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Dielectric anomaly in NaV_2O_5: evidence for charge ordering

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Abstract

We found a high-frequency dielectric and magnetic anomaly in NaV_2O_5 at the phase transition into the spin-gap state. The dielectric constant anomaly is of the antiferroelectric type, which is in agreement with the models assuming the zigzag charge ordering in the *ab*-plane.

Keywords: Charge ordering: spin gap; Low-dimensional magnetism; NaV_2O_5

1. Introduction

Two inorganic compounds, $CuGeO_3$ and NaV_2O_5, show 1D behavior of magnetic susceptibility and exhibit a phase transition into a spin-gap state. The opening of a spin gap is accompanied by the doubling of the lattice period in the magnetic chains direction. The phase transition in $CuGeO_3$ is generally considered to be a spin-Peierls transition. For some time NaV_2O_5 was considered as the second-inorganic spin-Peierls system. The 1D-magnetic structure of NaV_2O_5 was first associated with the chains of V^{4+}-ions (spin $S = \frac{1}{2}$) along the *b*-axis of the orthorombic crystal separated by the chains of nonmagnetic V^{5+} ions [1]. A ladder-type structure with equivalent V-sites was proposed on the base of new structure refinement studies of the high-temperature phase (see, e.g. Ref. [2]). The ladders are oriented along the *b*-axis with the rungs along the *a*-axis. There is one electron per rung and the spin-$\frac{1}{2}$ chains are formed by the electrons localized on V–O–V molecular orbitals on rungs and coupled by the exchange interaction along the *b*-direction. In this structure, the charge of V-ions fluctuates, its average value being 4.5.

Recent experimental data [3] as well as the theoretical arguments [4–6], suggest that the phase transition in NaV_2O_5 can be a result of some charge ordering (CO) rather than the spin-Peierls instability. Two kinds of CO were considered, of a ferroelectric [4] and an antiferroelectric [5,6] type (V^{4+}-chains or V^{4+}-zigzags, respectively). The chain model suggests an extra mechanism of a spin-gap opening (e.g. spin-Peierls transition). The opening of a spin gap in zigzag scenario is a direct CO consequence because of the exchange alternation. To confirm or reject the CO scenario we studied the dielectric constant at microwave and far-infrared frequencies.

2. Results and discussion

Step-like anomalies in real parts of the dielectric constants ε_a and ε_c where found while ε_b remained constant near $T_c = 35$ K (see Fig. 1). The analogous anomaly for ε_a was detected in Ref. [7] at the frequency 1 KHz, while a λ-type anomaly was observed for ε_c. This discrepancy may be ascribed either to weak conductivity of the

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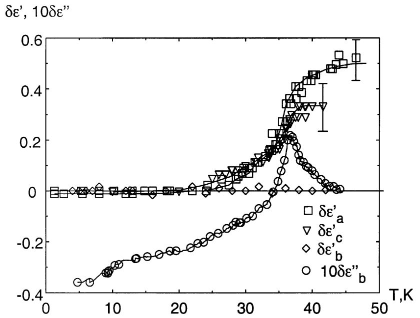

Fig. 1. Temperature dependences of changes in real and imaginary parts of the dielectric constants ε_i', ε_i'' at the frequency 36 GHz.

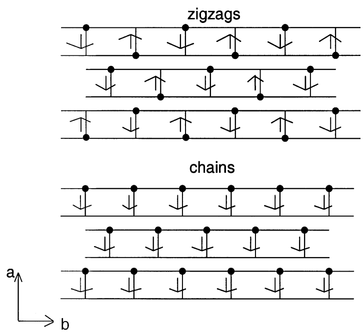

Fig. 2. Schematic form of the chain-type (a) and zigzag-type (b) charge ordering in NaV_2O_5 according to Ref. [5]. Solid lines represent paths of electron hopping in ladders, circles mark maxima of the electron density in the ordered state. The directions of the local dipoles on the rungs of the ladders are marked by arrows.

sample which could affect 1 kHz-results, or to relaxation effects in CO state that provide a frequency-dependent ε-value.

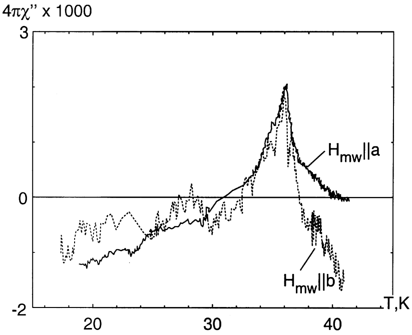

Fig. 3. 36 GHz magnetic losses versus temperature for two polarizations of microwave magnetic field.

The observed step-like anomaly in ε_b is in agreement with the zigzag CO and contradicts the ferroelectric chain structure. Schemes of both CO configurations are shown in Fig. 2. A surprising feature of the microwave magnetic susceptibility is a strong anomaly in magnetic losses near T_c (Fig. 3), indicating a coupling between the charge and spin degrees of freedom. Details of the experimental procedure and of the model justification are described in Ref. [8].

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