

A study of magnetoelectric activity in the antiferromagnet Nd_2CuO_4

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In this paper we investigate the high-frequency and static magnetoelectric properties of the antiferromagnet Nd_2CuO_4 , specifically the effect of an electric field on its microwave magnetic susceptibility and the shift such a field produces in its antiferromagnetic resonance frequency. We find that the first effect is strongly dynamic in character, i.e., the influence of an electric field on the static susceptibility is considerably smaller than it is in the microwave region. Although a linear magnetoelectric effect is possible due to a postulated structural distortion of the tetragonal lattice, we did not observe such an effect.

1. INTRODUCTION

Nd_2CuO_4 has an unusual noncollinear antiferromagnetic structure of the “planar cross” type with an ordering temperature of $T_N = 250$ K for the copper-ion spins.^{1,2} This structure can be represented as two interpenetrating collinear antiferromagnets whose antiferromagnetism vectors are perpendicular to one another. Stabilization of this structure can arise either from the exchange interaction of four magnetic ions³ or from the relativistic interactions that determine the anisotropy. In the second case, the structure of the crystal depends to a considerable degree on the magnetizations of the sublattices formed by Nd^{3+} ions.⁴ The magnetic subsystem of Nd^{3+} ions exerts a polarizing effect on the copper sublattices. In addition, the ions of neodymium are coupled to one another by the exchange interaction, which is much weaker than for the copper ions. The ordering of the neodymium subsystem caused by this interaction takes place at a temperature around 1.7 K (Ref. 5), although the polarization of the neodymium sublattices is appreciable even at temperatures lower than 10 K (Ref. 6).

To first approximation, the crystal structure of Nd_2CuO_4 is described by a tetragonal body-centered lattice with two formula units per unit cell. In Ref. 1 neutron-diffraction reflections were observed at temperatures above T_N , corresponding to a rather small structural displacement of the crystal lattice. This distortion corresponds to a shift of the copper ions from the vertices of the basis square, so that the unit cell is doubled in two mutually perpendicular directions. In this case, the magnetic structure loses the element of inversion symmetry, although the symmetry of the lattice after the distortion remains tetragonal. This distortion of the lattice makes it possible for a linear magnetoelectric effect to exist, i.e., the expansion of the thermodynamic potential is augmented by terms of the form $\beta_{ik} E_i H_k$.

In Ref. 3 a linear magnetoelectric effect was predicted for magnetic structures of the “planar cross” type when the lattice is distorted in this fashion. Although manifesta-

tions of linear magnetoelectric effects known up to now are associated with relativistic interactions, the effect can also arise from the exchange interaction of four ions, in which case the decisive factor for the existence of the magnetoelectric effect is the presence of the structural distortion which eliminates inversion as a symmetry operator. Note that the presence of such lattice distortions was disputed in Ref. 7.

In a previous paper⁸ we observed a dynamic magnetoelectric effect in our study of antiferromagnetic resonance. We inferred this effect from the excitation of one of the antiferromagnetic resonance modes by a high-frequency electric field. This magnetoelectric activity could be due either to the presence in Nd_2CuO_4 of the relativistic magnetoelectric effect referred to above, as was predicted in Ref. 8, or to the noncentral position of the magnetic Nd^{3+} ions.⁴ Because ions at such a position are subject to an electric crystal field component that is odd with respect to the coordinates, the sample can exhibit a dynamic dielectric response at the magnetic resonance frequencies corresponding to resonance of the neodymium subsystem.⁹

In this paper we continue our study of the static and dynamic magnetoelectric properties of Nd_2CuO_4 , including a search for the exchange-induced linear magnetoelectric predicted in Ref. 3 and for shifts in the magnetic resonance frequency in an electric field. Studies of the static and dynamic magnetoelectric properties complement one another, since observation of a shift in the antiferromagnetic resonance frequency in an electric field allows us to distinguish the effect of an electric field on the anisotropy constant from its effect on the g factor. For example, Kita *et al.*¹⁰ measured the modulus of the static magnetoelectric effect along with the shift in antiferromagnetic resonance frequency in an electric field for the antiferromagnet Cr_2O_3 , thereby allowing the different contributions to the magnetoelectric effect to be distinguished.

2. EXPERIMENTAL METHOD

In order to study the effect of a static electric field on antiferromagnetic resonance and microwave magnetic susceptibility, we placed a sample of Nd_2CuO_4 in a rectangular resonator with dimensions $20 \times 7.2 \times 3.4$ mm at the location of the maximum microwave magnetic field for the TE_{014} mode. The frequency of this mode was 36 GHz for our resonator. An electric field was applied to the sample by using a strip of copper foil with thickness 0.1 mm and dimensions 1.5×4 mm², located within the resonator at a distance 1 mm from the wall with dimensions 20×7.2 mm². The sample was placed between the copper foil and the wall of the resonator, and was electrically isolated from both surfaces. Using a thin insulated wire passed through an aperture in the end wall of the resonator, we applied potentials up to 200 volts to the copper strip with respect to the resonator wall. The copper strip and the feed wire were mounted in such a way that their presence did not disrupt the distribution of fields of the TE_{014} mode. The Q of the resonator, together with the sample, the copper strip, and the wire, was 1200 at liquid helium temperatures.

A crystal microwave detector fixed the microwave power passing through the resonator and developed a voltage U proportional to this power. When the resonator is tuned to its intrinsic frequency, changes in the sample losses lead to a proportional change in the signal from the detector. When it is tuned in the wings of the resonance curve, changes in both the imaginary and real parts of the microwave susceptibility of the sample contribute to the detector signal.

Samples with dimensions $0.7 \times 1.5 \times 1$ mm³ were cut from the same single crystal of Nd_2CuO_4 that was the source of samples for the antiferromagnetic resonance study.⁸ These samples were quite small, so that the change in intrinsic frequency of the resonator due to changes in the magnetic susceptibility of the sample at the magnetic resonance frequency could be neglected (this change was small compared to the width of the resonance curve of the resonator). The edge of the sample with length 0.7 mm parallel to the [100] crystallographic axis was oriented along the electric field that appeared when a voltage was applied to the strip. It is noteworthy that our samples of Nd_2CuO_4 had a small conductivity, which prevented us from creating a static electric field. As we show below, the screening time of an external electric field by charge carriers in the sample was 15 minutes at a temperature of 1.2 K, and about 1 second at a temperature of 8 K.

In order to increase the sensitivity to small changes in the microwave susceptibility, and also to avoid the screening action of the sample conductivity, we used a modulation method. The sample was subjected to an AC electric field with amplitude E_{ac} and frequency $f_{ac} = 2.6$ kHz. The AC component of the microwave signal from the detector δU was amplified using a phase-sensitive amplifier, whose reference signal came from the voltage created by the field with amplitude E_{ac} . If the effect of the electric field on the microwave susceptibility of the sample is linear, then the signal from the detector should contain an AC component that oscillates with the frequency of the electric field f_{ac} . If

the effect of the electric field is quadratic, then AC components should appear in the signal that oscillate at double the field frequency $2f_{ac}$. We will denote the amplitude of signal oscillations at the frequency f_{ac} by α_1 and at the doubled frequency $2f_{ac}$ by α_2 .

We checked for the absence of parasitic modulation of the Q factor and the characteristic frequency of the resonator by the field E_{ac} in trials using the resonator without a sample and with a sample of the antiferromagnet MnCO_3 , in which the microwave signal used to sweep the antiferromagnetic resonance line was unaffected by the electric field.

In order to study the static magnetoelectric properties we used a SQUID magnetometer.¹¹ Here a constant electric voltage was applied directly to the sample, which was glued between two copper electrodes using a conducting glue. The dimensions of the samples used in these experiments were roughly $2 \times 2 \times 1$ mm³. The electric field was applied in the [100] and [001] directions. By using two modifications of the electrodes we were able to measure the change in magnetic moment of the sample both parallel and perpendicular to the electric field. Using a solenoid that surrounded the samples, we were able to make these measurements in magnetizing fields of up to 200 Oe. In zero magnetizing field we looked for the classical magnetoelectric effect (i.e., a magnetic moment induced by an electric field), while in nonzero magnetizing fields we looked for the second-order effect, i.e., the effect of an electric field on the magnetic susceptibility.

The axis of sensitivity of the magnetometer was parallel to the external magnetizing field. The output SQUID signal was proportional to the change in magnetic moment of the sample.

3. EXPERIMENTAL RESULTS

3.1. Change in the microwave magnetic susceptibility under the action of an electric field

We found that the electric field had an appreciable effect on the microwave magnetic susceptibility at temperatures below 10 K. When an AC electric field was applied with frequency f_{ac} , we observed oscillations in the signal from the microwave detector at a frequency $2f_{ac}$. The relative magnitude of the change of the transmitted signal was 2×10^{-5} at a temperature of 1.2 K for an amplitude $E_{ac} = 2000$ V/cm. The amplitude of the response at frequency f_{ac} was 7 to 10 times smaller and changed in an uncontrolled fashion depending on the way the sample was glued.

In Fig. 1 we show how the amplitude α_2 of signal oscillations from the detector at the doubled frequency depends on the relative detuning between the microwave oscillator frequency and the resonator. The phase of these oscillations was shifted by 180° with respect to the phase of the squared electric field intensity oscillations (i.e., the signal from the detector decreased when the electric field was applied). For a signal tuned to the resonant frequency of the resonator, at which the change in the signal is determined only by the change in the imaginary part of the

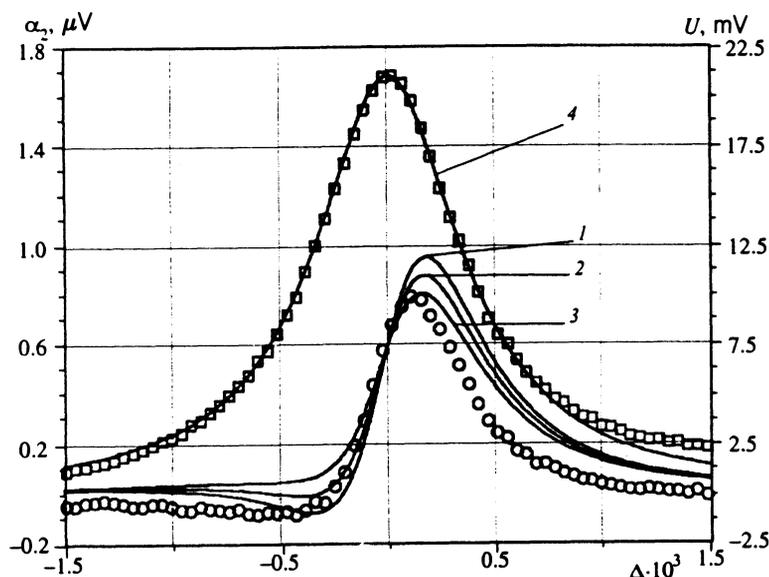


FIG. 1. Dependence of the microwave detector signal U (squares) and amplitude of its AC component α_2 (circles) on the relative detuning of the microwave oscillator and resonator. Curves 1, 2, and 3 are calculated using Eq. (1) for values of the ratio $\delta\chi''/\delta\chi'$ equal to 0.8, 1, and 1.2 respectively; curve 4 is a Lorentz curve for $Q=1200$. The quasistatic electric field with maximum intensity 2000 V/cm is parallel to the [100] axis, the microwave magnetic field is directed along [001], the temperature is 1.2 K, $f=36$ GHz, and $H=0$.

microwave magnetic susceptibility, this implies that the electric field acts to increase the magnetic losses in the sample. The resonance curve of the resonator was plotted in the course of this experiment, and was well described by a Lorentz curve corresponding to a Q of 1200. It is clear from this figure that the maximum in the response α_2 is shifted with respect to the resonance frequency f_0 of the resonator. This implies that the electric field changes not only the imaginary part but also the real part of the magnetic susceptibility. The change in the imaginary part of the magnetic susceptibility $\delta\chi''$ determines the change in magnetic losses in the sample; the impact of such changes in the imaginary part on a signal passing through the resonator is largest at the resonance frequency. The change $\delta\chi'$ in the real part of the susceptibility when an electric field is applied causes a shift in the intrinsic frequency of the resonator; the impact of this factor on the signal passing through the resonator is a maximum at the steepest point on the resonance curve.

Analysis of the effect of both these factors leads to the following expression for the relative change in the transmitted microwave signal as a function of the oscillator detuning:

$$\frac{\delta U}{U_0} = \frac{16\pi Qv/V}{1+4\Delta^2 Q^2} \left[\delta\chi''(1-8\Delta^2 Q) + \delta\chi' \frac{4\Delta Q}{1+4\Delta^2 Q^2} \right] \quad (1)$$

here $\Delta = (f - f_0)/f_0$ is the relative detuning of the microwave frequency of the oscillator f with respect to the resonator frequency, Q is the Q factor of the resonator, and v and V are the volumes of the sample and resonator respectively.

Based on the value of α_2 at $\Delta=0$, we find the value of $\delta\chi'' = (2 \pm 0.2) \times 10^{-7}$ cgs. This value is approximately 10^{-4} of the value of the static susceptibility $\chi_{||}$ given in Ref. 12 and extrapolated to a temperature of 1.2 K. We then choose the value of the ratio $\delta\chi''/\delta\chi'$ that gives the best agreement between calculations based on Eq. (1) and the observed function $\alpha_2(\Delta)$. Curves 1, 2, and 3 in Fig. 1

were plotted using Eq. (1) and values of the ratio $\delta\chi''/\delta\chi'$ equal to 0.8, 1, and 1.2 respectively. Satisfactory agreement is observed when $\delta\chi'' = \delta\chi'$.

Thus, the electric field gives rise to a quadratic increase in the real and imaginary parts of the microwave magnetic susceptibility.

The temperature dependence of α_2 and α_1 at $\Delta=0$ is shown in Fig. 2. Here we plot the temperature dependence of the microwave detector signal U , from which it is clear that the magnetic losses in the temperature interval 1 to 10 K increase as the sample is cooled.

As we said previously, the presence of an intense signal at twice the frequency of the applied electric field indicates an effect on the susceptibility that is quadratic in the field. If we add a constant bias E_{dc} to the AC field with amplitude E_{ac} , then the square of the total field will contain a term that oscillates at frequency f_{ac} with amplitude $2E_{dc}E_{ac}$. Therefore, a response should appear at the fundamental frequency f_{ac} , whose value is determined by the ratio

$$\alpha_1 = 2\alpha_2 \frac{E_{dc}}{E_{ac}} \propto E_{dc}E_{ac}. \quad (2)$$

We actually observed a response at the fundamental frequency corresponding to the linear effect resulting from adding a DC voltage to the applied AC voltage. When a DC voltage was applied, a linear response α_1 appeared and decayed over a period of time that depended strongly on temperature. At a temperature of 1.2 K this time was about 15 minutes, at 4 K it was 10 seconds, and at 8 K it was less than 1 second. The magnitude of the response α_1 immediately after switching on E_{dc} was proportional to both E_{dc} and E_{ac} , and was found to be in agreement with the magnitude of the response at the doubled frequency α_2 according to Eq. (2). The time dependence of the signal α_1 at a temperature of 1.2 K after applying the DC field is shown in Fig. 3.

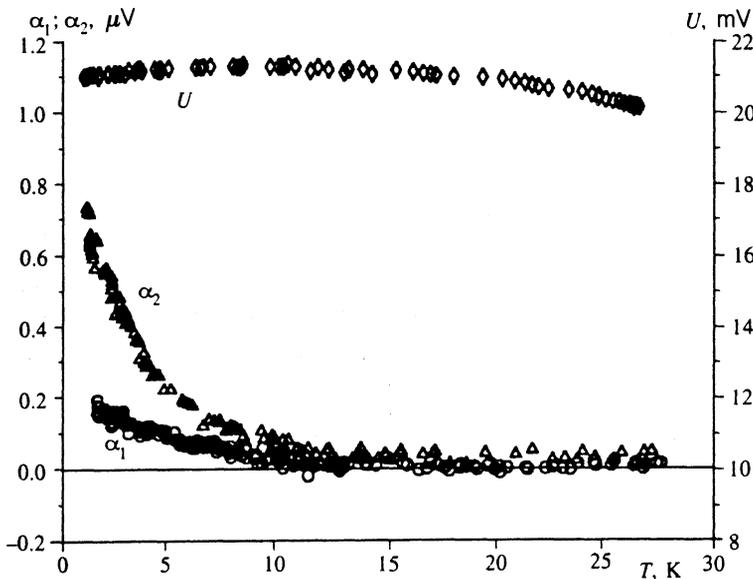


FIG. 2. Dependence of the microwave signal U and amplitude of its AC components α_1 and α_2 on temperature. $E_{ac} \parallel [100]$, the microwave magnetic field is parallel to the $[001]$ axis, $f=36$ GHz, and $H=0$.

It is natural to assume that the observed slow decay of the linear effect is connected with screening of the DC electric field by charge carriers in the sample. This assumption is also confirmed by the following observations. When a DC voltage is applied at temperatures above 15 K, and the sample is then cooled down to 1.2 K, we find that the DC electric field has no effect on the quantity α_1 . However, when E_{dc} is switched off after the cooling, a signal appears at the frequency f_{ac} with the same amplitude, but opposite in sign, to the analogous signal in experiments when E_{dc} was switched on at 1.2 K. We explain this behavior of the effect by the rapid screening of an external electric field at high temperatures, when the conductivity is rather large. In this case charge collects at the surface of the sample. As a result of cooling, the conductivity decreases and after switching off the external field the surface charge is reab-

sorbed over a rather long period of time. In the process of reabsorption, the electric field due to surface charge, which acts on the interior bulk of the sample, gradually decreases, leading to the appearance of a signal at frequency f_{ac} .

It is obvious that the observed time for the linear response α_1 to decrease is the time for screening of an external electric field. At temperatures below 10 K this time is much smaller than the period of oscillation of the field E_{ac} ; consequently, the behavior of the conductivity does not significantly affect the value of the electric field in the sample.

Our observation of a rather small linear effect in the microwave response α_1 to an electric field is probably explainable by nonuniformity of strain in the sample, which leads to a bulk polarization charge that creates a bias field E_{dc} ; as we explained above, this in turn gives rise to a linear effect.

We studied the effect of a quasistatic electric field on the high-frequency susceptibility at microwave frequencies in the interval 36 to 48 GHz. In this case we did not observe an appreciable change in the magnitude of the effect, and its sign was unchanged, i.e., at all the frequencies in this interval we observed a quadratic increase in the magnetic microwave susceptibility with the applied electric field.

3.2. The search for static magnetoelectric effects

We also made an attempt to observe the effect of an electric field on the static magnetic susceptibility and magnetization of the sample in zero external field.

We looked for changes in the static magnetization due to a DC electric field E in zero external magnetizing field and in a magnetizing field H for all five combinations of directions of the fields H and E along the $[100]$ and $[001]$ axes. In these experiments we deposited contacts on the sample, and a small electric current flowed through the sample when a voltage was switched on. Typical IV characteristics of the sample are shown in Fig. 4.

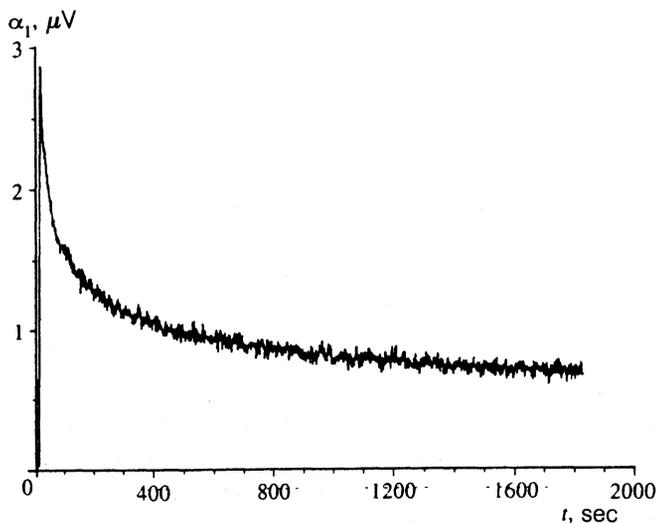


FIG. 3. Time dependence of the amplitude of the AC component of the microwave signal α_1 after switching on the constant electric field. $T=1.2$ K, $E \parallel [100]$, $f=36$ GHz, $H=0$. $E_{ac}=2000$ V/cm, $E_{dc}=3000$ V/cm.

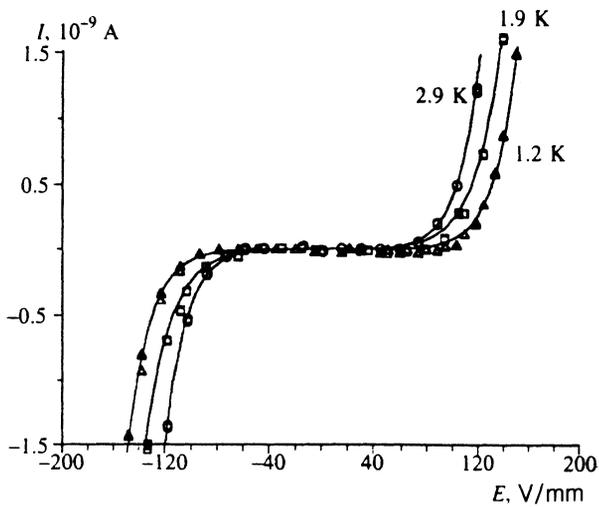


FIG. 4. Electric field dependence of the current in the [001] direction for various temperatures. The sample cross section was $1 \times 1 \text{ mm}^2$.

The dependence of the output signal from the magnetometer on the voltage applied to the sample is shown in Fig. 5 for various temperatures. Here the magnetizing field was equal to 80 Oe. For electric field intensities up to roughly 500 V/cm we observed no noticeable effect of an electric field on the magnetic moment of the sample. When the electric field was increased further, a non-zero signal appeared at the output of the SQUID magnetometer, connected with heating of the sample by the electric current flowing in it and with changes in the magnetization due to the dependence of the susceptibility on temperature. Evidence that this signal was connected specifically with sample heating was the correlation between the shape of its voltage dependence and that of the IV characteristics, as well as the change in sign of the SQUID signal as we

passed through the temperature 1.73 K. Our observations indicate that for a field directed in the ab plane the susceptibility χ_{\perp} has a peak at this temperature. (This peak in the susceptibility was also observed at 1.7 K for a polycrystalline sample.⁵) Therefore, at temperatures below the peak temperature heating leads to an increase in the magnetization in an external field; at temperatures above 1.73 K heating decreases the magnetization, which also is observed in experiment. The susceptibility χ_{\parallel} (for directions of the magnetic field parallel to the [001] axis) has a monotonic temperature behavior. In this case the sign of the SQUID signal does not change as the temperature increases for large voltages on the sample. The change in direction of \mathbf{H} leads to a change in the sign of the ordinate for all the curves shown in Fig. 5.

We monitored the signs of the derivatives $d\chi_{\perp}/dT$ and $d\chi_{\parallel}/dT$ and the peak temperature for the susceptibility χ_{\perp} based on the sign of the drift of the SQUID signal for slow heating or cooling of the sample with the magnetic field switched on.

The magnetometer noise that appears on these plots is due to the considerable degree of temperature instability and the strong dependence of the susceptibility on temperature. In the experiments with a magnetizing field, noise from the magnetometer limited the sensitivity with which we could measure the effect of an electric field on the magnetic susceptibility. Nevertheless, these data allow us to establish an upper bound on the effect of an electric field on the magnetic susceptibility: at a temperature of 1.2 K, a field of 500 V/cm produces a change in the static susceptibility that does not exceed 0.2 of the value of $\delta\chi'$ determined from the microwave experiments described above in the same electric field.

In the absence of a magnetizing field, we saw no noticeable effect of the electric field on the magnetization for electric field intensities up to 5000 V/cm. This leads to the

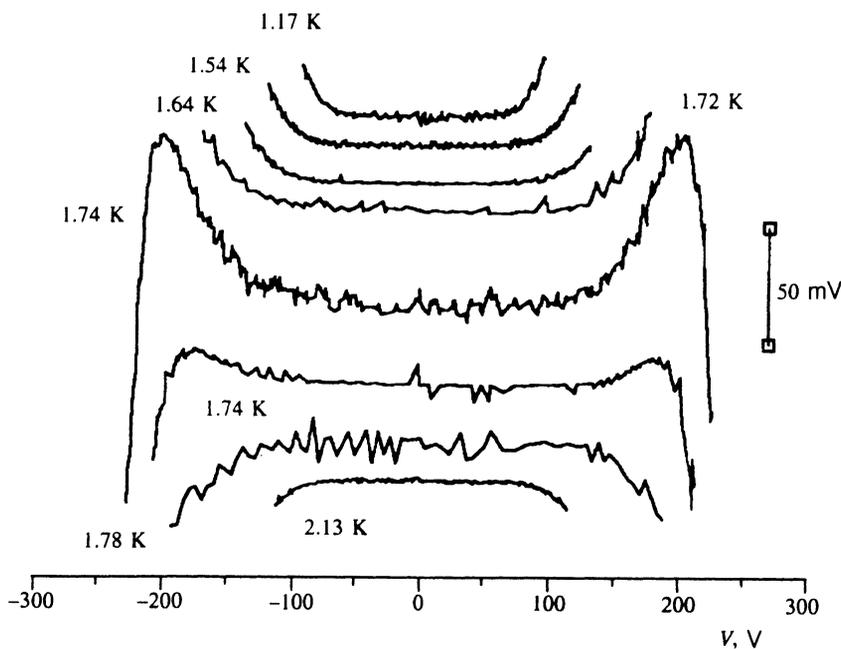


FIG. 5. Dependence of the output signal of a SQUID magnetometer on the voltage across the sample in a magnetizing field of 80 Oe for various temperatures. $\mathbf{E} \parallel [100]$, $\mathbf{H} \parallel [010]$. The size of the sample was 2 mm parallel to \mathbf{H} , 1 mm parallel to \mathbf{E} , and 2 mm in the direction perpendicular to both fields.

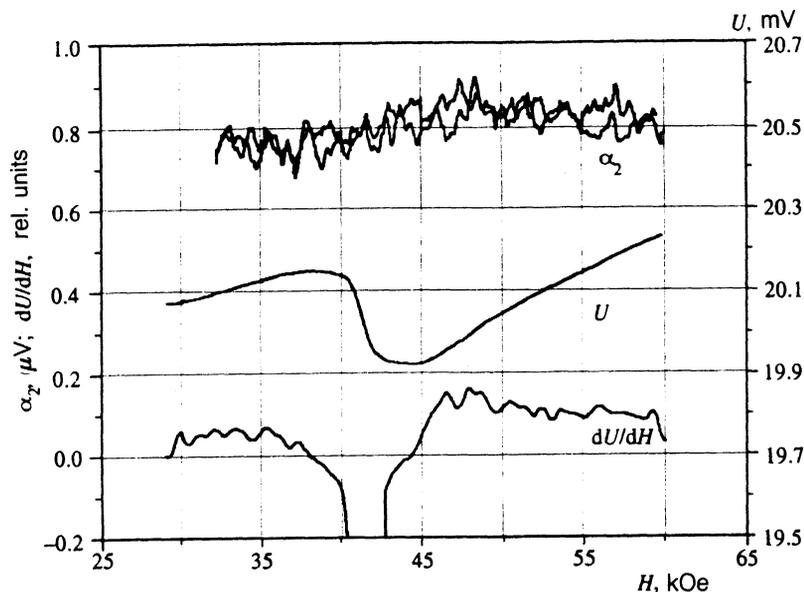


FIG. 6. Dependence of the microwave signal, its derivative with respect to magnetic field, and the AC component α_2 , on magnetic field. $T=1.2$ K, $E_{ac} \parallel [100]$, $f=36$ GHz, and the amplitude value of $E_{ac}=200$ V. $H \parallel [100]$.

following estimate for the modulus of the linear magneto-electric effect: the value of $4\pi M/E$ is at most 6×10^{-6} cgs units, which is roughly 100 times smaller than the maximum value of the analogous quantity for polycrystalline Cr_2O_3 (Ref. 13).

It is noteworthy that the effect of changes in the susceptibility due to heating by an electric current cannot explain the quadratic signal α_2 observed in the microwave experiments, which we described in Sec. 3.1. Because the IV characteristics exhibit breakdown behavior (see Fig. 4), the heating effect should have a sharply expressed threshold character. However, the signal α_1 exhibits a well-defined region of linear proportionality in its dependence on E_{ac} and E_{dc} , which indicates that the effect of the field on the microwave magnetic susceptibility is smooth rather than having a threshold.

In our experiments with the SQUID magnetometer, the changes we observed in the susceptibility for fields greater than 1000 V/cm allowed us to estimate the sample heating based on the known dependence of χ_0 on T . This amounted to no more than 10^{-2} K for $E=2000$ V/cm in the interval of temperatures investigated. Using known values of the power released in the sample and converted to heat, and the heat capacity of Nd_2CuO_4 (Ref. 14), we estimate that the thermal relaxation time of the sample is of order 0.1 second. For this relaxation time, the temperature oscillations in the sample at frequency $2f_{ac}$, which equaled 5.2 kHz, are 10^{-5} K. It is clear from the temperature dependence of the microwave signal passing through the resonator (Fig. 1) that this heating can lead to a relative change in the microwave signal of no more than 10^{-7} , i.e., much smaller than the observed value of $\delta U/U \approx 10^{-4}$. Thus, our estimates also confirm that the effect of the electric field on the magnetic susceptibility observed at microwave frequencies cannot be connected with heating of the sample by an AC conduction current excited by the field E_{ac} .

3.3. Dependence of the antiferromagnetic resonance field on electric field

In our search for evidence that the electric field affects magnetic resonance, we chose the most sharply expressed antiferromagnetic resonance line, which is observed in Nd_2CuO_4 for $H \parallel [100]$ (Refs. 8 and 15). For this direction of the field an abrupt jump occurs in the magnetization at $H=H_{srt}$ (Refs. 16 and 8). At a temperature of 1.2 K the spin reorientation field is 44 kOe. The value of H_{srt} has a sharp maximum for fields directed along [100] and decreases rapidly as the field direction changes. At a frequency of 36 GHz, the antiferromagnetic resonance field slightly exceeds H_{srt} . In the range of fields below H_{srt} the absorption of microwave power does not depend on frequency.^{8,17} Because of the difference in magnetic structure of the crystal in fields below and above H_{srt} , an abrupt discontinuity in the microwave absorption occurs in this field. When the field is scanned at a frequency of 36 GHz over fields above H_{srt} , we observe the right-hand limb of the antiferromagnetic resonance line. The left-hand limb of the line was not observed at this frequency, since the sample is in a different magnetic state in fields below H_{srt} . At higher frequencies the antiferromagnetic resonance field differs from the spin reorientation field, in which case the jumps in the absorption and the antiferromagnetic resonance line were observed separately and both limbs of the line could be scanned.^{8,15}

If the antiferromagnetic resonance magnetic field depends on electric field, we should observe an AC component in the microwave signal proportional to the derivative of the absorption intensity with respect to magnetic field when we use the modulation method with an AC field E_{ac} . Analogously, if H_{srt} depends on electric field, by rights we should expect an AC microwave signal proportional to the derivative of the function that describes the discontinuity in absorption.

In Fig. 6 we show the magnetic-field dependence of the response α_2 and a plot of the antiferromagnetic resonance line, i.e., the magnetic-field dependence of the microwave signal passing through the resonator, and also the derivative of this signal with respect to magnetic field. We see that the electric field has no effect on the absorption of microwave power in the neighborhood of the spin reorientation field. However, in the right-hand limb of the antiferromagnetic resonance line there is a contribution to the signal α_2 with a maximum value of $0.1 \mu\text{V}$ that is proportional to the derivative of the absorption intensity with respect to magnetic field. From a comparison of the value of this component of the signal and the value of the change in microwave signal as we pass through the antiferromagnetic resonance line, we find that the shift in the antiferromagnetic resonance line is quadratic with respect to electric field, positive, and comes to roughly 0.5 Oe in a field of 2000 V/cm .

4. DISCUSSION

4.1. Shift of the antiferromagnetic resonance in an electric field

The antiferromagnetic resonance mode described in the previous section is probably connected with oscillations of the spins of the Nd^{3+} ions, since its intensity falls off rapidly as the temperature increases in the interval from 1 to 3 K (Ref. 8). The Nd^{3+} ion is a Kramers ion, and the electric field can change the g factor but cannot affect the energy of the ground state in zero magnetic field. Starting from these considerations, we may conclude that the observed shift in the antiferromagnetic resonance field is caused by a decrease in the component g_{xx} of the g tensor in an electric field directed along the x axis ([100]). The change in the g factor is quadratic because we observe the signal at a frequency $2f_{ac}$. In order to estimate the magnitude of the change in the g factor we will assume that the four-sublattice antiferromagnetic structure of neodymium ions is equivalent to a two-sublattice structure in fields above H_{srt} (all the sublattices are flipped over by the perpendicular field). In this case the antiferromagnetic resonance frequency ω should be a known function of magnetic field:

$$\omega^2 = \gamma^2 (H^2 - H_{srt}^2), \quad (3)$$

where γ is the magnetomechanical ratio, which is proportional to the g factor. From this expression we find that the shift in the antiferromagnetic resonance field ΔH is connected with the relative change in the g factor by the expression

$$\frac{\delta\gamma}{\gamma} = \frac{\Delta H}{2(H - H_{srt})}. \quad (4)$$

From this we find $\delta g/g = (5 \pm 2) \times 10^{-5}$.

This observed shift in the g factor coincides in order of magnitude with shifts observed in experiments that measure the effect of an electric field on paramagnetic resonance^{18,19} in fields on the order of 1000 V/cm , and is an order of magnitude larger than it is in antiferromagnetic

Cr_2O_3 (Ref. 10). However, it should be noted that the authors of Refs. 18 and 19 observed an effect that was linear in field and associated with the odd component of the crystal electric field. Quadratic effects observed in experiments on paramagnetic resonance²⁰ are two to three orders of magnitudes smaller.

4.2. The effect of an electric field on the microwave magnetic susceptibility

In principle, we could perhaps ascribe the dependence of the susceptibility on electric field to the aforementioned effect of the electric field on the g factor. We note only that the microwave experiments involve a different field configuration: the electric field is perpendicular to the microwave magnetic field along which the susceptibility was measured. Parallel orientation of the fields was difficult for us to implement, since the strip of foil used to create the electric field ought to be placed parallel to the microwave magnetic field lines so as not to disrupt the distribution of high-frequency field in the resonator. If the measurement of the microwave susceptibility were to be explained by its effect on the g factor, then the effect of the electric field would manifest itself in changes of the static susceptibility. Since we have seen that the effect of the field on the static susceptibility is considerably smaller than its effect on the dynamic susceptibility in the microwave range (or is entirely absent), the observed effect of the electric field on the magnetic susceptibility must be essentially a dynamic effect.

We offer the hypothesis that the effect of the field on the dynamic susceptibility is connected with its effect on the spectrum of one or several spin-wave branches. Let us note first that there are appreciable magnetic losses in the range of frequencies we investigated^{8,17} that do not depend (or depend weakly) on frequency. This suggests that these losses might be connected with two-magnon processes, in which a magnon of the lower branch absorbs a microwave photon and is converted to a magnon of the upper branch. The gaps of the spin-wave branches are at 42 GHz (Ref. 8) and around 70 GHz (Ref. 15), respectively. If these frequencies depend on electric field, then the density of states of magnons that participate in these processes will also depend on electric field, and this circumstance could explain the fact that the dynamic susceptibility is more prone to changes in the electric field than is the static susceptibility.

The temperature dependence of $\delta\chi''$ coincides qualitatively with the temperature dependence of the magnetization of the neodymium sublattices.⁶ This fact suggests that the effect is associated with the neodymium subsystem in a significant way. According to Krivoruchko *et al.*,⁴ the magnetic anisotropy energy, which largely determines the magnetic structure of Nd_2CuO_4 , is determined by neodymium ions and exchange between neodymium and copper spin subsystems. Apparently, the electric field affects the energy gaps by way of this exchange interaction.

The absence of a linear magnetoelectric effect in the samples of Nd_2CuO_4 we investigated is clearly connected

with the absence of a distortion of the tetragonal structure, without which the linear magnetoelectric effect is forbidden by the crystal symmetry.

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