INTERPLAY OF ANTIFERROQUADRUPOLAR AND ANTIFERROMAGNETIC ORDER IN TmTe

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Neutron diffraction is the reference technique for probing long-range order formed on a lattice of atoms or magnetic moments. It is shown that, under certain conditions, it can also be invaluable for studying more exotic types of ordered structures involving electron charge distributions. Although the determination is indirect in this case, detailed information on the ordered state of quadrupole moments can be derived from the symmetry properties of the response to an applied magnetic field. This method is demonstrated in the case of the antiferroquadrupolar phase of TmTe.

Besides their magnetic dipole moments, lanthanide elements with incomplete 4*f* electron shells are known to also possess higher-order moments (quadrupole, octupole, etc.). In a classical picture, this reflects the non-sphericity of the electron charge distribution. In the case of solids, pair interactions between 4*f* quadrupoles located at neighboring sites can occur either directly through their electrostatic potentials (usually weak), or indirectly through various channels such as lattice strains (cooperative Jahn-Teller effect), conduction electrons in metals (RKKY-type coupling), higher-order exchange terms, etc. For most real systems, conventional magnetic interactions dominate, and the 4*f* dipole-moment lattice orders in a long-range magnetic structure at low temperature. Accordingly, the quadrupole moments will have non-zero values in the magnetic state, but this is only the result of dipole ordering. More rarely, quadrupole interactions can prevail and produce a phase transition on their own, whose primary order parameter is a component, or combination of components, of the quadrupole moment tensor. In some intermetallic compounds (TmZn, CeAg), as well as in typical Jahn-Teller systems (rare-earth zircons), the value of the order parameter is uniform at all sites, and the order is thus denoted "ferroquadrupolar" in analogy with magnetism. On the other hand, staggered types of quadrupole order, loosely termed "antiferro-quadrupolar" (AFQ), have been reported so far only for a small number of metallic compounds ($TmGa_3$, CeB_6). However, their study is of particular interest because the tensor nature of the quadrupole moment operator, as well as the possible interplay between magnetic and quadrupole order parameters, may result in a rich variety of physical situations. To characterize these phases, microscopic information is even more crucial than in the case of magnetism because one has to determine not only the wave-vectors and Fourier components of the structure, but also which components of the quadrupole moment tensor constitute the order parameter.

At first sight, neutron experiments do not seem well suited to probing quadrupole order because the neutron does not interact directly with electrostatic charge distributions. However, as was shown in the work of Effantin et al. on CeB_6 [1], this obstacle can be partly circumvented by concentrating on the response of the *dipole* moment lattice to a magnetic field applied along a high-symmetry direction in a single-crystal: indeed, this response is strongly constrained by the preexisting quadrupole order, and therefore contains relevant information, in the first place concerning the wave-vector of the quadrupole structure. We have applied this strategy to the magnetic semiconductor TmTe, which was recently reported to undergo a phase transition at $T_Q \sim 1.8$ K [2] (far above the Néel temperature of approximately 0.4 – 0.6 K) whose characteristics are suggestive of quadrupole order.

High-field measurements have been performed on the lifting detector diffractometer 6T2 using the Saclay-Grenoble 12-tesla split-pair cryomagnet, The data shown in Figure 1 clearly indicate that superstructure magnetic peaks associated with the zone-boundary wave-vector $\boldsymbol{k}$ = (1/2,1/2,1/2), which were absent in zero field, grow below T_Q as H is increased parallel to the [110] direction.

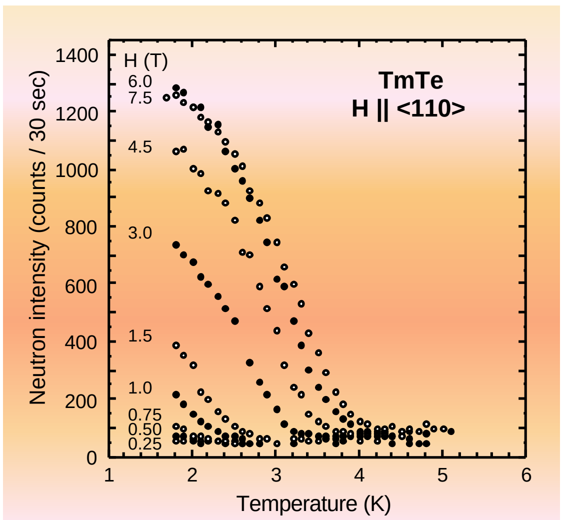

Figure 1. Intensity of the magnetic Bragg peak (3/2, -1/2, 1/2) induced by the external field H || [110].

This result is very important because it establishes the possibility for a *uniform* field to induce a *staggered* magnetic component in the ordered state below T_Q, whereas it induces only a component at $\boldsymbol{q}$ = 0 in the paramagnetic state. This lends considerable support to the above assumption of an underlying quadrupole order. By tracing these intensities as a function of temperature for different values of H, we were able to delineate the quadrupolar phase diagram for the three main symmetry directions, H || [001], [110], and [111] (Figure 2), and found good agreement with existing specific-heat results.

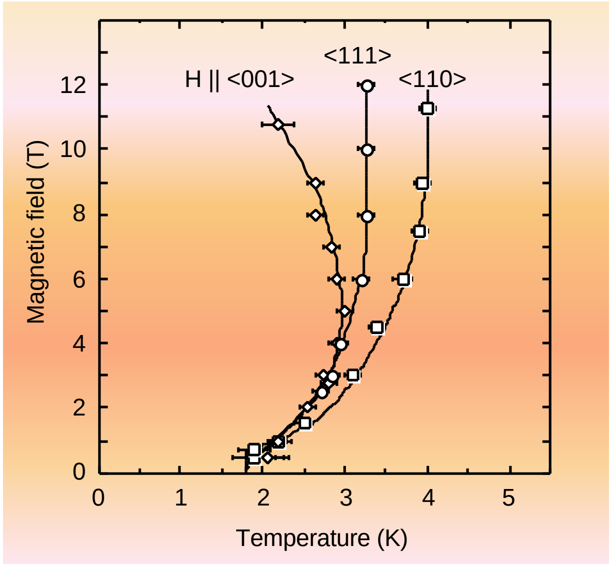

Figure 2. Quadrupolar phase diagram from the neutron diffraction results.

Furthermore, the quality of the data allowed us to fully disentangle the contribution of the different *k*- and *S*-domains, and to establish the direction of the staggered magnetic component μ_{AF} for H || [110], and [111]. In both cases, the best refinement was obtained by assuming μ_{AF} to be oriented along the two-fold axis $[\bar{1}10]$ perpendicular to both the wave-vector $\boldsymbol{k}$ and the field direction. For H || [001], on the other hand, the induced magnetic component is much weaker and the direction of μ_{AF} cannot be reliably determined. Using the group-theoretical analysis developed by Shiina et al. [3], it can be concluded that the latter results are compatible with only one type of order parameter, namely O_2^2. A schematic illustration of the field response for H applied along (110) is given in Figure 3.

Recently, the measurements have been extended to temperatures around 0.1 K and it was found that the magnetic phase forming below T_N is of the canted type: it gives rise to two magnetic components, one antiferromagnetic with the same wave-vector $\boldsymbol{k}$ = (1/2,1/2,1/2) as the quadrupolar structure, and the other ferromagnetic. This effect had actually been predicted by Shiina et al. [3] for the case where "in-plane" magnetic couplings (*i. e.* bilinear interactions involving the x and y components of the dipole moment) dominate. Its experimental observation further supports the type of quadrupole order suggested above.

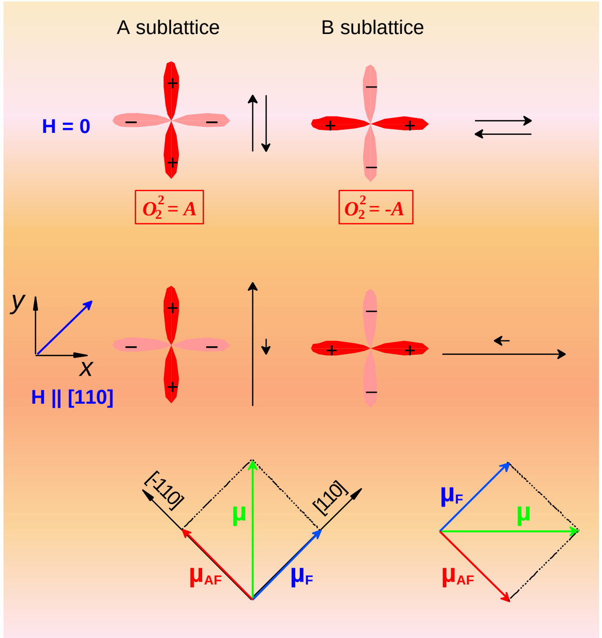

Figure 3. Schematic representation of the effect of a magnetic field H || [110] on the AFQ phase of TmTe; for H || [001], the symmetry is not broken and, ideally, only a uniform magnetic component is expected.

The results of this work [4] demonstrate that neutron diffraction, combined with a large external magnetic field, can provide a very powerful, albeit indirect, tool for studying quadrupole order in solids. Depending on systems, other techniques, such as synchrotron x-ray scattering may offer attractive alternatives. In the case of TmTe, however, the relatively low value of T_Q, as well as the risk of surface oxidation, make neutron diffraction the most straightforward method at the present time.

Further developments of this work, in particular measurements of the excitations in an applied field, will be aimed at clarifying the nature of the interactions responsible for the quadrupole ordering. Higher-order superexchange interactions have been suggested in Reference [3] but this assumption remains to be confirmed.

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