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Photoacoustic and photopyroelectric investigation of the antiferromagnets CoO and Cr₂O₃ in the neighbourhood of the Neel Temperature

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Abstract : The critical behaviour of the dynamic and static thermal properties are investigated in the neighbourhood of the Néel temperature of single crystal antiferromagnetic CoO and Cr₂O₃. Photoacoustic front and pyroelectric rear detection of front illumination induced temperature oscillations are used to determine critical exponents and amplitudes of the specific heat capacity anomaly. Simultaneously measured thermal conductivity data are also presented.

1. INTRODUCTION

In a lot of materials the temperature dependence of the balance between energy and entropy causes a phase transition, characterized by a drastic change of symmetry. At the critical temperature, the essential physical parameters governing the transition become scale invariant, resulting in an infinite correlation length. Consequently, the critical behaviour of these properties is independent of the details of the interactions and can be classified in universality classes which are only marked by general system properties, such as the dimensionality of the system, the type of order parameter and the interaction range. Magnetic materials offer a favourable tool to investigate universality since a lot of theoretical models were calculated for systems consisting of spins. We have investigated the paramagnetic-antiferromagnetic transition of CoO and Cr₂O₃, which are both typical transition metal oxide antiferromagnets, however, differ in interaction properties.

2. THE ANTIFERROMAGNETIC STRUCTURE OF CoO AND Cr₂O₃

The Co²⁺ ions in a cobalt oxide crystal occupy positions in a fcc structure and possess unpaired 3d electrons which are responsible for a magnetic dipole moment. CoO is a (111)-antiferromagnet, meaning that the magnetic moments of all Co²⁺ ions in one (111)-plane are parallel and all of them are antiparallel to the magnetic moments in the next (111) plane. The antiferromagnetic order parameter consists of 12 components. The explanation for the antiferromagnetic nature of transition metal oxides was given by Kramers and Anderson [1] in the 'super exchange interaction'. Direct interaction between the strongly localized spins of metal ions is very weak. Consequently, the spin-spin interaction happens via the two 'superfluous' 2p electrons of the intermediate oxygen ion. The magnetic moments of the two metal ions orient themselves, because of Hund's rule, antiparallel to the two respective oxygen electrons which are

oriented antiparallel due to the Pauli principle. As a consequence, the metal ions have an antiferromagnetic configuration.

Cr_2O_3 is a uniaxial antiferromagnet with a complex corundum structure and an orthorhombic unit cell. Below the Néel temperature the Cr_2O_3 spins are parallel to the c-axis (111). Several theoretical attempts were made to estimate the relative strength of the nearest neighbour interaction in Cr_2O_3 . Qualitatively, one expects that the super exchange interaction (via O^{2-} superfluous electrons) is present as well as a direct spin-spin interaction via 3d orbital overlap. The indirect interaction is supposed to be weak but decays slowly compared to the direct exchange interaction. Therefore, contrary to CoO in Cr_2O_3 the spin-spin interaction is not restricted to nearest neighbours.

3. EXPERIMENTAL SETUP

3.1 Photoacoustic front detection

In order to get simultaneous information on specific heat capacity and thermal conductivity it is necessary that the thermal diffusion length μ is comparable to a characteristic length of the sample. Often, front detection photoacoustics is performed when μ is of the same order of magnitude as the optical absorption length β^{-1} . However, in the case of CoO and Cr_2O_3 , the samples are optically very opaque. For the samples we used the important characteristic length is the sample thickness l , which is comparable to μ , i.e. the samples are thermally thin. Assuming a large sample-gas effusivity ratio and $\beta^{-1} \ll \mu$, the photoacoustic pressure signal, caused by periodically amplitude modulated light ($I = I_0 \exp(i\omega t)$) at the front surface of the sample can be written as [2] :

$$P(C, k) = A_{\text{sys}} \frac{I_0(1-R)}{\sqrt{2\omega}(1+i)e_s} \frac{e^{\sigma l} + e^{-\sigma l}}{e^{\sigma l} - e^{-\sigma l}} e^{i\omega t}, \quad (1)$$

with A_{sys} a sample independent complex factor which is to be determined by a calibration procedure, I_0 the light intensity, R the sample reflectivity, ω the modulation frequency, e_s the sample effusivity, and $\sigma = (1+i)\sqrt{\omega/2\alpha_s}$, with α_s the sample diffusivity. At low enough modulation frequencies, thermal diffusivity and effusivity can be determined from amplitude and phase of the signal via iterative inversion of expression (1). At high modulation frequencies, the signal becomes independent of thermal diffusivity, and is simply inversely proportional to the sample effusivity. Contrary to the rear signal, the front signal is not exponentially attenuated with increasing modulation frequency, allowing high frequency measurements with sufficient signal. Another advantage compared to rear detection is that the dependence of the signal on diffusivity and effusivity is comparable; i.e. the calculated effusivity is only slightly dependent on the accuracy of the calculated diffusivity. Furthermore, this is a non-contact method, where other techniques need a coupling fluid.

3.2 Photopyroelectric rear detection

In this case, the temperature oscillation of a periodically front illuminated sample is detected at the sample rear surface by a pyroelectric device, which generates a current proportional to the amplitude of the thermal wave reaching the device after passing through the sample and the sample-pyro thermal coupling fluid. The complex amplitude of the signal is given by [3] :

$$I(C, k) = \frac{I_0(1-R)p}{L_p \rho_p C_p (e_s e_p^{-1} + 1)} e^{-\sigma l}, \quad (2)$$

with p the pyroelectric coefficient, L_p, ρ_p, C_p, e_p , respectively, the transducer thickness, density, specific heat capacity and effusivity.

Expression (2) can easily be inverted in order to calculate analytically the sample diffusivity and effusivity from the amplitude and the phase of the signal.

4. RESULTS AND CONCLUSIONS

According to Salomon [4] the small anisotropic tetragonal distortion of the CoO crystal structure below the Néel temperature can induce, via spin-orbit coupling, an anisotropy in the spin-spin interaction, leading to Ising behavior, at least close to the critical temperature. This is confirmed by temperature dependent specific heat capacity measurements but in contradiction with renormalization group calculations [5] (ϵ expansions around dimension 4) which predict the absence of a stable fixed point for scale transformations in parameter space, preventing a second order transition. According to Landau symmetry arguments, the absence of a second order character implies a first order character. Experimentally, a first order transition is only found for MnO and FeO [6], experimental results in literature for NiO and CoO are second order.

Fig.1: Photoacoustic data and fit curve for the specific heat capacity and thermal conductivity of CoO near the Néel temperature.

The photoacoustically determined results for specific heat capacity and thermal conductivity of a 325 μm thick CoO disk are given in Fig.1. The thermal conductivity for which high resolution literature results in the neighbourhood of a critical point are very rare shows a dip at $T_N = 15.8^\circ\text{C}$, indicating an influence of spin-spin interactions on phonon scattering. Such an influence is not surprising, since energy transfer between the spins and the lattice happens by spin-phonon interaction.

We have tried to fit the temperature dependence of the specific heat capacity by a power law; i.e., $C(T) = B + A(T_N - T)^{-\alpha}$ for $T < T_N$ and $C(T) = B + A'(T - T_N)^{-\alpha}$ for $T > T_N$, with α the critical exponent, B the background specific heat capacity and A, A' the critical amplitudes. It was impossible to fit the the

critical behaviour of our specific heat capacity data to Ising behaviour. We found a Heisenberg like specific heat critical exponent $\alpha = -0.12 \pm 0.014$. Moreover, the fitted value $A'/A=1.6 \pm 0.6$ for the critical amplitude ratio is close to the theoretically expected Heisenberg value 1.4.

Fig.2: Photopyroelectric data and fit curve for the specific heat capacity and thermal conductivity of Cr_2O_3 near the Néel temperature.

Since Cr_2O_3 is anisotropic, one can theoretically expect crossover behaviour from Heisenberg to Ising approaching T_N . Bruce et al. [7] find Heisenberg behaviour with $\alpha = -0.12$. However, they do not find the Heisenberg critical amplitude ratio and cannot confirm crossover behaviour. From ultrasonic sound velocity data, Bachelierie et al. [8] found an Ising critical exponent for the specific heat capacity. A fit of our photopyroelectrically determined data (Fig.2) results in $\alpha = -0.102 \pm 0.004$ for the specific heat capacity around $T_N = 35.38$ °C. This value is Heisenberg like, as well as the value for the critical amplitude ratio $A'/A=1.3 \pm 0.1$. However, this result which was obtained with a temperature resolution of 0.2°C does not exclude a possible crossover to Ising closer to T_N . Marinelli et al. [9] recently found crossover behavior in high resolution photopyroelectric experiments. For the thermal conductivity, we found a change in slope at T_N , similar to Bruce et al. [7].

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