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Magnetism of powder samples and of single crystals

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ABSTRACT

The temperature dependence of the magnetic order parameter of NiF_2, $MnCO_3$ and K_2NiF_4 measured on powder samples is compared with data obtained on single crystals. It is found that the dynamic dimensionality of the NiF_2 and $MnCO_3$ powder samples is one-dimensional (1D) for all temperatures $T<T_N$. This shows that the powder grains are mono-domain particles. In the multi-domain single crystals isotropic 3D dynamic symmetry is observed. In order to explain isotropy in the bulk single crystals a dynamic averaging process over all domain orientations has to be postulated. For single crystal material of K_2NiF_4 a critical exponent of $\beta=0.14\pm0.01$ has been reported. Our neutron diffraction measurements on powder material yield to a very good approximation mean field critical behaviour indicating isotropic dynamics in the critical range. This unexpected result points to a dynamic averaging process over all differently oriented powder grains. As we have argued earlier, the observed universality in the dynamics of ordered magnets is due to a boson guiding field. Averaging over all powder grains, requests that the mean free path of the field bosons is larger than the size of the grains. Additionally, it must be assumed that the field quanta are able to tunnel across the interface between adjacent grains. In polycrystalline bulk samples of the 2D ferromagnet Rb_2CrCl_4 and of the 1D antiferromagnet $KCuF_3$ also mean field critical behaviour is identified. As a consequence, a similar averaging process as in the K_2NiF_4 powder sample can be assumed to average over the mosaic structure of polycrystalline bulk material of Rb_2CrCl_4 and $KCuF_3$. High quality single crystals of Rb_2CrCl_4 and of $KCuF_3$ exhibit critical exponent of $\beta\sim0.3$. Dependence of the critical exponents on the mesoscopic morphology of the sample is considered as a typical indication of a boson controlled dynamics, and may partly explain the surprisingly broad distribution of critical exponent values reported in literature.

1. Introduction

Since development of Renormalization Group (RG) theory by K.G. Wilson we know that in ordered magnets with a three-dimensional spin the dynamics is as for a continuous medium [1]. Spins and interactions between spins are not important for the dynamics [2]. This is well known from the experimental fact that the critical exponents of magnets of the same symmetry class are independent of spin and lattice structure. As experiments show, the dynamics remains independent of the atomistic degrees of freedom for all temperatures down to $T\rightarrow0$ [3,4]. This dynamic behaviour is called universality. On the other hand, magnon dispersions are different for different spin structures. As a consequence, magnons cannot be the relevant excitations for the universal dynamics. Another frequently noticed disagreement with spin wave theory is that magnons at zone boundary (the near neighbour interactions) show virtually no anomaly at the ordering temperature, and persist far into the paramagnetic range [5–7].

When spins and interactions between spins are unimportant for the dynamics it is logical to conclude that thermal energy is no longer in the system of the interacting spins (magnons) but has changed to the delocalised excitations of the magnetic continuum. Quite generally, the excitations of a continuous medium are bosons. The medium can be either an elastic, electric or magnetic continuum. We have called all types of bosons of the continuous magnetic medium GSW bosons, giving tribute to J. Goldstone, A. Salam and S. Weinberg [8]. In other words, the dynamics of the ordered spin system is determined by a boson guiding field. In fact, universality is the typical thermodynamic behaviour of a field of freely propagating bosons.

GSW bosons can be assumed to be essentially magnetic dipole radiation emitted upon precession of the magnetic moments. Since integer and half-integer moments precess differently, the emitted GSW bosons have different properties and the dynamics of the field depends on whether the sources of the field quanta, the spins, have integer or half-integer quantum numbers [3,4]. As a consequence, the coupling strength between localised magnetic moments and delocalised field quanta is of the order of the emission probability for magnetic dipole radiation and therefore is weak. Since absorption probability is correspondingly weak, the mean free path of the GSW bosons is large. GSW bosons therefore

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propagate to a good approximation ballistic and resemble a gas confined to the volume of the sample. GSW bosons can be viewed as long distance exchange particles between spins. Due to their large mean free path GSW bosons average over all structures with dimensions smaller than their mean free path. As a consequence the ratio between mean free path of the bosons and the length scale of the morphological structures is decisive for the observed universality class. This is in contrast to the atomistic length scale of spin wave theory. On atomistic length scale grain boundaries and mosaic structures are nearly not important. Spin wave theory gives reasonable descriptions of the material specific magnon dispersions. In other words, we have to distinguish between the local excitations due to the discrete atomistic structure (magnons) and the global excitations of the magnetic continuum (field bosons). It should be noted that in spite of exchange interactions to nearest neighbours only the dispersions of magnons extend until $q\rightarrow 0$ because the atomistic propagation mode from spin to spin allows the magnons to travel, in principle, over macroscopic dimensions. However, in contrast to the GSW bosons magnons get scattered at any point defect. This strongly limits their mean free path.

Universality means that the thermodynamic observables follow over a large temperature range a simple power function of the distance from critical temperature. Since $T=0$ also is a critical temperature the universal power functions at $T=0$ are power functions of absolute temperature. In ordered magnets with three-dimensional spins the two universal power functions at $T=0$ and at $T=T_c$ overlap and give complete description of the order parameter for all temperatures. In other words, magnons never are relevant for the dynamics of the long range ordered state. The only known exceptions with atomistic dynamics are genuine Ising magnets. Since Ising spins cannot precess they are unable to emit magnetic dipole radiation. As a consequence, the GSW boson field gets not populated and magnons remain the only relevant excitations for the dynamics. It should be noted that Ising magnets occur extremely rare in nature [9]. For all other magnets with three-dimensional spins field theories instead of spin wave theory are necessary for the explanation of the observed universal power functions of temperature [10–12].

The weak coupling between GSW boson field and spin system is very important because it thermally couples the spins to the dynamics of the field. In other words, the non relevant spin system receives its thermodynamics from the relevant boson guiding field. In this way the universal dynamics of the field becomes observable using spin sensitive methods such as elastic neutron scattering, NMR or Mössbauer spectroscopy. The spins, so to say, are indicators of the dynamics of the field.

In contrast to magnons, direct experimental observation of the GSW boson field is very difficult. As experiments on standing magnetic waves in thin ferromagnetic films indicate [13], in 3D magnets with half-integer spin the dispersion of the GSW bosons is a linear function of wave vector. Linear dispersion is in contrast to the quadratic magnon dispersion predicted by spin wave theory and proves that the standing waves are resonating GSW boson states and not magnons. Linear dispersion means that the GSW bosons are mass less. Mass less particles have neither charge nor magnetic moment and therefore are invisible to neutrons. It is much surprising that field particles without magnetic moment are able to control the dynamics of the spins. For all other universality classes the dispersion of the GSW bosons seems to be non linear [13].

Ballistic propagating field particles have dispersion relations that are a simple power function of wave vector for all energies. In particular the excitation spectrum is generally continuous, i.e. gap less. This results into universal power functions of absolute temperature for the thermal decrease of the magnetic order parameter also for magnets with large magnon gap. Note that for magnets with magnon gap spin wave theory predicts exponential function of temperature for order parameter and magnetic heat capacity.

It is important to note that the dimensionality of the boson field is fundamentally different from the dimensionalities known from spin wave theory. Since the field is the relevant excitation spectrum the dimensionality of magnets has to be characterised by the dimensionality of the field. In spin wave theory it is distinguished between the dimensionality of the exchange interactions, the dimensionality of the spin and the dimensionality, or components, of the order parameter. These typical atomistic categories are unimportant for the field. We can assume that the dimensionality of the field is given by the anisotropy of the dispersion relations of the GSW bosons. One-dimensional GSW boson fields occur in axial crystals only. The anisotropy of the magnetic continuum has much similarity with the anisotropy of the elastic continuum. In the isolated magnetic domain all moments are aligned along one axis. Since GSW bosons get emitted along precession axis of the spins there are bosons travelling only along this axis. As a consequence, the boson field of the isolated magnetic domain is one-dimensional. This applies, in principle, to all saturated ferromagnets.

In order that the multi-domain bulk material has three-dimensional dynamic symmetry a dynamic averaging process over all domain orientations is necessary. We can assume that this averaging process is managed by the GSW bosons. Condition for the averaging process is that the mean free path of the GSW bosons is larger than the linear dimension of the domains. In a recent neutron scattering study of a fairly large single crystal of the cubic antiferromagnet $RbMnF_3$ we observed that the sublattice magnetisation exhibits the dynamic universality class of a one-dimensional magnet [14]. For half-integer spin of the Mn^{2+} ion ($S=5/2$) the 1D universality class is $T^{5/2}$ [3,4]. This amazing result suggests that the averaging process over all domain orientations does not work in $RbMnF_3$. The reason for this seems to be that due to the extremely weak magneto-elastic coupling the domains in $RbMnF_3$ can be assumed to be fairly large. When the linear dimension of the domains is larger than the mean free path of the GSW bosons the macroscopic sample consists of a relatively small number of decoupled domains and exhibits 1D dynamic symmetry of the isolated domain, in contrast to the isotropic magnon dispersions [15]. This illustrates that the local dimensionality of the magnons (3D) can be different from the global dimensionality of the boson field (1D).

Postulation of a dynamic averaging process over all differently oriented magnetic domains seems to be physically compelling. However, a dynamic averaging process over differently oriented grains of a powder sample is not self evident. Our neutron scattering study of a powder sample of K_2NiF_4 have yielded a critical behaviour of mean field type ($\beta=1/2$), in contrast to $\beta=0.14\pm 0.01$ reported for single crystalline material [16]. Since mean field critical behaviour is characteristic of isotropic 3D systems this observation provides strong evidence for a dynamic averaging process over all powder grains which are, of course, not in dense contact with each other as are domains in bulk material. In order to explain isotropic mean field critical behaviour of the K_2NiF_4 powder sample we have to assume that the GSW bosons are able to tunnel across the interface between neighbouring grains. This seems to be a necessary condition for the formation of an isotropic dynamic average. Tunnelling processes are not unlikely in view of the fact that the GSW bosons have much similarity with magnetic dipole radiation. The GSW boson field has radiation character! Beyond surface of the grains GSW bosons exist as evanescing waves. If the next grain is within reach of the evanescing wave tunnelling to the adjacent grain is possible. However, dynamic averaging is observed at stable fixed point (SFP) $T=T_c$ only but not at SFP $T=0$. This illustrates that the boson fields at the two SFP's are different.

At this occasion it has to be emphasised that classification of the critical behaviour in terms of atomistic models is inappropriate