# Correlation of spin, orbital, and structural states in the quasi-two-dimensional Heisenberg antiferromagnet $Eu_2CuO_4$

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The dielectric susceptibility and static magnetic properties of a $Eu_2CuO_4$ crystal in external constant electric and magnetic fields are investigated experimentally. The physical situation is analyzed and the experimental data are intrepreted on the basis of the self-consistent interaction of the spin and orbital subsystems. It is shown that the orbital-glass state induced by two-dimensional Heisenberg antiferromagnetic spin fluctuations, in turn, alters the state of the spin subsystem for $T>T_N$. Phase separation, a two-dimensional weak ferromagnetic moment, and restricted regions of quasi-two-dimensional states appear in it. © *1996 American Institute of Physics.* [S1063-7761(96)02608-X]

## 1. INTRODUCTION

The correlations of spin, orbital, and structural states for magnetic crystals with Jahn–Teller $3d$ ions have been adequately studied (see the review in Ref. 1) for the most part crystals with Jahn–Teller ions having an orbital singlet ground state, in which the spin-orbit interaction is governed by the quadrupole orbital angular momenta, have been investigated.[1]

In this paper we consider the physical situation in the quasi-two-dimensional Heisenberg antiferromagnet $Eu_2CuO_4$ with Jahn–Teller $Cu^{2+}$ ions, for which the presence of a degenerate ground orbital state has been postulated. According to crystal-field theory with consideration of the local environment, when the $Cu^{2+}$ ions are found in an octahedral environment of $O^{2-}$ ions, as in $La_2CuO_4$, their ground orbital state is an orbital singlet that arises from the doublet ground orbital state $\Gamma_3(e_g)$ in the cubic lattice.[2] In the case of the tetragonal crystals of $R_2CuO_4$ (where $R$=Nd, Pr, Sm, Gd, or Eu, and the crystal structure is of the $T'$ type[3]), the $Cu^{2+}$ ions do not have an octahedral environment, and they form square lattices in $CuO_2$ layers, having only four $O^{2-}$ ions in their local environment. The closest ions to the $Cu^{2+}$ ions in neighboring layers are the rare-earth $R^{3+}$ ions, rather than apical $O^{2-}$ ions (as in $La_2CuO_4$). In this case the ground orbital state of the $Cu^{2+}$ ions should apparently not be determined by the local environment alone and has not yet been firmly established.

To explain the experimental data from observations of the orbital-glass state in $Eu_2CuO_4$ in Ref. 4, we assumed that the ground orbital state of the $Cu^{2+}$ ions is a tetragonal doublet, which arises from the triplet ground orbital state $\Gamma_5'(t_{2g})$ in the cubic lattice (the notation follows Ref. 2). As has been reported (see, for example, Ref. 2), in cases in which the spin-orbit interaction surpasses the orbital-lattice interaction, the ground cubic orbital triplet $\Gamma_5'$ splits in the tetragonal lattice into a ground orbital doublet (a tetragonal doublet) and an excited singlet state.

The hypothesis that the $Cu^{2+}$ ions are in a degenerate ground orbital state leads to a new physical situation in a crystal of $Eu_2CuO_4$ and possibly in other tetragonal crystals of $R_2CuO_4$. When the ground orbital state is chosen for the $Cu^{2+}$ ions, the mean orbital angular momentum in the ground state is nonzero, and the linear spin-orbit interaction is effective.[2]

An important feature of all the quasi-two-dimensional states constructed on the basis of $CuO_2$ planes is the fact that the value of the exchange integral $J_{ij}$ of the two-dimensional Heisenberg antiferromagnetic exchange interaction $J_{ij}(\mathbf{S}_i\mathbf{S}_j)$ exceeds the value of the spin-orbit coupling constant $\lambda$ ($\lambda\mathbf{SL}$). The exchange integral for $La_2CuO_4$ is $J/k_B \simeq 1250$–$1500$ K.[5] The spin-orbit coupling constant for $Cu^{2+}$ ions having a singlet ground orbital state is $\lambda/k_B \simeq 600$–$800$ K.[2] The value of the spin-orbit coupling constant for $Cu^{2+}$ ions having a degenerate ground orbital state is unknown. To construct a model of an orbital glass it was assumed in Ref. 4 that a perturbative scheme can be devised for the parameter $\lambda/J$. It was also assumed in Ref. 4 that the orbital-orbital interaction through spin fluctuations is stronger than the interactions of the orbitals with phonons. The possibility of realizing an analog of the Jahn–Teller effect involving magnons and replacement of the vibronic (orbital-phonon) states by spin (orbital-magnon) states then arises.[4]

The discovery of an orbital-glass state in a $Eu_2CuO_4$ crystal over the broad temperature range $T>T_N$ was reported in Ref. 4. It was assumed there that the Néel temperature for $Eu_2CuO_4$ is $T_N \simeq 150$ K.[4,6–8] There are diverse opinions in the literature regarding the value of $T_N$ for $Eu_2CuO_4$. Our experimental data[4,6,8] were interpreted under the assumption that $T_N \simeq 150$ K. The same value was obtained by investigating the Mössbauer effect.[7] The value $T_N \simeq 270$ K was given in a recently published paper[9] on an investigation of Bragg neutron scattering in $Eu_2CuO_4$. A small jump in the intensity of the Bragg peak was observed at $T \simeq 150$ K, but its nature was not explained. For $T>150$ K there was a practically linear drop in the intensity of the Bragg peak with the temperature up to $T \simeq 270$ K, above which the scattering intensity dropped sharply. We assume that for $T>150$ K there is no quasi-two-dimensional long-range antiferromagnetic order and that there are only restricted regions of quasi-two-

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dimensional order, which can be responsible for the Bragg scattering observed in Ref. 9 for $T>150$ K. This is considered in greater detail in Sec. 3.

Unlike the usual situation, in which a spin- or structural-glass state exists in crystals with random disorder in the arrangement of the ions,[10] in $Eu_2CuO_4$ an orbital-glass state is observed for an ordered crystal. According to the model in Ref. 4, an orbital glass is created by an orbital-orbital interaction through two-dimensional Heisenberg antiferromagnetic spin fluctuations for $T>T_N$. This interaction has a long-range character. Its radius of action is determined by the correlation radius of the two-dimensional Heisenberg antiferromagnetic spin fluctuations $\xi(T)$. Because of the antiferromagnetic character of the spin fluctuations, the orbital–orbital interaction through these fluctuations changes sign after every lattice spacing. We assumed in Ref. 4 that an interaction having these properties can lead to the formation of a glassy state in an ordered crystal. We note that the correlation radii of two-dimensional Heisenberg antiferromagnetic spin fluctuations for $T>T_N$ are described by the formula $\xi=a\exp(2\pi\rho_s/k_BT)$ (Ref. 5). Here $2\pi\rho_s$ is the antiferromagnetic spin stiffness, and $a$ is the lattice constant. Also, we have $2\pi\rho_s\simeq J$. If we assume that the value of $J$ for $Eu_2CuO_4$ is the same as that for $La_2CuO_4$, we have $\xi\sim 2000a$ at $T\sim 150$ K. Thus, the glassy state of the orbital subsystem is induced by two-dimensional Heisenberg antiferromagnetic spin fluctuations and exists over the broad temperature range $T>T_N$.

On the other hand, the state of the spin subsystem itself depends on the state of the orbital subsystem. In fact, when microwave spin dynamics were studied in Ref. 8 in the same temperature range in which an orbital-glass state was observed, well defined ($\gamma\ll\omega_0$) uniform ($q=0$) ferromagnetic spin-wave excitations were discovered ($\omega_0$ is the frequency, $\gamma$ is the damping, and $q$ is the wave vector). It was shown in Ref. 8 that such excitations can appear for $T>T_N$ by virtue of the random Ising fields $h_i^z=\lambda\langle\sigma_i^z\rangle$ in the orbital-glass state on the spin subsystem. Here $\langle\sigma_i^z\rangle$ is the average value of the local frozen orbital angular momentum in the orbital-glass state ($\sigma_i^z=\pm 1$ is the projection of the orbital angular momentum operator $\mathbf{L}$ onto the ground tetragonal doublet). Thus, there is a self-consistent interaction between the spin and orbital subsystems in the crystal.

This paper contains the results of a further experimental investigation of the low-frequency dielectric susceptibility when external constant electric and magnetic fields are applied, as well as the results of an investigation of the static magnetic properties of crystals with different conductivities in constant magnetic fields of different strength and orientation. In Ref. 4 an analysis of the experimental data obtained by studying the dielectric susceptibility in the absence of external fields showed that we are, in fact, dealing with a glassy state. However, it was noted there that along with the usual properties for spin or structural glasses, the orbital glass in a $Eu_2CuO_4$ crystal has some specific properties, whose nature was not discussed in Ref. 4. We are dealing with the coexistence of two different glassy states and the existence of a critical temperature and a critical frequency, at which the properties of the orbital subsystem vary abruptly.[4]

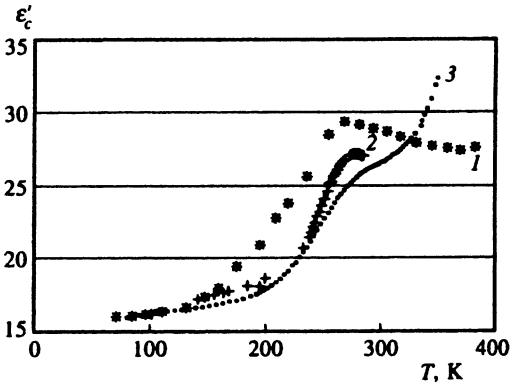


FIG. 1. Temperature dependence of the real part of the dielectric constant ($\varepsilon_c'$) at $f=230$ Hz for various values of the applied external constant electric field $E_=$: *1*—$E_==0$; *2*—$E_==7\times10^3$ V/cm; *3*—$E_==2\times10^4$ V/cm.

In this paper we discuss these special features of an orbital glass with consideration of the self-consistent interaction of the spin and orbital subsystems. We also consider the static magnetic properties obtained both in Ref. 6 and in the present work, and we compare the temperature dependences of the static magnetic susceptibility and the intensity of the Bragg neutron scattering peak.[9]

The paper consists of three sections: an introduction, a section describing the experimental data, and a section devoted to an analysis of the properties of the orbital and spin subsystems.

## 2. EXPERIMENTAL DATA

### 2.1. Influence of external electric and magnetic fields on the orbital-glass state

The influence of external constant electric ($E_=$) and magnetic ($H_=$) fields on the low frequency (in the frequency range 70 Hz–1 MHz) dielectric susceptibility of a $Eu_2CuO_4$ crystal in the temperature range 77–400 K was studied. Highly insulating (with conductivity $\rho<10^{-7}$ $\Omega\cdot\text{cm}^{-1}$ at 300 K) single crystals were selected for the measurements. The measurements in a field $E_=\neq 0$ were performed on the same samples used to obtain the data on the dielectric susceptibility in the absence of an external constant electric field in Ref. 4. The same samples were also used in the measurements of the static magnetic susceptibility. Since the $Eu_2CuO_4$ crystals are thin plates perpendicular to the $c$ axis (the typical dimensions of the plates are $2\times1.5\times0.1$–$0.2$ mm), only the dielectric susceptibility along the $c$ axis of the crystal ($\varepsilon_c$) was determined. The external field $E_=$ was also applied along the $c$ axis. The temperature and frequency dependences of $\varepsilon_c$ in the presence of applied electric fields of different strength ($E_=\leqslant 2\times10^4$ V/cm) were studied.

In Ref. 4 step-like anomalies of the real part of the dielectric susceptibility were discovered in the absence of a constant electric field on the plots of $\varepsilon_c'(T)$ at a fixed frequency, and strong low-frequency dispersion of these anomalies was observed (see the inset in Fig. 2). As is seen from Figs. 1 and 2, the application of an external constant electric field along the $c$ axis of the crystal results in dis-

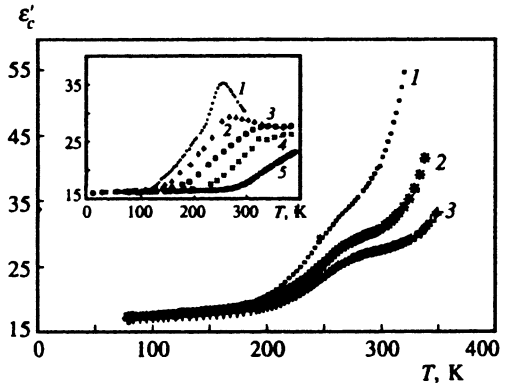


FIG. 2. Temperature dependence of $\varepsilon_c'$ in a field $E_{=}=2\times10^4$ V/cm at various frequencies: *1*—$f=73$ Hz, *2*—$f=140$ Hz, *3*—$f=270$ Hz. Inset: same dependence, but in a field $E_{=}=0$ at various frequencies: *1*—70 Hz, *2*—230 Hz, *3*—1 kHz, *4*—20 kHz, *5*—1 MHz.

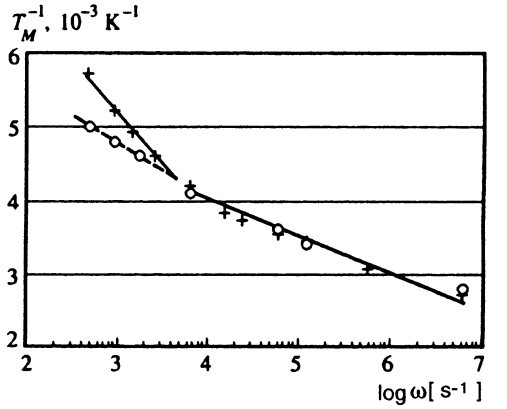


FIG. 3. Dependence of $1/T_M$ on log $\omega$ for the cases of $E_{=}=0$ (crosses) and $E_{=}=2\times10^4$ V/cm (circles).

placement of the anomaly on the plot of $\varepsilon_c'(T)$ toward higher temperatures. Also, as is seen from Figs. 1 and 2, the displacement of the anomaly is especially great in the low-temperature part of the plot of $\varepsilon_c'(T)$. A comparison of the plots in Fig. 2 and in the inset in Fig. 2 reveals that the frequency dispersion on the plots of $\varepsilon_c'(T)$ for $E_{=}=0$ begins at $T\simeq120$ K, while in the case of $E_{=}\neq0$ this dispersion begins at $T\simeq190$ K. A sharp increase in comparison to the case $E_{=}=0$ is observed in $\varepsilon_c'$ at temperatures above 300–350 K when a field is applied ($E_{=}\neq0$). The dielectric loss tangent also increases. We assume that the sharp increases in $\varepsilon_c'$ and $\tan\delta$ for $T>300$ K is caused by the influence of the field on the thermally activated impurity conductivity. We shall not consider this region below. We note that for $T<300$ K a field $E_{=}\neq0$ scarcely alters the dielectric loss tangent.

The dispersion observed for the real part of the dielectric susceptibility both in the case of $E_{=}=0$ (inset in Fig. 2) and in the case of $E_{=}\neq0$ (Fig. 2) is characteristic of spin and structural glasses.[10] We assume that the low-frequency dispersion is caused by the presence of relaxers with a broad set of relaxation times ($\tau_{\min}\ll\tau\ll\tau_{\max}$). We perform an analysis of the experimental data in the case of an applied external electric field just as was done in Ref. 4 for the case of $E_{=}=0$. Let $T_M$ be the temperature of the maximum of an arbitrary plot of $\varepsilon_c'(T)$ at a fixed frequency $\omega$ in the region of the step-like anomaly on $\varepsilon_c'(T)$. Then, as in the absence of a field, the dependence of the relaxation time on the temperature satisfies the Arrhenius law, and we have $\tau=1/\omega=\tau_0\exp(U_A/k_BT_M)$, where $U_A$ is the activation barrier. As in the absence of a field, there are two linear segments on the plot of the dependence of $1/T_M$ on log $\omega$ with a discontinuity at $T_M=T_{cr}\simeq250$ K and $\tau=\tau_{cr}\simeq2\times10^4$ s (Fig. 3). The high-frequency segments ($\tau<\tau_{cr}$) of the plots of the dependence of $1/T_M$ on log $\omega$ for both $E_{=}=0$ and $E_{=}=2\times10^4$ V/cm are similar, while the low-frequency segments ($\tau>\tau_{cr}$) for $E_{=}=0$ and $E_{=}\neq0$ differ. The values of $U_A$ and $\tau_0$ for the temperature range $T<T_{cr}$ and $\tau>\tau_{cr}$ are as follows:

$$E_{=}=0:\quad U_A=0.13\ \text{eV},\quad \tau_0=1.2\times10^{-7}\text{s}, \tag{1}$$

$$E_{=}=2\times10^4\ \text{V/cm}:\quad U_A=0.26\ \text{eV},\quad \tau_0=1.3\times10^{-10}\ \text{s}. \tag{2}$$

In the ranges of values $\tau<\tau_{cr}$ and $T>T_{cr}$ the values of $U_A$ and $\tau_0$ for both $E_{=}=0$ and $E_{=}\neq0$ are as follows:

$$U_A=0.37\ \text{eV},\quad \tau_0=4\times10^{-14}\ \text{s}. \tag{3}$$

Glassy systems usually exhibit one linear segment in the dependence of $q/T_M$ on log $\omega$ (Ref. 10). Thus, an external electric field brings the slopes of the low-frequency and high-frequency segments of the dependence of $1/T_M$ on log $\omega$ closer to one another and gives the observed glassy state a more familiar appearance.

Figure 4 presents plots of the frequency dependence of $\varepsilon_c'$ for the cases of $E_{=}=0$ and $E_{=}=2\times10^4$ V/cm at one fixed temperature. Having a series of such plots for several temperatures, we can obtain the density of states of the relaxers $g(1/\omega_0T)$ in the presence of an electric field, as was done in Ref. 4 for $E_{=}=0$. When an external electric field is applied, just as in the case $E_{=}=0$, there are two segments with a constant density of states: $g_1$ for states having $(1/\omega)_{cr}<1/\omega<(1/\omega)_{max}$ and $g_2$ for states having $(1/\omega)_{min}<1/\omega<(1/\omega)_{cr}$. An abrupt change in the density of

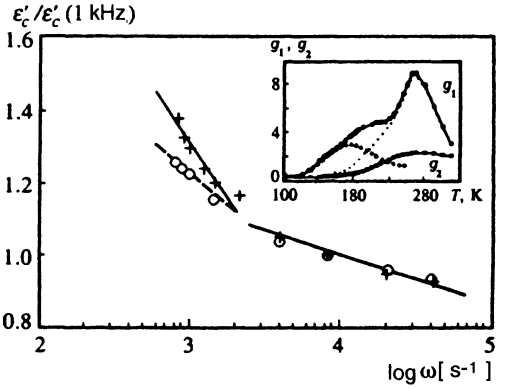


FIG. 4. Dependence of $\varepsilon_c'$ in the relative scale $\varepsilon_c'/\varepsilon_c'$(1 kHz) on log $\omega$ for the cases of $E_{=}=0$ (crosses) and $E_{=}=2\times10^4$ V/cm (circles). Inset: temperature dependences of the orbital-glass densities of states $g_1$ and $g_2$ for $E_{=}=0$ obtained in Ref. 4.

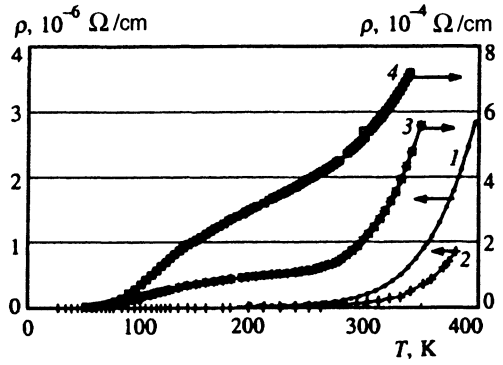


FIG. 5. Temperature dependence of the conductivity for four different samples. The details are given in the text.

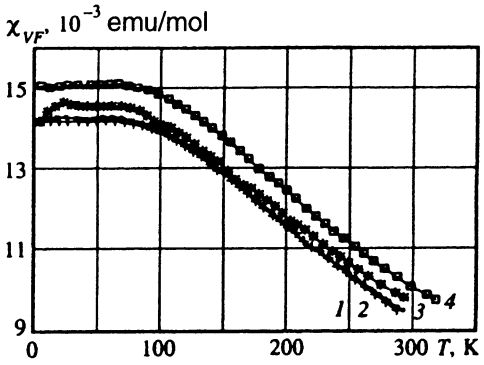


FIG. 6. Temperature dependence of the Van Vleck static magnetic susceptibility for four samples (*1–4*) with different conductivities (see Fig. 5). The external magnetic field $H_{=}=1$ T was applied parallel to the $c$ axis of the crystal.

states occurs at $1/\omega=(1/\omega)_{cr}\simeq 2\times 10^{-4}$ s. The inset in Fig. 4 presents the temperature dependences of $g_1$ and $g_2$ for $E_{=}=0$ (the upper and lower curves, respectively). The upper curve can be divided naturally into two parts with maxima at $T\simeq 150$ K and $T\simeq 250$ K. The first maximum corresponds to $T_N$, and the second maximum corresponds to $T_M\simeq T_{cr}$. The application of a constant electric field results in the following changes in the density of states in the glass: $g_2(1/\omega,T)$ scarcely varies for $E_{=}\neq 0$, and $g_1(1/\omega,T)$ decreases as the field increases. The part of the density of states $g_1(1/\omega,T)$ corresponding to the low-temperature maximum near the value of $T_N$ for the crystal decreases with increasing values of $E_{=}$ and practically vanishes when $E_{=}=2\times 10^4$ V/cm. At the same time, the part of $g_1(1/\omega,T)$ corresponding to the maximum at $T\simeq T_{cr}$ decreases to a considerably lesser extent.

An external constant magnetic field $H_{=}\leqslant 1.5$ T aligned either along the $c$ axis or in the $xy$ plane has practically no influence on the orbital-glass state.

### 2.2. Static magnetic properties

The dielectric properties of $Eu_2CuO_4$ crystals were studied on samples having a very low conductivity. The presence of impurity conductivity due to defects in the crystal or nonstoichiometry with respect to oxygen produces an increase in the dielectric losses and a conductivity contribution to $\varepsilon_c$. Therefore, an orbital glass can be studied by measuring the low-frequency dielectric susceptibility only on stoichiometric insulating single crystals. The investigation of Eu $_2CuO_4$ crystals with various levels of impurity conductivity by measuring the static and dynamic magnetic susceptibility makes it possible to evaluate the variation of the spin state of the crystals as a function of the conductivity. We note that the nonstoichiometry with respect to oxygen in $La_2CuO_{4-\delta}$ creates a possibility for a metal-insulator transition and results in the appearance of superconductivity in such crystals. The conductivity also strongly influences the magnetic properties of $La_2CuO_{4-\delta}$, i.e., the value of the Néel temperature decreases with increasing conductivity.

When single crystals of $Eu_2CuO_4$ are grown,[6] variation of the crystallization temperature range makes it possible to obtain insulating crystals with different levels of impurity conductivity. Figure 5 presents plots of the dc conductivity for the four samples on which the magnetic investigations were conducted. Sample *2*, which has a very low conductivity, was optically transparent,[12] and the dielectric measurements were performed on it.[4] The microwave investigations of the dynamic magnetic susceptibility were performed on samples similar to *1–3*. The behavior of the dynamic magnetic susceptibility was similar for all the samples investigated.[8] Figures 6 and 7 present plots of the temperature dependence of the static magnetic susceptibility obtained in an external magnetic field $H_{=}=1$ T oriented along the $c$ axis or in a plane perpendicular to the $c$ axis for samples *1–4* (see Fig. 5). It is seen that the static magnetic properties scarcely depend on the conductivity. A jump in the static magnetic susceptibility is observed for all the samples at $T\sim 150$ when $H_{=}$ is oriented in a plane perpendicular to the $c$ axis. Varying the orientation of the external field in the plane perpendicular to the $c$ axis did not alter the static magnetic susceptibility. We note that no weak ferromagnetic moment was observed in any of the four samples for $H_{=}=0$ and for fields $H_{=}\leqslant 3$ T aligned along the $c$ axis or in a perpendicular plane at any temperature (before and after the jump at $T\sim 150$ K). The field dependence of the magnetic moment was linear and tended to zero as $H_{=}\to 0$. Figure 8 presents plots of the temperature dependence of the static magnetic susceptibility for sample *2* at various values

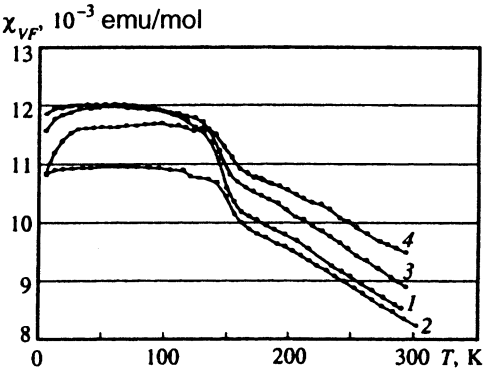


FIG. 7. Same dependence for the same four samples as in Fig. 6, but with an external field $H_{=}=1$ T applied perpendicularly to the $c$ axis of the crystal.

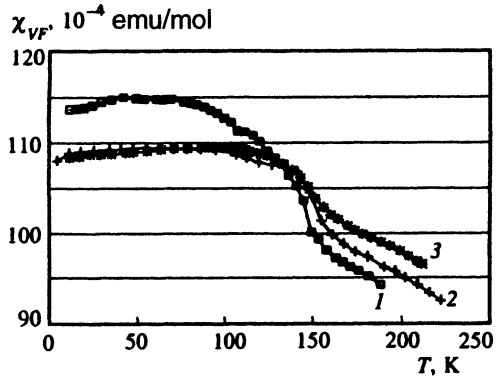


FIG. 8. Dependence of the Van Vleck static magnetic susceptibility in a plane perpendicular to the $c$ axis of the crystal for sample 2 in various external magnetic fields: *1*—0.2 T, *2*—1 T, *3*—2.5 T.

of the external constant magnetic field aligned in a plane perpendicular to the $c$ axis. It is seen that there is a jump in the static magnetic susceptibility at $T\sim 150$ K at all values of the applied magnetic field.

## 3. ANALYSIS OF THE PHYSICAL SITUATION AND INTERPRETATION OF THE EXPERIMENTAL DATA

In this section we analyze the physical situation with consideration of the self-consistent interaction of the spin and orbital subsystems. We first briefly recall the model of an orbital glass described in Ref. 4. We then examine the reciprocal influence of the random Ising fields appearing in the orbital subsystem in the orbital-glass state on the state of the spin subsystem. Two phases appear in the latter: a two-dimensional Heisenberg phase and a two-dimensional Ising antiferromagnetic phase. Finally, we examine how the phase separation of the states of the spin subsystem influences the properties of the orbital subsystem.

### 3.1. Reverse influence of the orbital subsystem on the spin subsystem

A model of a two-dimensional Ising orbital glass based on the Hamiltonian

$$\mathscr{H}^{2D}=\sum_{ij} \mathbf{J}_{ij}(\mathbf{S}_i\cdot\mathbf{S}_j)+\lambda\sum_i S_i^z\sigma_i^z \tag{4}$$

was proposed In Ref. 4. Here the first sum describes two-dimensional Heisenberg antiferromagnetic exchange with the exchange constant $J_{ij}$, which differs from zero at nearest neighbors, and the second sum describes the spin-orbit interaction. The model of an orbital glass in Ref. 4 underlies the hypotheses advanced in the first section of this paper. The Hamiltonian (4) was used to obtain the following effective Hamiltonian for the orbital subsystem in second-order perturbation theory with respect to $\lambda/J$:[4]

$$\mathscr{H}^{\sigma}_{\text{eff}}=\sum_i \lambda\langle S_i^z\rangle\sigma_i^z+\sum_{ij}\frac{\lambda^2}{J_{ij}}K_{ij}^{zz}\sigma_i^z\sigma_j^z. \tag{5}$$

Here $\langle S_i^z\rangle$ is the average value of the $z$ component of the spin in all the states, and $K_{ij}^{zz}=\langle S_i^zS_j^z\rangle-\langle S_i^z\rangle\langle S_j^z\rangle$ is the spin correlation function. If we introduce the antiferromagnetic order parameter $\mathbf{\Omega}_i$ such that $\mathbf{S}_i=(-1)^{n_i}\mathbf{\Omega}_i$ ($n=\pm 1$, depending on the subsystem to which the spin $\mathbf{S}_i$ belongs), then according to Ref. 5

$$\langle\mathbf{\Omega}_i\cdot\mathbf{\Omega}_j\rangle\sim\frac{1}{\sqrt{r_{ij}}}\exp\left(-\frac{r_{ij}}{\xi}\right), \tag{6}$$

where $\xi=a\exp(2\pi\rho_s/k_BT)$, and $r_{ij}$ is the radius vector. The summation in the second term in (5) is carried out over all the sites in the lattice. The first term in (5) describes the magnetization of the orbital subsystem by the mean field $\lambda\langle S_i^z\rangle$. The second term describes the orbital-orbital interaction through antiferromagnetic spin fluctuations. The role of this interaction is especially important when the mean field satisfies $\lambda\langle S_i^z\rangle=0$, i.e., for $T>T_N$. It was assumed in Ref. 4 that the orbital-orbital interaction through two-dimensional Heisenberg antiferromagnetic fluctuations, which is long-range and changes sign after every lattice spacing, is responsible for the appearance of an orbital glass.

Let us examine the reciprocal effect of the orbital subsystem in the orbital-glass state on the two-dimensional Heisenberg antiferromagnetic spin fluctuations, i.e., on the state of the spin subsystem for $T>T_N$. Now these fluctuations are influenced not only by the easy-plane anisotropy, which is caused by the exchange anisotropy[13,14] and leads to orientation of the antiferromagnetic spins in the $xy$ plane for $T<T_N$, but also by the Ising random fields $h_{i\sigma}^z=\lambda\langle\sigma_i^z\rangle$, where $\langle\sigma_i^z\rangle$ is the value of the local frozen orbital angular momentum in the orbital-glass state.

If the easy-plane anisotropy is of the same order as the interlayer antiferromagnetic exchange interaction $J^{\perp}\sim 10^{-5}$ J, as in the case of $La_2CuO_4$ (Ref. 5), the main role in the formation of the state of the spin subsystem is played by the interlayer interaction, which leads to a quasi-two-dimensional state with antiferromagnetic long-range order for $T<T_N$. At the same time, for $T>T_N$, outside the quasi-two-dimensional critical region, easy-plane anisotropy does not alter the character of two-dimensional Heisenberg antiferromagnetic fluctuations.[8] Let us consider how an Ising random field influences the state of two-dimensional Heisenberg spin fluctuations.

We assume that the orbital-glass state in an ordered crystal can be represented in the form of a set of coexisting orbital-density waves with a broad set of wave vectors $q^z_{\sigma\,\min}<q^z_\sigma<q^z_{\sigma\,\max}$. We also assume that in the orbital subsystem there is a set of molecular fields $h^z_\sigma(a^z_\sigma)$ with a Gaussian distribution $f(h^z_z)$.

We consider the response of a two-dimensional Heisenberg antiferromagnet for $T>0$ (or at $T>T_N$ for a quasi-two-dimensional Heisenberg antiferromagnet) to the Ising fields $h^z_\sigma(q^z_\sigma)$. In the temperature range $T>T_N$ of interest to us the spin subsystem is paramagnetic. However, as was already noted, for $T\ll 2\pi\rho_s$ there are two-dimensional Heisenberg antiferromagnetic fluctuations with correlation radii $\xi(T)\gg a$ in it. As a result, there is a local antiferromagnetic order parameter in the layers on scales $\eta\ll\xi(T)$. The response of a two-dimensional Heisenberg antiferromagnet for $T>T_N$ to an external magnetic field $H(q)$ depends greatly on the value of the wave vector $q$. In the case of a uniform

($q=0$) external field, the response of the spin subsystem is simply an isotropic paramagnetic susceptibility. In the case of a field with $q\neq 0$, the two-dimensional Heisenberg antiferromagnetic fluctuations have a local spin polarization scale $\eta=1/q$ along with the correlation radius $\xi(T)$. If $\eta\ll\xi(T)$ holds, then, according to Ref. 5, a transverse antiferromagnetic susceptibility, which is local on a scale $\eta\ll\xi(T)$, appears in the spin subsystem instead of the isotropic paramagnetic susceptibility when certain conditions are satisfied. These conditions are as follows : $T\ll 2\pi\rho_s$ and $q_s\xi(T)\geqslant 1$, where the wave vector is $q_s=q-q_{AF}$, and $q_{AF}$ is the antiferromagnetic vector in the reciprocal lattice. Also, according to Ref. 5, the local transverse antiferromagnetic susceptibility depends weakly on $q_s$ and is close in magnitude to the uniform transverse antiferromagnetic susceptibility $\chi_\perp^0$ at $T=0$. For $q_s\xi>1$ we have

$$\chi_\perp(q_s,T)\simeq\frac{2}{3}\chi_\perp^0. \tag{7}$$

At $q_s\xi=1$

$$\chi_\perp(q_s,T)=\chi_\perp^0, \tag{8}$$

and the longitudinal susceptibility is $\chi_\parallel(q_s,T)=0$.

A special situation arises in the case $q=q_{AF}$, i.e., when a staggered field $H_{st}(q_{AF})$ is applied. The condition $\eta\ll\xi(T)$ is satisfied at all temperatures $T\ll 2\pi\rho_s$. However, $q_s=0$ holds, and the condition $q_s\xi\geqslant 1$ cannot be satisfied at any temperature. In this case, in analogy to a three-dimensional Heisenberg antiferromagnetic (see Ref. 8), we assume that there is a susceptibility $\chi_{st}=[J(0)-J(q_s)]^{-1}$ in the limit $T\to 0$. In the limit $q_s\to 0$, we have $\chi_{st}\to\infty$. We assume that the same situation exists for $T\ll 2\pi\rho_s$ and when the wave vector $q_s$ deviates only slightly from $q_{AF}$. If the field $H_{st}$ is parallel to the $z$ axis, crossover to a two-dimensional Ising antiferromagnetic state should occur in the two-dimensional Heisenberg antiferromagnetic spin subsystem in the situation considered above.

As we postulated above, the fields $h_\sigma^z(q_\sigma^z)$ over a broad set of wave vectors $q_{\sigma\,\max}^z>q_\sigma^z>q_{\sigma\,\min}^z$ coexist in the orbital subsystem in the orbital-glass state, and the spin subsystem is influenced by these fields. In this case we have $q_s^z=q_\sigma^z-q_{AF}^z$.

If the condition $q_s^z\xi(T)\geqslant 1$ holds for $q_s^z$ in the range $q_{s\,\min}^z<q_s^z<q_{s\,cr}^z$ then in accordance with the foregoing discussion there is a transverse antiferromagnetic susceptibility $(2/3)\chi_\perp^0<\chi_\perp(q_s,T)<\chi_\perp^0$ which is local on a scale $\eta\ll\xi$, in the spin subsystem at $T\ll 2\pi\rho_s$. Here $q_{\sigma\,cr}^z$ corresponds to $q_s^z=q_{\sigma\,cr}^z-q_{AF}=1/\xi(T)$.

The transverse antiferromagnetic susceptibility $\chi_\perp(q_s,T)$ leads to the appearance of weakly ferromagnetic moments on a scale $\eta\ll\xi(T)$ in the layers:

$$m_{loc}^z\simeq\chi_\perp^0 h_\sigma^z(q_\sigma^z). \tag{9}$$

An antiferromagnetic order parameter is detected here in the $xy$ plane on the same local scales (as for $T<T_N$). The correlation radii of the two-dimensional Heisenberg antiferromagnetic fluctuations in the $xy$ plane are determined, as before, by $\xi(T)$. Since $\chi_\parallel=0$ holds, the components $S^z$ of the spin fluctuations do not interact with the fields $h_\sigma^z(q_\sigma^z)$. Thus, the fields $h_\sigma^z(q_\sigma^z)$ with the values $q_{s\,\min}^z<q_s^z<q_{s\,cr}^z$ do not alter the character of the two-dimensional Heisenberg antiferromagnetic spin fluctuations.

If there are fields $h_\sigma^z(q_\sigma^z)$ for which $q_\sigma^z$ falls within the range $q_{\sigma\,\max}^z>q_\sigma^z>q_{\sigma\,cr}^z$ and $q_{\sigma\,\max}^z\simeq q_{AF}$ holds in the two-dimensional spin subsystem, crossover from a two-dimensional Heisenberg state to a two-dimensional Ising state will occur in the layers on local scales. In these local regions the antiferromagnetic vector is parallel to the $z$ axis. Since we have $\xi(T)\gg a$ at low temperatures (near $T_N$), the phase volume of the two-dimensional Ising phase is small. In fact, the condition $q_s\xi(T)=1$ holds even when the deviation of $q_s$ from $q_s=q_{AF}$ is very small (of the order of $1/\xi$). The phase volume of the two-dimensional Ising phase increases only in the limit $T\to 2\pi\rho_s$. Thus, the two-dimensional Heisenberg antiferromagnetic phase predominates in the spin subsystem over the broad temperature range $T\ll 2\pi\rho_s$ of interest to us.

We note that the spin subsystem can also be influenced by the subsystem of thermally excited $Eu^{3+}$ ions in the magnetic $^7F_1$ excited state, which is at a distance $\sim 300$ cm$^{-1}$ along the energy scale from the nonmagnetic $^7F_0$ ground state. When this excited level is thermally populated and there is a sufficient concentration of thermally excited $Eu^{3+}$ ions in the Eu subsystem, clusters of correlated states can appear, as was observed in $Eu_2CuO_4$ crystals in Refs. 15 and 16. We assume that the correlations of the displacements of the $Eu^{3+}$ ions along the $z$ axis in the clusters are antiferroelectric. Owing to the interaction of these displacements with the orbital states of the $Cu^{2+}$ ions, an additional field $H_{st\,Eu}^z$ through which the Eu subsystem affects the spins of the $Cu^{2+}$ ions, can appear on local scales corresponding to the scales of the clusters of $Eu^{3+}$ ions. For $T\ll 2\pi\rho_s$ this field also causes crossover from a two-dimensional Heisenberg state to a two-dimensional Ising state of the Cu spin subsystem. We note that at temperatures $T\geqslant 200$ K the phase volume of the two-dimensional Ising phase appearing under the influence of the Eu subsystem can exceed the phase volume that appears under the influence of the orbital subsystem of the $Cu^{2+}$ ions.

We note that the correlation radius of the two-dimensional Ising spin fluctuations in the two-dimensional Ising phase [$\xi_{Is}\propto(T-T_c)^{-1/8}$ (Ref. 17)] is considerably smaller than the correlation radius of the two-dimensional Heisenberg spin fluctuations $\xi(T)$.

At temperatures $T>T_N$ an interaction $J^\perp m_i^z m_k^z$ (the sites $i$ and $k$ are the nearest $Cu^{2+}$ ions in neighboring layers) appears on a scale $\eta\ll\xi$ in the two-dimensional Heisenberg antiferromagnetic phase owing to the presence of antiferromagnetic interlayer exchange with a constant $J^\perp$ ($J^\perp\ll J$). This interaction was absent for $T<T_N$ in the region of long-range antiferromagnetic order without weak ferromagnetism. As a result, there is mutual antiferromagnetic orientation of the weak ferromagnetic moments that are local on scales $\eta\ll\xi(T)$ in neighboring layers. Restricted regions of quasi-two-dimensional order with antiferromagnetic vectors aligned in $xy$ planes should then appear. Long-range quasi-two-dimensional antiferromagnetic order cannot appear in the absence of an Ising random field $f(h_\sigma^z)$ (Ref. 18). We

assume that the Bragg neutron scattering observed in Ref. 9 at $T>T_N$ can be caused by the presence of such restricted quasi-two-dimensional regions.

We note that the quasi-two-dimensional states at $T<T_N$, at which antiferromagnetic long-range order is realized, and at temperatures $T>T_N$, when there are restricted regions of a quasi-two-dimensional state, are different in nature, although the symmetry of the order parameter is the same in both cases. At temperatures $T<T_N$ the quasi-two-dimensional long-range order is caused by the weak interlayer antiferromagnetic exchange interaction $J^{\perp}S_iS_k$. Also, in contrast to the case of $La_2CuO_4$, the relative positions of the $Cu^{2+}$ ions in neighboring $CuO_2$ layers in the tetragonal crystals of $R_2CuO_4$ is such that the interlayer interaction vanishes in the mean-field approximation when the spins in the layers have an antiferromagnetic arrangement.[13,14] Therefore, the coupling between the layers can be obtained outside the mean-field approximation (for example, with consideration of the quantum zero-point fluctuations[14]). In the case $T>T_N$ the coupling between the layers is mediated by the antiferromagnetic correlation of the weak ferromagnetic moments in neighboring layers.

Thus, in the low-temperature region ($T<T_N\simeq 150$ K) there is quasi-two-dimensional antiferromagnetic long-range order with spins lying in the $xy$ plane in the spin subsystem. There is then no weak ferromagnetism. An orbital-glass state[4,8] appears in the quasi-two-dimensional critical region near the Néel temperature and exists over a broad temperature range $T>T_N$. The reciprocal effect of the orbital subsystem of the copper ions and the thermally excited states of the europium ions on the spin subsystem of the $Cu^{2+}$ ions results in a separation of phases in the latter, i.e., the appearance of phases with two-dimensional Heisenberg and two-dimensional Ising antiferromagnetic fluctuations. The former phase is characterized by the existence of local two-dimensional weak ferromagnetic moments parallel to the $z$ axis and restricted regions of quasi-two-dimensional order with antiferromagnetism vectors fixed in the $xy$ plane. In the latter phase (the two-dimensional Ising phase) the local antiferromagnetic vector is parallel to the $z$ axis of the crystal.

### 3.2. Properties of an orbital glass

As was shown in Ref. 4 (see also the preceding section) an orbital-glass state appears as a result of the long-range, orbital-orbital interaction of alternating sign through two-dimensional Heisenberg antiferromagnetic spin fluctuations described by the second term in the Hamiltonian (5). Here we assumed that the orbital–orbital interaction through spin fluctuations, which is enhanced by its long-range action, surpasses the orbital-orbital interaction through phonons. Let us examine this situation in greater detail. If the correlation radius of the two-dimensional Heisenberg antiferromagnetic spin fluctuations $\xi(T\to 2\pi\rho_s)\to a$, the orbital–orbital interaction through these fluctuations will be very weak: $J_\sigma^s(T\Rightarrow 2\pi\rho_s)\simeq\lambda^2/J$. For a square lattice in a layer at temperatures $T\ll 2\pi\rho_s$ the interaction $J_\sigma^s$ is enhanced by its long-range action, since at these temperatures $\xi(T)\gg a$ [see Eqs. (5) and (6)]. In this case $J_s^s\gg J_s^{ph}$ holds and splitting of the tetragonal doublet occurs on spin excitations (the spin Jahn-Teller effect). The constant $J_\sigma^{ph}$ characterizes the orbital-orbital interaction through phonons.

As was shown in the preceding section, at temperatures such that an orbital glass exists the spin subsystem breaks down into two coexisting phases, viz., a two-dimensional Heisenberg phase and a two-dimensional Ising phase. The orbital-glass state considered in Ref. 4 and above in this paper is induced specifically by two-dimensional Heisenberg antiferromagnetic fluctuations. In a two-dimensional Ising phase the correlation radius of the spin fluctuations and, accordingly, the strength of the orbital-orbital interaction through the spin fluctuations are considerably smaller. If the orbital-orbital interaction through phonons is weaker than the interaction through two-dimensional Ising spin fluctuations, the usual vibronic Jahn-Teller effect is realized in the orbital subsystem for the phase volume occupied by the two-dimensional Ising phase. Local structural distortions should then occur. Although the volume of the two-dimensional Ising antiferromagnetic phase is small at low temperatures, it exists at any temperature in the region where the orbital glass exists (i.e., in the quasi-two-dimensional critical region near $T_N$ and for $T>T_N$). Local structural distortions should appear over this entire temperature range. We assign the glassy state with the density $g_2$ that we observed experimentally (see the inset in Fig. 4) to an orbital state induced by the two-dimensional Ising phase, while the orbital glass with the density $g_1$ is induced by the two-dimensional Heisenberg phase. As a result, the ratio of the densities of states $g_1$ and $g_2$ simultaneously characterizes the ratio of the phase volumes of the two-dimensional Heisenberg and two-dimensional Ising phases, respectively, in the spin subsystem.

The presence at all temperatures of the critical scale $1/q_{s\,cr}$ for spin polarization by the field $h(q^z_{\sigma\,cr})$, at which crossover from the two-dimensional Heisenberg state to the two-dimensional Ising state occurs in the spin subsystem, leads to the appearance of the critical lifetime $\tau_{cr}=1/\omega_{cr}$ observed in our experiments. To interpret the plots of the frequency dependence of the dielectric susceptibility of the crystal, we assume that the maximum response occurs for the states which satisfy the condition $\omega\tau(T)=1$. Therefore, at every temperature the response of the orbital glass with the density of states $g_1$ is dominant in the low-frequency range $\omega<\omega_{cr}$, and the response of the orbital glass with the density of states $g_2$ is dominant for $\omega>\omega_{cr}$. At $\omega=\omega_{cr}$ the responses of these states coincide. The linear plots of $\varepsilon_c'$ as a function of $\ln\omega$ and of $1/T_M$ as a function of $\log\omega$ (see Ref. 4 and Figs. 3 and 4 in this paper) exhibit discontinuities at $\omega=\omega_{cr}$, because the values of $U_A$ and $\tau_0$ for the $g_1$ and $g_2$ states differ significantly [see Eqs. (1) and (3)]. The value of $U_A$ for the $g_1$ states is close to the value of the antiferromagnetic spin stiffness, and $\tau_0\simeq 10^{-7}$ s. At the same time, the value of $U_A$ for the $g_2$ states characterizes the energy of a local structural distortion, and $\tau_0\simeq 10^{-14}$ s. As we see, the values of $\tau_0$ correlate with the scales of the respective interactions. We note that in the orbital subsystem there is also a critical temperature $T_{cr}$, which corresponds in our case to $T_{cr}\simeq 250$ K. We assume that this is the temperature at which

the condition $q_s\xi(T=T_{cr})=1$ holds for the most probable value of the field $\bar{h}_z^z(\bar{q}_\sigma^z)$ in the Gaussian distribution $f(h_\sigma^z)$. At temperatures $T>T_{cr}$ the orbital density of states $g_1$ drops (see the inset in Fig. 4).

### 3.3. Influence of external electric and magnetic fields on the properties of an orbital glass

As was noted above, the glassy states with the densities $g_1$ and $g_2$ coexist in the orbital subsystem. The coupling of the orbital states with the lattice is considerably weaker for the $g_1$ states than for the $g_2$ states. However, in both of these cases the polarizability of the crystal is governed by the orbital-lattice interaction. In our case of a ground orbital state in the form of a tetragonal doublet, the orbital-lattice interaction reduces to an orbital-orbital interaction through phonons, which is realized through the quadrupole orbital angular momenta (see Ref. 1).

For the $g_1$ states the ground-state splitting of the tetragonal doublets is caused by the interaction of the orbitals with spin fluctuations, and the weak interaction of the orbitals with phonons results in small lattice distortions, which correlate with the ground-state splitting of the doublets. Thus, the polarizability of the crystal that we studied provides information on the orbital-glass state in this case. In addition, the weak and random coupling of the orbitals with the lattice produces the anomalous softness of the lattice. For the $g_2$ states the interaction of the orbitals with phonons is the main mechanism for splitting the tetragonal doublets, and it makes the lattice stiffer.

An external constant electric field $E_=$ parallel to the $z$ axis results in the appearance of a nonzero uniform polarization uniform distortion $P=\varepsilon_c' E_=$. The crystal field experiences uniform distortion for the $g_1$ states, resulting in uniform splitting of the tetragonal doublet along with weak and random splitting in the glassy state. In this case the height of the activation barrier $U_A$ increases, and the value of $\tau_0$ in the Arrhenius law varies [see Eqs. (2) and (3) in Sec. 2.1].

We also note that the field $E_=$ has an especially strong influence on the $g_1$ orbital states due to the orbital-orbital interaction through quasi-two-dimensional critical antiferromagnetic spin fluctuations, which have a density maximum near $T_N$ (Ref. 8). The homogeneous ferroelectric polarization appearing in a field $E_=\neq 0$ gives rise to a uniform orbital angular momentum $\langle\sigma_i^z\rangle_E\neq 0$. Here a uniform magnetizing field $\lambda\langle\sigma_i^z\rangle_E\neq 0$ that acts on the spin subsystem also appears along with the random fields $\lambda\langle\sigma_i^z\rangle$. This field can freeze the quasi-two-dimensional critical spin fluctuations and practically eliminate the states with the density $g_1$ that have a maximum at $T\simeq 150$ K, as is observed experimentally (see Figs. 1 and 4). However, this field cannot freeze the two-dimensional Heisenberg antiferromagnetic fluctuations ($\lambda/k_B\ll 2\pi\rho_s$) and the orbital glass induced by these fluctuations (i.e., the $g_1$ states with a density maximum at $T\simeq 250$ K). In fact, in a field $E_=\simeq 2\times 10^4$ V/cm the low-frequency dispersion attesting to the appearance of an orbital-glass state appears at $T\simeq 190$ K (see Fig. 2), rather than at $T\simeq 120$ K (as when $E_==0$). According to Ref. 8, crossover from a three-dimensional state to a two-dimensional state occurs in the system of antiferromagnetic spin fluctuations at $T\simeq 190$ K. The influence of an external field $E_=\neq 0$ is considerably weaker for the $g_2$ states, since the stiffness of the lattice is greater in this case.

As was noted in Sec. 2.1, an external magnetic field $H_=\leqslant 1.5$ T aligned either along the $z$ axis or in the $xy$ plane does not influence the orbital-glass state. This is apparently because the applied external field is considerably weaker than the internal magnetic fields present in the crystal. In addition, the external field is uniform, i.e., it has a weak influence on the spin subsystem at temperatures $T>T_N$, for which there is only an isotropic paramagnetic susceptibility in the present case.

### 3.4. Static magnetic properties

According to Refs. 6 and 19, the measured static magnetic susceptibility in $Eu_2CuO_4$ is determined mainly by the Van Vleck paramagnetic susceptibility of the $Eu^{3+}$ ions. The antiferromagnetic susceptibility of the subsystem of $Cu^{2+}$ ions is small due to the high spin stiffness (for example, for $La_2CuO_4$ we have $\chi\sim 10^{-4}$ emu/mole, which is considerably smaller than the Van Vleck susceptibility of the $Eu^{3+}$ ions [$\chi_{VV}\sim 10^{-2}$ emu/mole (Refs. 6 and 19)]. The latter is determined by the mixing of the magnetic excited states $^7F_n$ ($n=1,2,\ldots$) with the nonmagnetic $^7F_0$ ground state of the $Eu^{3+}$ ions both by components of the crystal field and by the effective field of the Eu-Cu exchange interaction.

When the external constant magnetic field was oriented in a plane perpendicular to the $z$ axis, a jump in the static magnetic susceptibility was observed in Ref. 6 at $T\simeq 150$ K, which we attribute to the abrupt variation of the spin state of the $Cu^{2+}$ ions and, accordingly, to the variation of the Eu-Cu internal exchange field and the Van Vleck susceptibility of the $Eu^{3+}$ ions. In fact, investigations of the crystal structure of $Eu_2CuO_4$ do not reveal a structural phase transition at $T\simeq 150$ K, at which the values of the crystal-field splittings of the levels of the $Eu^{3+}$ ions could vary abruptly. A calculation of the temperature dependence of the Van Vleck susceptibility associated with the crystal field[19] gives a smooth dependence free of jumps for the susceptibility both along the $z$ axis and in a plane perpendicular to it. Therefore, it is natural to assume that the observed susceptibility jump is caused by variation of the state of the spin subsystem of the $Cu^{2+}$ ions.

Let us discuss the possible nature of the abrupt variation of the state of the spin subsystem of the $Cu^{2+}$ ions, taking into account the self-consistent influence of the spin and orbital subsystems. The quasi-two-dimensional antiferromagnetic long-range order in the Cu subsystem is destroyed at $T=T_N\simeq 150$ K. However, usually there should be no susceptibility jumps upon such a transition. In our case at temperatures $T>T_N$ the spin subsystem passes into an inhomogeneous state, in which the same antiferromagnetic state as at temperatures $T<T_N$ is maintained on local scales $\eta\ll\xi(T)$. At temperatures $T<T_N$ there is a homogeneous quasi-two-dimensional antiferromagnetic state with antiferromagnetism vectors fixed in the $xy$ plane, which is characterized by a uniform transverse antiferromagnetic susceptibility, i.e., $\chi_\perp^0$

As was shown above (see also Ref. 8), near $T_N$ (in the region where the quasi-two-dimensional critical fluctuations with large correlation radii exist), as well at temperatures $T \gg T_N$, where there are two-dimensional Heisenberg antiferromagnetic spin fluctuations with correlation radii $\xi \gg a$, two phases coexist in the spin subsystem. For the two-dimensional Heisenberg antiferromagnetic phases on local scales $\eta \ll \xi(T)$ in the layers, only part of the phase volume has a transverse antiferromagnetic susceptibility when an external uniform magnetic field is applied in the $xy$ plane. In addition, as was shown in Sec. 3.1, $\chi_\perp(q_s,T)=(2/3)\chi_\perp^0$ holds for states that satisfy the condition $q_s\xi(T)>1$. As a result, there is abrupt variation of $\chi_\perp(q_s,T)$ near $T_N$, and a magnetic moment $m^{\rm pl} \simeq \chi_\perp^0 H_=$ develops in the plane. The values of the effective Eu-Cu exchange field $(\mathbf{V}_{ij}\mathbf{m}_i^{\rm Cu}\mathbf{m}_j^{\rm Eu})$ and, accordingly, of the Van Vleck susceptibility also vary abruptly.

Due to the antiferromagnetic interaction between the layers, the magnetic moment components $m_i^z$ have antiparallel orientations, and the mean value of the magnetic moment is $\langle m^z\rangle=0$. In this case the relatively weak external magnetic field oriented along the $z$ axis has practically no influence on the Van Vleck susceptibility. However, when the strength of the field $H_=$ parallel to the $z$ axis is sufficiently great (comparable to the effective field of the antiferromagnetic interlayer coupling for the Cu subsystem $(h_j^z=\Sigma_i J_{ij}^\perp m_i^z)$], a metamagnetic transition, under which the antiferromagnetic arrangement of the magnetic moments in neighboring layers transforms into a ferromagnetic arrangement, is possible. When $T>T_N$ holds, such a transition can occur in the restricted quasi-two-dimensional regions appearing as a result of the interlayer correlations of the weak two-dimensional local magnetic moment. A nonzero mean weakly ferromagnetic moment $\langle m^z\rangle \neq 0$ then appears, and a jump in the Van Vleck susceptibility should be observed. Such transitions were not observed in $Eu_2CuO_4$ in the fields $H_= \leqslant 3$ T that we used.

Thus, no Van Vleck susceptibility features are expected or observed experimentally in the physical situation considered above for $T>T_N$, provided that $T \ll 2\pi\rho_s$ holds (including $T \simeq 250$–$270$ K). In fact, the volume of the two-dimensional Heisenberg antiferromagnetic phase, which determines the influence of the spin subsystem on the Van Vleck susceptibility, only varies (decreases) smoothly in the layers over this entire temperature range.

Another situation exists for the temperature dependence of the Bragg scattering intensity,[9] since scattering only from quasi-two-dimensional states is studied at temperatures $T>T_N \simeq 150$ K. As was shown above, the phase volume of the quasi-two-dimensional states decreases most abruptly at temperatures $T \geqslant T_{\rm cr} \simeq 250$, although for $T>T_{\rm cr}$ the quasi-two-dimensional states are maintained up to very high temperatures. We assume that the abrupt drop in the intensity of the Bragg peak at $T \simeq 270$ K observed in Ref. 9 is attributable to the abrupt decrease in the phase volume of the quasi-two-dimensional states at those temperatures.

Additional investigations are needed to ascertain the real value of the Néel temperature (i.e., the temperature below which quasi-two-dimensional antiferromagnetic long-range order, rather than restricted regions of quasi-two-dimensional order, exists).

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