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ANTIFERROMAGNETIC RESONANCE IN MnO

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The low-frequency branch of antiferromagnetic resonance in MnO was observed experimentally. Its spectrum was investigated and the angular and temperature dependences of the resonance field and the width of the resonance curve were determined. The experimental results were found to be in good agreement with the hypothesis of the existence of an "easy-plane" magnetic anisotropy and of T-type domain structure.

1. INTRODUCTION

MnO is a typical member of a group of crystals, known as NiO-type antiferromagnets, which have a special magnetic structure. This group includes the divalent oxides of the 3d metals Mn, Co, and Ni. In the paramagnetic state these oxides have the fcc lattices and their antiferromagnetic ordering below the Néel temperature is accompanied by a slight distortion of the original crystallochemical structure.

Although NiO-type antiferromagnets are the classical objects for investigating antiferromagnetism (see, for example, reviews[1,2]) and much work has been done on these crystals, they are still attracting interest, because of several unsolved problems. Thus, for example, the low-frequency branch of antiferromagnetic resonance has not yet been observed in any member of this group of crystals. The present paper describes an investigation of this low-frequency branch in the case of MnO. In earlier brief communications[3,4] we mentioned the first observations of the low-frequency branch of antiferromagnetic resonance in NiO and MnO. The high-frequency branch of antiferromagnetic resonance in MnO lies, like the corresponding high-frequency branches of the other crystals in the same group, in the infrared part of the spectrum and it has been investigated by many workers.[5-7]

According to the current ideas, the transition of MnO to the antiferromagnetic state at $T_N = 120°K$[1] is accompanied by a rhombohedral distortion of the crystal lattice, namely, a distortion along one of the space diagonals $\langle 111 \rangle$ of the original cube.[2] This distortion can be represented by a quantity δ which is the deviation of the angle between the edges of the unit cell from its initial value of $\pi/2$. In the case of MnO at 77°K the distortion is $\delta = 6.5 \times 10^{-3}$ rad.[9] Although the distortion of the cubic structure is slight, nevertheless the magnetic properties become strongly anisotropic and exhibit an axial anisotropy of the "easy-plane" type. The anisotropy energy is of the dipole-dipole origin and the anisotropy constant K_1 is of the order of 10^7 erg/cm^3.[10]

The phase transition at the Néel point is of first order[9] and it produces not only magnetic ordering but also alters the crystallochemical structure. Below T_N the spins in any one (111) plane, perpendicular to the shorter [111] diagonal, are aligned in parallel; in alternate (111) planes the spins are antiparallel.

In the paramagnetic state a crystal of MnO has four equivalent $\langle 111 \rangle$ directions and, therefore, below T_N twinning may give rise to splitting into macroscopic regions (T domains) each of which has its own compression axis: $[111]$, $[\bar{1}11]$, $[1\bar{1}1]$, or $[11\bar{1}]$; consequently, each such region has its own easy plane.

MnO not only exhibits a strong magnetocrystalline anisotropy of the easy-plane type but also a weak sixth-order anisotropy in the easy plane.[11] This stimulates the formation of S domains[3] in which the directions of spins are deviated through an angle which is a multiple of 60°. The anisotropy energy associated with the existence of the S domains is small and a weak magnetic field of the order of several kilo-oersteds[11] is sufficient to induce a uniform spin ordering (single S domain) in the easy plane, i.e., within a T domain. Since our resonance measurements were carried out in fields of the order of tens of kilo-oersteds, the S domain structure should have had no influence on the results obtained.

Thus, the antiferromagnetism of MnO is of special nature because it has a strong easy-plane anisotropy, but the relative positions of the easy planes in different macroscopic regions are governed by the cubic symmetry of the original crystallochemical structure. This special magnetic structure should affect various properties of MnO crystals including, naturally, the spectrum of the low-frequency branch of antiferromagnetic resonance. It should be noted that MnO does not exhibit weak ferromagnetism and that it differs in this respect from the majority of the easy-plane antiferromagnets whose low-frequency branch of antiferromagnetic resonance has been investigated.

2. GROUND STATE AND SPECTRUM OF RESONANCE FREQUENCIES

We shall interpret the experimental data using the model of a collinear, two-sublattice, planar antiferromagnet and we shall ignore the weak anisotropy in the easy plane. The energy of such an antiferromagnet can be written in the form

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[1] Different measurement methods give values of T_N in the range 116-122°K.

[2] Above and below T_N, MnO crystals are fully isomorphous with NiO whose structure has been investigated more thoroughly. [8]

[3] The domain notation for MnO is as follows: T represents twin domains and S represents spin domains.

$$W = \tfrac{1}{2}BM^2 + \tfrac{1}{2}(aL_z^2 + bM_z^2) - \mathbf{MH}_0, \qquad (1)$$

which includes the exchange energy, the anisotropy energy, and the Zeeman energy in an external static magnetic field $\mathbf{H}_0$. Here and later we shall use the following notation:

$$\mathbf{M} = \mathbf{M}_1 + \mathbf{M}_2, \quad \mathbf{L} = \mathbf{M}_1 - \mathbf{M}_2,$$
$$H_E = BM_s, \quad H_A{}^a = 2aM_s, \quad H_A{}^b = 2bM_s, \qquad (2)$$
$$\mathbf{M} = \mathbf{M}_0 + \mathbf{m}(t), \quad \mathbf{L} = \mathbf{L}_0 + \mathbf{l}(t),$$

where $\mathbf{M}_1$ and $\mathbf{M}_2$ are the magnetization vectors of the two sublattices; $M_S = |\mathbf{M}_1| = |\mathbf{M}_2|$ is the absolute value of the sublattice magnetization; H_E is the exchange field; H_A^a and H_A^b are the effective anisotropy fields; $\mathbf{M}_0$ and $\mathbf{L}_0$ are the equilibrium values of $\mathbf{M}$ and $\mathbf{L}$.

The ground state can be found from the minimum of the energy (1) subject to the two additional conditions $\mathbf{ML} = 0$ and $M^2 + L^2 = 4M_S^2$, which follow from the definitions of $\mathbf{M}$ and $\mathbf{L}$ given in Eq. (2).

Since the problem considered has the axial symmetry, we can assume (without any loss of generality) that $\mathbf{H}_0$ lies in the zy plane making an angle ψ with the easy plane, which coincides with the xy plane. Then, we find that in the ground state

$$M_{0x} = 0, \qquad L_{0x} = L_0 \approx 2M_s\sqrt{1 - \left(\frac{H_0}{2H_E}\right)^2}, \quad \chi_\perp = \frac{1}{B},$$
$$M_{0y} = \frac{H_y}{B} = \frac{H_0\cos\psi}{B}, \qquad L_{0y} = 0, \qquad \chi_\parallel = \frac{1}{B+b},$$
$$M_{0z} = \frac{H_z}{B+b} = \frac{H_0\sin\psi}{B+b}, \quad L_{0z} = 0, \qquad \frac{\Delta\chi}{\chi} = \frac{H_A{}^b}{2H_E} \qquad (3)$$

where $\chi_\perp$ and $\chi_\parallel$ are the static susceptibilities in the easy plane and along the difficult axis, respectively; $\Delta\chi$ is the difference between these susceptibilities.

It is evident from Eq. (3) that the direction of $\mathbf{M}$ is practically identical with the direction of $\mathbf{H}_0$ for any value of ψ and the static susceptibility is almost isotropic to within H_A^b/H_E. If we use the standard terminology for the easy-axis antiferromagnets, we can say that only the "transverse" susceptibility exists in a planar crystal. It follows from the results of measurements reported in[12-14] that the static susceptibility of NiO-type ferromagnets subjected to sufficiently strong fields (when the anisotropy in the easy plane becomes unimportant) depends weakly on the temperature and exhibits only a slight anisotropy.

The problem of small oscillations of the magnetization and of finding the resonance frequencies can be solved with the aid of the Landau-Lifshitz equations. The condition for the existence of a nontrivial solution of these equations after they are linearized leads to the expression

$$\left(\frac{\omega}{\gamma}\right)^4 - \left(\frac{\omega}{\gamma}\right)^2\left[\frac{B+b}{B}H_0^2\cos^2\psi + aBL_0^2 + \left(\frac{B}{B+b}\right)^2 H_0^2\sin^2\psi\right] + aBL_0^2\frac{B+b}{B}H_0^2\cos^2\psi = 0, \qquad (4)$$

which is analogous to the equation obtained by Borovik-Romanov and Prozorova[15] for an antiferromagnet with a weak magnetic moment. If $\psi = 0$, i.e., if $\mathbf{H}_0$ lies in the easy plane, Eq. (4) yields the following expressions for the two branches of the spectrum:

$$\left(\frac{\tilde{\omega}_1}{\gamma}\right)^2 = \frac{B+b}{B}H_0^2 = \frac{\chi_\perp}{\chi_\parallel}H_0^2, \qquad (5)$$

$$\left(\frac{\tilde{\omega}_2}{\gamma}\right)^2 = aBL_0^2 = 2H_A{}^aH_E\left[1 - \left(\frac{H_0}{2H_E}\right)^2\right], \qquad (6)$$

which are analogous to the well-known formulas given, for example, in Turov's monograph.[16]

If $\psi \neq 0$, it is shown in[15] that the two resonance branches will interact and repel each other, particularly in the region of intersection of $\tilde{\omega}_1$ and $\tilde{\omega}_2$. In the case of MnO, the activation energy of the high-frequency branch is high and it corresponds to 27.5 cm^{-1} at about 0° K.[5] Therefore, the intersection of the branches (5) and (6) in the case of MnO occurs either in very strong magnetic fields (~ 300 kOe) and correspondingly high frequencies (infrared range) or in the direct vicinity (within 1 deg) of T_N in the millimeter range (the second possibility follows from the temperature dependence of $\tilde{\omega}_2$[5]). Thus, if we exclude very high fields and the immediate vicinity of T_N, we may assume that $\tilde{\omega}_2 \gg \tilde{\omega}_1$.

When this assumption is made, Eq. (4) yields the following expressions for the eigenfrequencies of the spin system in an easy-plane antiferromagnet with an arbitrary value of ψ:

$$\text{LF branch:} \quad \left(\frac{\omega_1}{\gamma}\right)^2 = \frac{B+b}{B}H_0^2\cos^2\psi \approx H_0^2\cos^2\psi, \qquad (7)$$

$$\text{HF branch:} \quad \left(\frac{\omega_2}{\gamma}\right)^2 = aBL_0^2 + \frac{B}{B+b}H_0^2\sin^2\psi \approx \qquad (8)$$
$$\approx 2H_A{}^aH_E\left[1 - \left(\frac{H_0}{2H_E}\right)^2\right] + H_0^2\sin^2\psi.$$

Usually the low-frequency branch of antiferromagnetic resonance in an easy-plane crystal does not have a zero activation energy. There are various reasons why a gap may appear in the spectrum of the low-frequency branch. They include a weak anisotropy in the easy plane, spontaneous magnetostriction,[17] interaction between the electron and nuclear spin systems at low temperatures,[18] etc. Formally, the gap Δ can be included in the following manner:

$$(\omega/\gamma)^2 = H_0^2\cos^2\psi + \Delta^2. \qquad (9)$$

3. EXPERIMENTAL METHOD AND SAMPLES

We studied antiferromagnetic resonance in MnO using a radiofrequency spectrometer with a pulsed magnetic field, similar to that described in[19]. The frequency was varied in the 30–80 GHz range. The temperature of a sample was varied from that of boiling nitrogen to 100° C. The sample was placed at the center of a rectangular reentrant multiwave resonator. The sample could be rotated about an axis perpendicular to an external static magnetic field $\mathbf{H}_0$. The magnetic component of the microwave field was perpendicular to $\mathbf{H}_0$ and it coincided with the axis of rotation of the sample. The external magnetic field could be increased right up to 100 kOe.

We investigated antiferromagnetic resonance of MnO single crystals grown by the Verneuil method or by epitaxial growth from the vapor phase on different crystallographic planes of MgO substrates. The single crystals grown by the Verneuil method were cut along the {100} planes into cubes of ~ 1 mm edge or spheres of ~ 1 mm diameter were machined with the aid of an abrasive.[20] The epitaxial single crystals were disks of ≈ 2.5 mm diameter and ≈ 0.2 mm thick; some of them were studied still attached to the MgO substrates.

The results obtained for different samples were similar. The majority of the gaps given in the present paper represent the results of measurements carried out on a disk of 2.6 mm diameter and 0.17 mm thick, which was cut from an MnO crystal grown epitaxially on the (111) face of an MgO substrate. After cutting and polishing, the disk was annealed in hydrogen at T = 1100°C and cooled slowly. This heat treatment improved the stoichiometry of the sample and tended to reduce the internal stresses.

Since the results of resonance measurements depended strongly on the presence of the T-domain structure and on the nature of this structure, whenever possible we studied the samples (transparent thin disks) in transmitted polarized light at temperatures above and below T_N using an MIN-4 microscope.

It was possible to observe the T-domain structure in polarized light because below T_N the crystals were optically uniaxial due to the spontaneous compression along the [111] axis. Since the optical axes of different T domains were inclined at various angles with respect to the plane of polarization of light, the angular dependences of the birefringence could be used to identify T domains with different easy planes.

It was reported in[21] that thin epitaxial NiO single crystals grown on the (111) surface of MgO had the single-domain structure. This was due to the appearance of stresses in NiO-MgO bicrystals which led to preferential compression of the whole NiO film along just one of the possible ⟨111⟩ axes so that a single T domain was formed. In the case of MnO-MgO this situation did not arise because the relationship between the lattice parameters of MnO and MgO was different and the thermal expansion coefficients of these crystals were also different. Polydomain crystals of manganese oxide grew on the (100) and (111) faces of MgO. MnO crystals grown on the (111) face of magnesium oxide consisted of all four possible types of T domain (this was found in polarized light studies).

4. RESULTS OF MEASUREMENTS OF ANTIFERROMAGNETIC RESONANCE AND DISCUSSION

Figure 1 shows a typical oscillogram of antiferromagnetic resonance in an MnO single crystal. The horizontal coordinate represents a signal proportional to the external magnetic field H_0 lying in the (111) plane and making an angle of ~ 18° with the $[2\bar{1}\bar{1}]$ direction. This oscillogram has several absorption peaks which appear because, for a fixed orientation of a crystal, the field H_0 forms different angles ψ_i ($i = 1, 2, 3, 4$) with the easy plane of each of the four types of T domain. Therefore, it follows from Eq. (9) that for an arbitrary orientation of a crystal there should be four resonance peaks (at a given frequency) and they should occur at different values of H_0 corresponding to antiferromagnetic resonance in different types of T domain. When a single crystal is rotated relative to H_0, these peaks shift along the field axis, merge with one another, and form one, two, or four absorption lines.

The dependence of the resonance field on the angle of rotation of a sample φ can be calculated for any rotation axis from Eq. (9). One has to bear in mind the relationship between the angles ψ_i for each T domain and the angle φ. The solution of this geometrical problem is plotted in Fig. 2 for the case when the rotation axis is [111]. The calculated dependences are obtained on the assumption that the g factor is $g = 2$ and that $\Delta = 0$. The same figure includes the experimental results, which are in good agreement with the calculations.[4]

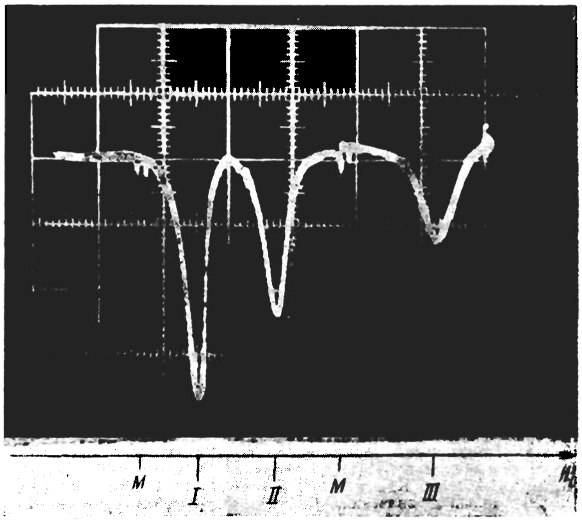

FIG. 1. Oscillogram of antiferromagnetic resonance in an MnO single crystal. The horizontal scale represents a signal proportional to H_0. The line denoted by I represents two merged absorption lines corresponding to resonance in T domains with (111) and $(1\bar{1}1)$ easy planes; II represents resonance in T domains with $(11\bar{1})$ easy planes; III represents resonance in T domains with $(\bar{1}11)$ easy planes. The two pairs of narrow peaks (denoted by M) are calibration marks of the H_0 field. T = 80°K, ω = 69.5 GHz.

The [111] axis was not chosen accidentally. The angular dependence obtained for this axis is most interesting since it allows us to observe separately the antiferromagnetic resonance lines for all four types of T domain. Moreover, the position of any one absorption line is independent of the angle of rotation of the sample and it corresponds to the minimum resonance field and (as will be shown later) to the minimum width at a given frequency. Furthermore, the smallest number of the resonance lines (two lines) observed for this rotation axis corresponds to H_0 coinciding with one of the $[01\bar{1}]$ directions. In principle, we can determine the relative number of T domains of a given type from the relative intensities of the resonance lines.

Figure 3 shows the results of measurements of the frequency dependence of the resonance field for the low-frequency branch of MnO. These results were obtained using H_0 directed along the $[01\bar{1}]$ axis. The measurements were carried out at T = 80°K. At all frequencies in the 30—80 GHz range we observed two resonance lines corresponding to $\psi = 0°$ [resonance in T domains with (111) and $(\bar{1}11)$ easy planes] and $\psi = 55°$ [resonance in T domains with $(1\bar{1}1)$ and $(11\bar{1})$ easy planes]. The theoretical dependences calculated on the assumption that $g = 2$ and $\Delta = 0$ are represented by the continuous and dashed lines corresponding to $\psi = 0°$ and $\psi \approx 55°$. It is evident from Fig. 3 that Δ must be close to 0 and its

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[4] The angular dependence of the resonance field, which we obtained earlier [3] for the [100] rotation axis, is also in agreement with the calculations.

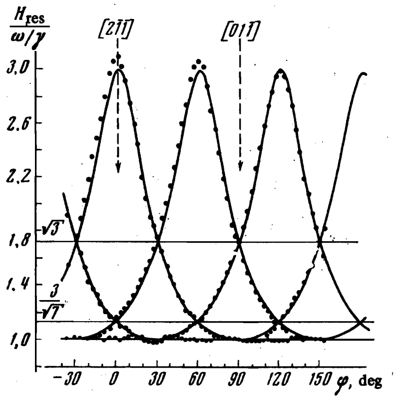

FIG. 2. Dependence of the antiferromagnetic resonance field of an MnO single crystal on the angle of rotation of the crystal relative to H_0. Rotation axis [111]; H_0 lying in the (111) plane; φ is the angle between H_0 and [2$\bar{1}\bar{1}$]; T = 80°K, ω = 69.5 GHz. The points represent experimental values and the continuous curves are calculated.

influence on the resonance field (bearing in mind the experimental error) is of importance only at the lowest of the frequencies used (30.74 GHz). Since, at this frequency, H_{res} = 10.4 ± 0.4 kOe (for the line corresponding to $\psi = 0°$), it follows from Eq. (9) that Δ does not exceed 5 kOe.

Let us now consider the angular dependence of the width of the antiferromagnetic resonance line. It is evident from the oscillogram in Fig. 1 that the resonance lines obtained at a given frequency for different values of H_0 have different widths 2ΔH, which are measured at the mid-amplitude of the absorption peak. We shall assume that the dissipation parameter Δω is independent of the angle ψ between H_0 and the easy plane. Then, $\Delta H = \Delta\omega(dH/d\omega) = \Delta H^0/\cos\psi$, where ΔH^0 is the half-width of the line for H_0 lying in the easy plane. An analysis of the experimental results obtained at 69.5 GHz and T = 80°K confirms that the suggested angular dependence of ΔH is indeed obeyed within an experimental error of ±10%.

The temperature dependences of the resonance field H_{res} and of the width of the antiferromagnetic resonance line were determined in H_0 directed along the [01$\bar{1}$] axis. When this orientation was employed, one of the absorption lines should have corresponded to $\psi = 0°$ and, as indicated by the angular dependence of ΔH discussed above, this line should have had the smallest width observable in the sample at a given temperature and frequency. Our measurements were carried out at 70.4 GHz in the temperature range 80—320°K. The temperature was determined to within ±1 deg. During any one measurement of ΔH and H_{res} the temperature was maintained to within ±0.1 deg. The temperature dependences obtained in this way are plotted in Fig. 4. We also observed two absorption lines corresponding to $\psi = 0°$ (lower row of black dots) and $\psi \approx 55°$ (upper row), whose positions along the field axis was independent of temperature up to 121°K.

The formulas for the resonance frequencies show that, in contrast to the high-frequency branch, the expression for the low-frequency spectrum does not include the anisotropy field and, consequently, H_{res} for the low-frequency branch should be independent of temperature. This is correct only when Δ^2 is small compared with H_{res}^2, which is definitely true of our measurements at 70.4 GHz. Above T_N one of the resonance lines ($\psi \approx 55°$) disappears because the anisotropy plane and the domain structure both disappear. The remaining absorption line no longer represents antiferromagnetic resonance but electron spin resonance (ESR). This disappearance of the resonance line provides a basis for a precision method of measuring T_N of MnO and similar crystals.

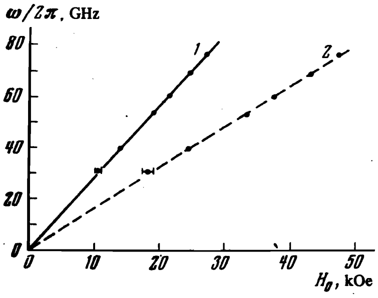

FIG. 3. Frequency dependence of the antiferromagnetic resonance field of an MnO single crystal. T = 80°K. H_0 lying in the (111) plane along the [01$\bar{1}$]. The continuous and dashed lines represent the theoretical dependences $\omega(H_0)$ for T domains whose easy planes are (111), ($\bar{1}$11), (1$\bar{1}$1), (11$\bar{1}$), respectively, inclined at angles $\psi_{1,2} = 0°$ and $\psi_{3,4}$ = 54°44'. 1) $H = \omega/\gamma$; 2) $H = (\omega/\gamma)(1/\cos 54°44')$.

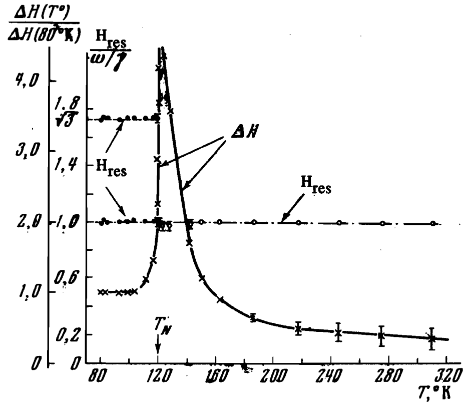

FIG. 4. Temperature dependence of the resonance field and the width of the absorption line for an MnO single crystal. H_0 || [01$\bar{1}$]; ω = 70.4 GHz. The black and white dots represent H_{res} in the antiferromagnetic and paramagnetic states, respectively. The crosses are the values of ΔH. At 80°K, ΔH = 1.2 kOe. The calculated dependences of the resonance field in the antiferromagnetic and paramagnetic states are represented by the dashed and chain lines, respectively.

The relative value of ΔH, given in Fig. 4, was obtained for that antiferromagnetic resonance line which "transformed" at T_N into an ESR line (for this line $H_{res} \approx \omega/\gamma$ both below and above T_N). At T = 80°K the half-width ΔH for the sample investigated was 1.2 kOe. However, the measurements were carried out using pulsed magnetic fields so that the value of ΔH was determined with a considerable error. In the paramagnetic region the line became narrower and the relative amplitude of the absorption of microwave power in the resonator became greater. In this case the value of ΔH could not be determined at mid-amplitude of the line. A suitable correction was made for ΔH but the error in the determination of this quantity unavoidably increased.

Nevertheless, the results plotted in Fig. 4 show clearly that the width of the antiferromagnetic resonance line is practically constant up to 100° K but ΔH begins to rise on approach to T_N. In the immediate vicinity of T_N the value of ΔH rises very rapidly; it increases severalfold within 1 deg. Above T_N the line width decreases with temperature but the fall is less rapid than the rise on the low-temperature side of T_N. A very interesting feature is the continuous fall of ΔH in the paramagnetic region even at temperatures removed by hundreds of degrees from T_N.

All the experimental results, particularly the discovery of the low-frequency "zero-gap" branch of antiferromagnetic resonance as well as the angular, frequency, and temperature dependences of this branch provide the most direct, clear, and unambiguous (compared with the results obtained by other methods) confirmation of the correctness of the accepted ideas on the magnetic structure of MnO which is a planar antiferromagnet with T domains.

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105