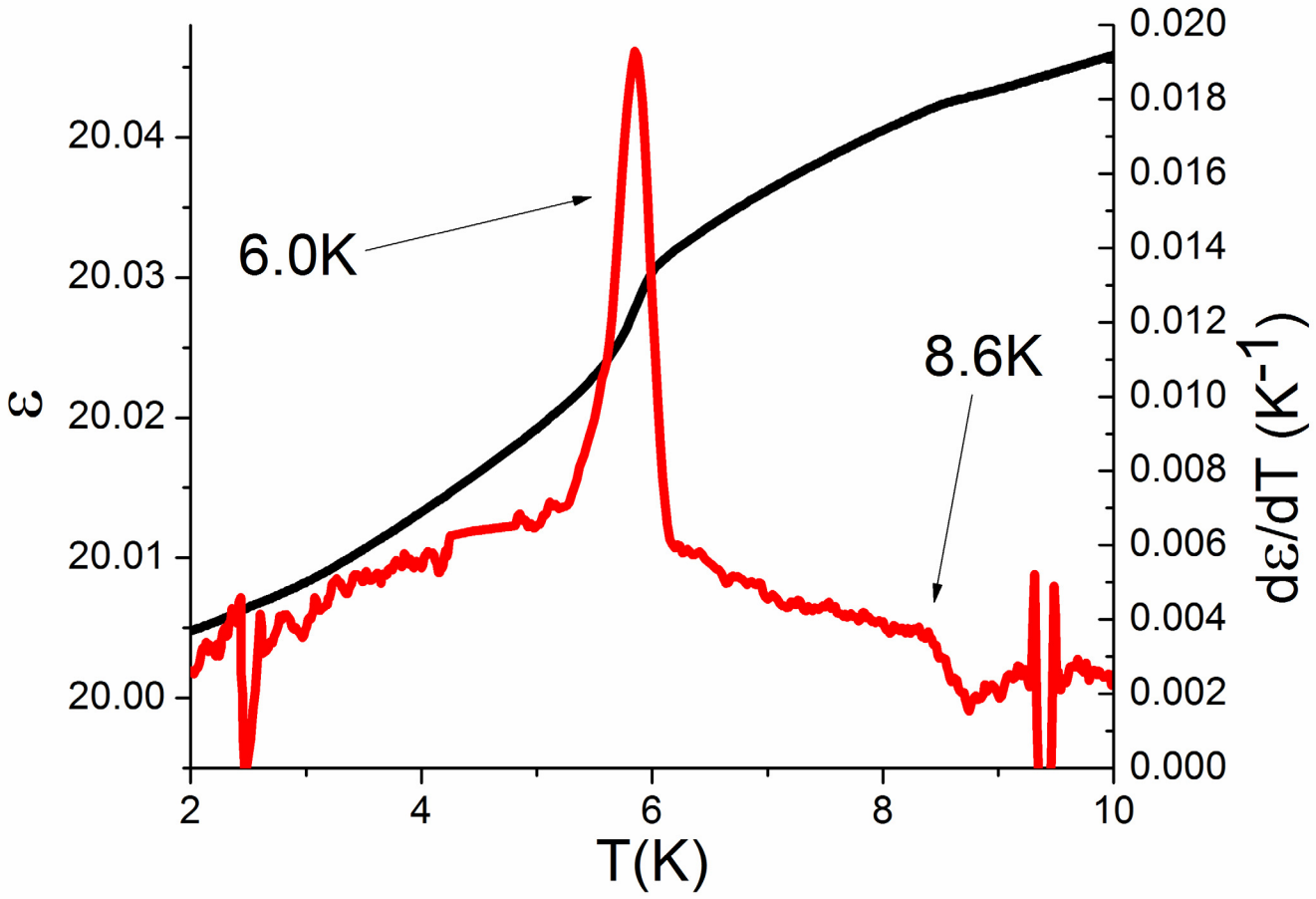

Fig. 3 The temperature dependence of the dielectric constant (black) shows anomalies at each of the transition temperatures. The first derivative of the dielectric constant (red) shows these anomalies more clearly.