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High-frequency dielectric and magnetic anomaly at the phase transition in NaV_2O_5

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We found anomalies in the temperature dependence of the dielectric and magnetic susceptibility of NaV_2O_5 in the microwave and far-infrared frequency ranges. The anomalies occur at the phase transition temperature T_c, at which the spin gap opens. The real parts of the dielectric constants ϵ_a and ϵ_c decrease below T_c, while ϵ_b remains constant. The change in ϵ_a is proportional to the square of the ordering parameter below T_c. The dielectric anomalies found indicate a redistribution of charges at the phase transition. The character of the dielectric constant anomalies is of the antiferroelectric type, which is in agreement with the models assuming the zigzag charge ordering in the ab plane below T_c. The anomaly of the microwave magnetic losses is probably related to the coupling between the spin and charge degrees of freedom in vanadium ladders. [S0163-1829(99)08021-2]

I. INTRODUCTION

Two inorganic compounds, $CuGeO_3$ and NaV_2O_5, were extensively studied as quasi-one-dimensional (1D) magnets containing the chains of spins $S=\frac{1}{2}$ coupled by the antiferromagnetic Heisenberg exchange. Both materials show 1D behavior of magnetic susceptibility at high temperatures and the phase transition into the spin-gap state at low temperatures. Furthermore, the opening of the spin gap below T_c is accompanied by lattice deformations and, in particular, by the doubling of the lattice period in the chain direction.

The phase transition in $CuGeO_3$ is generally considered to be a spin-Peierls transition. The driving force of this transition is the collective spin-lattice instability with the gain in the energy of the spin chains exceeding the loss in the lattice energy.[1,2] The experimental data on $CuGeO_3$ are in a good agreement with theoretical results for antiferromagnetic spin-$\frac{1}{2}$ chains coupled to the lattice.[3] Some deviations from the ideal spin-Peierls behavior found for $CuGeO_3$ are also well understood. They are related to considerable interchain interactions (the interchain exchange is about 0.1 of the intrachain exchange[3]) and the strong next-nearest-neighbor exchange (about 0.3 of the nearest-neighbor exchange[4]).

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 $S=1/2$) along the b axis of the orthorombic crystal separated by the chains of nonmagnetic V^{5+} ions.[5,6] This structure with inequivalent vanadium sites was recently questioned in Refs. 7–10. On the basis of the structure refinement studies of the high-temperature phase of NaV_2O_5 the ladder-type structure with equivalent V sites was suggested.[7,8] The ladders are oriented along the b axes 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,[11] as well as the theoretical arguments,[12–14] suggest that the phase transition in NaV_2O_5 can be a result of some charge ordering rather than the spin-Peierls instability. Two kinds of such an ordering were considered. In the model of Ref. 12 the electrons below T_c become localized on one leg of each ladder, as in the initially proposed high-temperature structure.[5] In this model one has to invoke an extra mechanism to account for the spin gap at low temperatures. In the second model[13,14] the electrons (or the V^{4+} ions) form zigzags on each ladder. The opening of the spin gap in this scenario is a direct consequence of the charge ordering. The optical spectra of NaV_2O_5 (Refs. 15,16) were first interpreted as an indication of the presence of inequivalent V sites even at room temperature. However, these spectra could also be explained by the presence of only a short-range charge order with relatively slow charge fluctuations. An extra confirmation that the phase transition in NaV_2O_5 is not an ordinary spin-Peierls transition comes from the study of thermal conductivity: in contrast to the spin-Peierls system $CuGeO_3$, the thermal conductivity of NaV_2O_5 has a huge anomaly below T_c, which can be naturally explained by the charge ordering.[17]

Further information about the behavior of the charge subsystem close to the phase transition point can be gained by studying the dielectric constant. Measurements of the dielectric constant at the frequency of 1 KHz along three principal directions were performed in Ref. 18. An anomaly with a strong anisotropy with respect to the electric field orientation signifying the rearrangement of the charge in NaV_2O_5 has been found.

In this paper we present combined measurements of the real and imaginary part of the dielectric and magnetic susceptibility of NaV_2O_5 at the microwave frequency of 36 GHz and of the refractive index in the far-infrared range. For comparison the measurements of the same parameters were performed also for $CuGeO_3$. Our results on NaV_2O_5 confirm the presence of strong anomalies in the dielectric constant along the a and c directions, although the type of the anomaly for the c direction is different from the one found in Ref. 18. The anomaly in ϵ_a is naturally explained by the picture of the zigzag charge ordering suggested in Ref. 13,14. Possible reasons for the anomaly in ϵ_c are discussed and further experiments to check this picture are suggested.

II. EXPERIMENT

We studied the complex microwave dielectric and magnetic susceptibilities by measuring the temperature dependence of the resonance frequency and of the Q factor of the microwave TE_{104} cavity of the size $20\times7.2\times3.4$ mm. The resonance frequency f of the empty cavity is 36 GHz. The presence of a sample shifts the resonance frequency of the cavity. When the sample in the form of a thin plate is placed at the maximum of the microwave electric field $\mathbf{E}_{mw}$, with the plane of the plate parallel to the electric field, the resonance frequency of the cavity should be shifted by the value δf:

$$\frac{\delta f}{f}=-2\frac{(\epsilon'-1)v}{V}. \tag{1}$$

Here v,V are the volumes of the sample and of the cavity, respectively, ϵ' is the real part of the dielectric constant.

When the sample plane is oriented perpendicularly to the field $\mathbf{E}_{mw}$, the shift of the frequency is given by the relation:

$$\frac{\delta f}{f}=-2\frac{(\epsilon'-1)v}{\epsilon' V}. \tag{2}$$

In this case the frequency shift is smaller due to the depolarization factor effect. Formulas (1) and (2) are valid in the quasistatic approximation when the sample is small in comparison with the length of the electromagnetic wave.

The imaginary part of the dielectric constant ϵ'' affects the Q factor and results in a diminishing of the microwave power U transmitted through the cavity. The relative change of U is given by the relation:

$$\frac{\delta U}{U}=-4Q\epsilon''\frac{v}{V}. \tag{3}$$

The analogous relations could be used to determine the changes of relative magnetic permeability, $\mu=\mu'+i\mu''$, in the case when the sample is placed at a maximum of the microwave magnetic field.

The sample used for the microwave susceptibility measurements had the dimensions $1\times1\times0.3$ mm and, thus, was much smaller than the half of the length of the standing electromagnetic wave in the cavity (5 mm). This fact enables us to separate the magnetic and dielectric susceptibilities by positioning the sample at different points of the cavity. The a and b directions were lying in the plane of the plate, the c axis being perpendicular to the plate.

The change of the refractive index n with temperature was measured in the frequency range between 1.2 and 3.0 THz by registrating the interference pattern at different temperatures in the transmittance spectra of thin (14–110 μm) plates of NaV_2O_5 single crystals cleaved perpendicular to the c direction. The spectra were measured using the BOMEM DA3.002 Fourier transform spectrometer at a resolution of 0.2–2.0 cm^{-1}. The incident light was polarized either along the a axis ($\mathbf{E}\|a$) or along the b axis ($\mathbf{E}\|b$). We have also performed $\mathbf{E}\|c$ measurements at room temperature.

The position ν_m (in wave numbers) of the mth maximum in the transmittance spectrum is given by the expression

$$\nu_m=\frac{m}{2dn}, \tag{4}$$

where d is the thickness of the plate. We determined the three principal values of the refractive index along the a, b, and c axes by measuring the distance between the adjacent maxima of the interference pattern.

The relative changes of the dielectric constant ϵ' were found from the measured shifts of the interference maxima according to the relation

$$\frac{\delta\epsilon'}{\epsilon'}=2\frac{\delta n}{n}=-2\frac{\delta\nu_m}{\nu_m}. \tag{5}$$

Single crystals of stoichiometric NaV_2O_5 used in our experiments have been obtained by a melt growth method using $NaVO_3$ as a flux. The details of the growth procedure are described in Ref. 19. Our samples were of the same growth procedure as used in Ref. 20. The temperature T_c of the phase transition obtained from the beginning of the magnetic susceptibility drop is 36 ± 0.5 K.

III. EXPERIMENTAL RESULTS

A. Microwave measurements

The shift of the resonant frequency of the cavity produced by the sample at $T=40$ K was of about 400 MHz both for $\mathbf{E}_{mw}\|b$ and $\mathbf{E}_{mw}\|a$. It corresponds to the dielectric constant values $\epsilon'_{a,b}=12\pm1$.

For $\mathbf{E}_{mw}\|c$, when the microwave electric field is perpendicular to the plate, the shift of the resonator frequency was only 40 ± 15 MHz. Either the small value of ϵ'_c or the depolarization effect could result in this frequency shift which is much smaller than in the case of $\mathbf{E}_{mw}\|a$ or $\mathbf{E}_{mw}\|b$. Thus, the

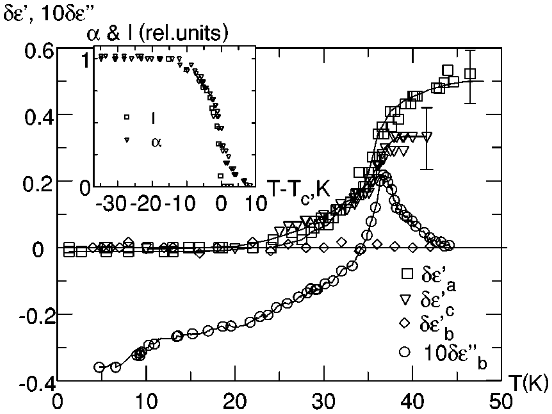

FIG. 1. Temperature dependence of the real and imaginary parts of the dielectric constant at the frequency 36 GHz. The inset demonstrates the comparison of the measurements of $\delta\epsilon_a'$ (present work) and of the intensity I of the x-ray reflexes (Ref. 25): $\alpha=1-\delta\epsilon_a'(T)/\delta\epsilon_a'(45K)$.

absolute value of ϵ_c' could not be estimated accurately enough: following the formula (2) we obtain the value ϵ_c' between 4 and infinity.

However, the frequency shift of the resonator at changing temperature can be measured with the accuracy of about 1 MHz and hence the relative change of ϵ_c at the phase transition was measured quite accurately. We used the value $\epsilon_c'=8$ obtained in quasistatic measurements[18] and in the far-infrared experiments described below, for the calculations of $\delta\epsilon_c$ according to the formula (2).

The temperature dependence of the change of the dielectric function was derived from the dependence of the resonator frequency shift with respect to the frequency at $T=1.5$ K. The value of the change of the imaginary part was derived from the temperature dependence of the transmitted signal relative to the reference value of this signal at 40 K.

The temperature dependence of the change of the dielectric susceptibility is shown in Fig. 1. The real part of the dielectric constants $\epsilon_{a,c}'$ decreases below the phase transition temperature, while the real part of ϵ_b does not show any observable change. The imaginary part of ϵ_b exhibits a well-pronounced peak. The change of the imaginary part of ϵ_a and ϵ_c were found to be smaller than the apparatus noise level.

A change in the response of the sample to the microwave magnetic field was only found for the imaginary part of the magnetic susceptibility. These results are shown in Fig. 2.

We reexamined the $CuGeO_3$ crystals studied in Refs. 21,22 and have not found any changes of the dielectric and magnetic susceptibilities of comparable values.

B. Far-infrared measurements

For the frequency of 1.2 THz we have obtained the following values of ϵ' at room temperature: $\epsilon_a'=15.0\pm0.6$, $\epsilon_b'=10.2\pm0.2$, $\epsilon_c'=7.5\pm0.2$. These data are close to those for ϵ' at zero frequency found earlier from the fitting of the reflectivity spectra of NaV_2O_5 single crystals by the model of independent oscillators.[23]

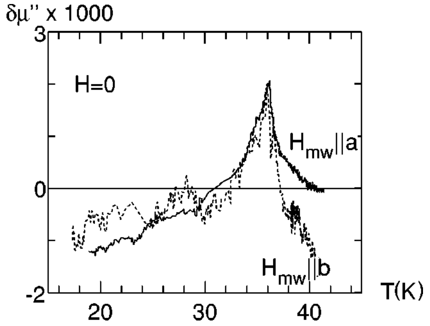

FIG. 2. Temperature dependence of the change of the imaginary part of the magnetic permeability at the frequency 36 GHz.

Figure 3 shows the temperature dependence of the relative change of the refractive index at different far infrared frequencies for the electric field polarized along the a and b axes. Simultaneously with the decrease of the refractive index n_a, the absorption present in the low-frequency region of the far-infrared spectrum of NaV_2O_5 above T_c decreases markedly when temperature decreases below T_c (see the inset of Fig. 3 and Ref. 16). We checked that despite these changes of the absorption coefficient, Eq. (5) allows us to determine the changes of n_a with sufficiently good precision. The relative change of the refractive index is of about 0.05 and corresponds to the change of the dielectric constant that is approximately twice as large as the value found at the microwave frequency of 36 GHz.

We have not found any observable changes of ϵ_b' (or n_b). Unfortunately, we could not observe the interference pattern for $\mathbf{E}\|c$ polarization at low temperatures and, hence, to detect the possible changes of ϵ_c'.

IV. DISCUSSION

The observed dielectric anomalies could, in principle, arise from (i) an anomalous thermal expansion of NaV_2O_5 at T_c,[24] (ii) a change of the lattice polarizability, and (iii) a rearrangement of electrons below T_c.

The change of the sample size at the phase transition was observed by studying the thermal expansion.[24] The relative change of the sample size as temperature passes through the

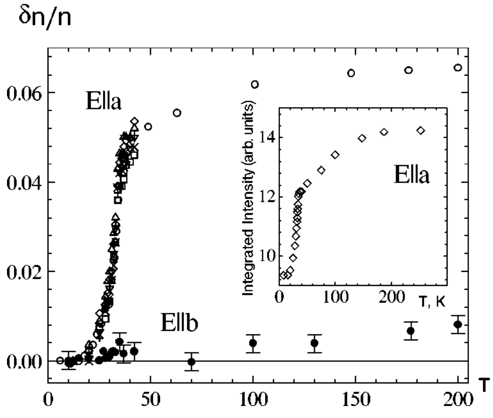

FIG. 3. Temperature dependence of the relative change of the refractive index n_a at the frequency 1.1 THz (□), 1.4 THz (△), 1.9 THz (∇), 2.2 THz (◇), 2.6 THz (○), 2.8 THz (×), 3.1 THz (+), and n_b at 3.1 THz (●). Inset presents the temperature dependence of the integrated intensity of the far-infrared absorption $I=\int_{1.0\text{ THz}}^{12\text{ THz}}\beta(\nu)d\nu$, β being the absorption coefficient.

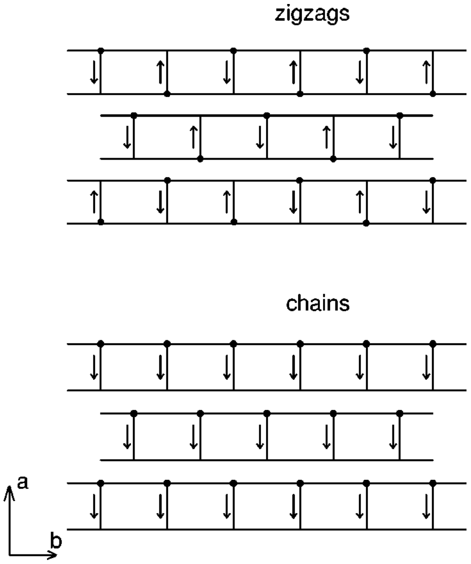

FIG. 4. Schematic form of the chain-type (bottom) and zigzag-type (top) charge ordering in NaV_2O_5 according to Ref. 13. Solid lines represent the possible paths of the electron hopping in ladders, circles mark maxima of the electron density in the ordered state. The directions of the local dipole moments appearing on the rungs of the ladders are marked by arrows on the left from the rungs.

transition point is of the order of 10^{-4}. This change of the size of the sample could produce the changes in the observed values of dielectric constant and of the refractive index of the same order of magnitude. The anomalies found in our experiments are at least 2 orders of magnitude larger. Thus the reason (i) could not result in the dependences shown in Figs. 1 and 2.

As for (ii), the changes in the lattice polarizability should be accompanied by the corresponding changes in the phonon spectrum below T_c.[16] Our estimates show that the new modes that appear below T_c cannot account for the observed change of ϵ' due to their relatively small oscillator strengths.

Our data show that the phase transition in NaV_2O_5 is marked not only by the opening of a spin gap and by the lattice period doubling, but also by a strong dielectric anomaly. This dielectric anomaly indicates that the charge subsystem plays an important role in the phase transition and agrees with the conclusion about the charge ordering at T_c. Therefore, we believe that the models of the structure[7,8] that do not consider a charge ordering and which explain the opening of the spin gap as a result of the spin-Peierls transition are not adequate. Note that the spin-gap opening in $CuGeO_3$, which seems to be a conventional spin-Peierls material, is not accompanied by changes in dielectric susceptibilities, as follows from our measurements.

Furthermore, analyzing the form of the temperature dependence of ϵ, one can choose an appropriate model of the charge ordering. The model with a localization of electrons on one leg of the ladder[12,15] (see Fig. 4, bottom) with a formation of chains of localized electrons corresponds to the ferroelectric ordering with the spontaneous electric polarization along the *a* axis. In this case the peak in the temperature dependence of ϵ_a' should be observed. Since the anomalies found in the real part of the dielectric constant have the form of steps (typical for the antiferroelectric transition) rather than peaks, we conclude that the charge ordering occurs without generation of a macroscopic dipole moment. Thus the experimental data agree with the antiferroelectric charge ordering in the form of the zigzag distribution of V^{4+} ions as described in Refs. 13,14 (see Fig. 4, top).

The zigzag models developed in Refs. 13,14 result in the identical type of the charge ordering but differ in the mechanism of a spin-gap opening. In Ref. 13 the spin-gap opening is related to the alternation of the exchange integral along the ladder that results from the charge ordering, while in Ref. 14 the spin gap is explained by the formation of singlet pairs in the ordered phase due to interladder interactions. The present experimental data cannot provide arguments to choose between these two interpretations.

The concept of the zigzag charge ordering enables one to explain most general features of the observed data. The decrease of the dielectric constant ϵ_a' can be attributed to the shrinking of the electron clouds along the *a* direction due to the localization of electrons on the distinct V sites, instead of being smeared along the V-O-V orbitals. The observed diminishing of the optical absorption coefficient in the frequency range studied (1.2–3.0 THz) below T_c is probably also connected with the above-mentioned shrinking of the electron clouds, the absorption itself being of an electronic origin (see, in particular, Ref. 23).

The coupled spin and charge degrees of freedom in vanadium ladders were described in Ref. 13 using the spin-isospin Hamiltonian. The isospin concept is introduced for the description of a charge ordering: isospin $\tau_i^z=+1/2$ corresponds to the electron localized on one end of the rung *i*, while $\tau_i^z=-1/2$ describes the electron localized on the other end. A charge ordering would result in the formation of the dipole moments d_i^z along the *a* direction on the rungs, with $d_i^z \propto \tau_i^z$. The zigzag ordering corresponds here to τ^z antiferromagnetism. The coupling of an external electric field along the *a* axis to the local (rung) order parameter is given by the term $-E_z\tau_i^z$, similar to the coupling $-H_z s_i^z$ in magnetic systems, and the resulting behavior of ϵ_a' should be similar to the behavior of the parallel magnetic susceptibility, $\chi_\parallel$, in an antiferromagnet. This consideration is in a qualitative agreement with the experimental data shown in Fig. 1.

Using the Landau mean-field approach near T_c, one can show that the decrease of the dielectric susceptibility is proportional to the square of the charge order parameter, i.e., the value of $\delta\epsilon_a'$ is proportional to the intensity of the additional x-ray reflections that appear at T_c. As shown in the inset of Fig. 1, there is, indeed, a good correlation below $\sim(T_c-2$ K) between $\delta\epsilon_a'$ and the x-ray intensity measured in Ref. 25. There is some deviation of the data close to and above T_c—the value of $\delta\epsilon_a'$ does not vanish at T_c and shows a tail up to $T-T_c\approx 8$ K. This deviation above T_c can naturally be ascribed to the existence of short-range charge ordered areas which are fluctuating in some frequency range. These fluctuations give rise to the dynamic dielectric susceptibility above T_c, but are averaged out and vanish when the susceptibility is measured on a large time scale as it was observed at 1 KHz in Ref. 18.

The nature of the behavior of the dielectric constant ϵ_c' is less clear, especially in view of the difference in the anomaly of ϵ_c' obtained in our measurements (see Fig. 1) and in the quasistatic measurements of Ref. 18, where the λ-type anomaly was observed for ϵ_c' instead of the steplike behavior shown in Fig. 1.

Both these measurements show the appearance of local dipole moments along the *c* direction at the transition temperature. The possible origin of such moments may be, e.g., the shifts of the V ions within O_5 pyramids along the *c* direction. The charge ordering (localization of the electrons on particular V ions) accompanied by the shifts of these ions along the *c* direction can modify the local dipole moments in the *c* direction.

From the crystal structure of NaV_2O_5 one would then again expect an antiferroelectric ordering of such dipoles, which would give the behavior of ϵ_c' consistent with the one observed in the present work (see Fig. 1). The reason for different type of the ϵ_c' anomaly observed in our measurements and in quasistatic measurements of Ref. 18 is not clear at present. A possible explanation might be related to the small but measurable conductivity of NaV_2O_5 single crystals. The conductivity has a large peak at T_c: it is 1.5 times higher than the conductivity both below and above T_c.[26] The conductivity contributes only to the imaginary part of the microwave dielectric constant and at 40 K the contribution is of the order of 10^{-7}. Thus, the conductivity is too small to affect the results of the microwave measurements. However, the influence of this small conductivity on the results of the electric capacity measurements at 1 KHz (Ref. 18) could be significant, because the observed value of conductivity results in the capacitor discharge time of about 10^{-3} s. Therefore, the electric-field potential within the capacitor might be disturbed by a leakage current and this fact might result in an additional change of the capacity. Further experiments are needed to elucidate this point and also to check whether the anomalies in ϵ_c' are indeed related to the shifts of V ions along the *c* axis below T_c as argued above.

The picture of the phase transition in NaV_2O_5 as being mainly of the charge-ordering type with some degree of a short-range order persisting above T_c is also qualitatively consistent with the behavior of the dielectric losses ϵ'' shown in Fig. 1. There should definitely exist slow enough charge fluctuations close to T_c, which would give rise to the dielectric losses. Why the anomaly in ϵ'' is most pronounced for the electric field directed along the *b* axis is not clear yet.

Correspondingly, a coupling of the charge (and lattice) degrees of freedom to spins, definitely present in this compound, should result in magnetic losses, which were indeed observed (see Fig. 2). This coupling originates from the Pauli principle, which relates the symmetry of the spatial and spin parts of the wave function of two exchange-coupled electrons from the neighboring rungs of the ladder.[13] As a result, triplet excitations on the ladder rungs could carry a dipole moment.[15]

Note that the peak in the imaginary part of the magnetic susceptibility is of significant value and exceeds several times the value of the real part of the susceptibility at T_c. Therefore, the fluctuations of the spin gap alone could not explain the peak of magnetic losses and such a peak is indeed not observed in $CuGeO_3$, where the charge ordering is absent.

V. CONCLUSIONS

The anomalies of the dielectric and magnetic susceptibility in the microwave and far-infrared frequency range were found in NaV_2O_5. The pronounced dielectric constant anomaly and the peak in magnetic losses at T_c indicate that the nature of the spin-gap opening in NaV_2O_5 is different from that of $CuGeO_3$. These anomalies are in agreement with the zigzag charge-ordering scenario of the phase transition in this material,[13,14] while the models that do not involve a charge ordering below T_c (Refs. 7,8), as well as the models with the ferroelectric charge ordering[12,15] can be rejected.

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