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Cyclotron resonance in magnetic crystals with phase separation regions

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The results of cyclotron resonance measurements in a set of magnetic crystals in which the presence of phase separation regions was already assumed are presented. It is shown that cyclotron resonance at room temperature exists in those crystals that have sufficiently high potential barriers at the boundaries of phase separation regions.

Keywords: electron resonance, phase separation regions, domain walls.

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1. Introduction

In the recently published study [1], the cyclotron resonance (CR) was reported to be observed in single crystals of iron-yttrium garnet (YIG) at room temperature. The studied single crystals were of high quality — Bragg angles in X-ray diffraction were $\Theta_b \sim 10''$. They also had low dielectric and magnetic losses. The phenomenon of CR indicated the presence of free electrons and the existence of small regions containing these electrons the volume of which was much smaller compared to that of the specimen. Such regions — Phase Separation Regions (PSR) are currently known to occur in many magnetic crystals. Previously, physical characteristics of crystals containing transition metal ions were described based on a theory that outlined various subsystems of the crystal (charge, spin, and orbital) as relatively independent. The interaction between them was usually taken into account in the form of small corrections to the main interactions within each subsystem. However, according to modern concepts, such crystals are strongly correlated systems, and an approach has been developed for them in which the interaction between different degrees of freedom was initially taken into account when writing the model Hamiltonian. At the same time, it is possible to describe a complex phase diagram of states for a number of crystals and, in particular, the occurrence, under certain conditions, of such an inhomogeneous state when the bulk of the crystal retains its initial homogeneity, but a certain number of small clusters (micro- or nanoscale) appear within which the ferromagnetic order of localized spins is realized and there are free or weakly bound electrons, causing increased local conductivity and the possibility of cyclotron resonance. The main part of the specimen, outside the clusters, as already noted, remains dielectric and retains the original magnetic order. The electron-electron interaction is critical in the formation of such states, triggering a double exchange that leads to the ferromagnetic ordering of PSR regions. The theory that takes into account the electron-electron interaction has been developed in-depth in relation to manganites $R\mathrm{Mn_2O_5}$ (R — rare-earth ions) [2–5].

In manganites, crystals of stoichiometric composition initially contain manganese ions of different valences Mn^{3+} and Mn^{4+}. Along with that, as a result of the balance of double exchange, Coulomb repulsion and Jahn-Teller energy, an inhomogeneous state with the formation of PSR turns out to be more beneficial in terms of energy. In other magnetic crystals with 3d ions, the specific mechanisms stimulating the appearance of double exchange necessary for the formation of PSR may be different. This may be the presence of a certain number of impurity ions that disrupt the charge balance or other factors, such as elastic deformations during the crystal growth, leading to the appearance of local electric fields and some „extra“ electrons. In [1], we suggested that the mechanism previously proposed in [6] is effective, according to which structural distortions occur in ferromagnets in the walls of antiparallel magnetic domains due to magnetostriction, leading to a violation of the central symmetry. As a result, local electric fields and free (or weakly bound) electrons appear, the mechanism of double exchange is activated, and as a result, regions of phase separation (PSR) occur. Since the phase separation process occurs with a balance of sufficiently strong interactions, of about $0.5-1.0$ eV, in principle, the resulting inhomogeneous state can exist up to room temperature and even higher. As already mentioned, phase separation may occur in many real crystals. There are potential barriers at PSR boundaries that provide effective shielding of the inner region from excitations existing at high temperatures in the main part of the crystal. This, as we believed in [1], provides the possibility of CR existence in YIG at room temperature. It is natural to assume that in other crystals containing PSR, at least in those in which magnetic ions have an immediate environment of oxygen ions, the potential barriers at PSR boundaries will be close in magnitude.

In this study we attempted to discover CR in some crystals, where PSR ($YCr0_3$ [7]), either had been discovered earlier or had been suggested to exist. The success, by and large, was problematic in setting up the goal for this task, since in a real experimental study one has to deal with a limited range of frequencies and magnetic fields of the measuring system (in our case, for instance, the frequency band 27−37 GHz, and range of magnetic fields 0−20 kOe). The problem is that if the assumption of the spherical shape of the Fermi surface is sufficiently justified for the cubic symmetry crystals, which made it possible to expect CR detection in a magnetic field close to the field of homogeneous ferromagnetic resonance (FMR), then for crystals with lower symmetry this is not fully reasonable. It should also be borne in mind that for different crystals, depending on the specific mechanism of PSR formation and other factors, the situation may vary significantly. For crystals with a perfect structure and virtually no impurity ions, such as YIG, it is likely that the PSRS will be mostly of the same size, although the presence of elastic deformations, magnetic history, and various random factors may lead to some variation in the size of these regions. In case of the impurity mechanism of PSR formation, such a spread is more likely.

In this paper, we did not seek to study in detail the CR characteristics in the measured crystals. It was important for us to detect CR and obtain a minimal set of data to identify the observed signals as a manifestation of cyclotron resonance. We selected orthorhombic crystals for the experiment, which were oriented so that the selected crystallographic axis **C** was directed along the magnetic field **H**. At the same time, it could be expected that with a small difference in the lattice cell parameters a and b, the cross section of the Fermi surface in the plane perpendicular to **C** axis would be close to circular and, accordingly, CR would be close to FMR in terms of field. It turned out that this condition is actually realized for our chosen group of crystals, and we were able to measure CR in them.

2. Samples and measurement procedure

We have studied the following monocrystalline specimens: $YCrO_3$, $YFeO_3$, $GdCrO_3$, $EuCrO_3$ and $NaNiF_3$. The single crystal $NaNiF_3$ was selected to test the effect of replacing the oxygen environment of magnetic ions with fluorine ions. All the crystals listed above have the structure of rhombic distorted perovskite. In the low temperature range, antiferromagnetic ordering with a weak ferromagnetic moment is realized in them. Néel temperature for the orthochromites and $NaNiF_3$ is $T_N = 140-180$ K and at room temperature they are paramagnetic. The single crystal $YFeO_3$ has Néel temperature of $T_N = 648$ K, thus, at a room temperature, its magnetic order is preserved. In all the crystals listed above, the weakly ferromagnetic moment is directed along **C** axis, which allowed them to be oriented along this axis magnetically. The specimens were shaped like plates of different thicknesses, with a volume of several cubic millimeters. At the same time, such specimens were selected that grew with a developed plane perpendicular to **C** axis. Since all crystals (with the exception of $YFeO_3$) were in a paramagnetic state at room temperature, the demagnetization effect, which is important for measuring YFeG, did not matter for them. In case of $YFeO_3$ it is also of small importance, since the weakly ferromagnetic moment is small (of about 10^{-2} of the sublattice magnetization). More detailed information about the specimens will be provided when discussing the results for specific crystals.

As in paper [1], the measurements were carried out using a magnetic resonance spectrometer with low-magnetic modulation. The measuring cell consisted of a system of two horns, between which a specimen was placed in a free space. A wave of TE_{10} type was excited in thehorns, so that the sample was in the field of the electromagnetic TEM wave, the plane of polarization of which was determined by the corresponding orientations of the horns. Two configurations with mutually orthogonal orientation of the electromagnetic field vectors **e** and **h** were used. As well-known, the configuration when e is perpendicular to the constant magnetic field **H** is optimal for CR excitation (C configuration), and configuration with **h** perpendicular to **H** — for FMR excitation (F configuration). It is precisely this polarization difference between the excitation conditions of CR and FMR that we used mainly for their identification. The electromagnetic field at the specimen location had an almost flat front, with a sufficiently high polarization isolation between the configurations Cand F — the ratio of microwave power passing through the measuring cell with the matched and crossed positions of the horns was about 10^3.

3. Measurements and discussion

First of all, let's consider the results for $YCrO_3$ crystal. As already noted, $YCrO_3$ is a rhombically distorted perovskite with a dedicated axis **C**. The spatial group $D_{2h}^{16}-Pbmn$, the lattice cell contains four formula units. Ions Cr^{3+} are located in oxygen octahedra. The axes of these octahedra are slightly deviated from **C** axis along which they are fixed in undistorted perovskites. The temperature of destruction of the antiferromagnetic order $T_N = 142$ K. Previously, we studied in detail the dielectric and magnetic properties of $YCrO_3$ [7] and it was shown that regions of phase separation in these crystals exist up to temperatures above 350 K. In the present study, as already noted, the axis **C** of the sample was oriented along **H**, and the developed plane (**a, b**) was perpendicular to the direction of propagation of the microwave wave. The dependences of the microwave power passing through the specimen on the magnetic field $P(H)$ were measured. The rate of change of the magnetic field was10−15 Oe/s. As in [1], the microwave power at the input of the measuring cell was measured in a relative manner by voltage at the detector. At the same time, it was possible

to compare power levels with different measurements, but not measure its absolute value. The value P_0 corresponds to power passing through the cell with a crystal in field $H = 0$.

Figures 1–5 shows the field dependences of signals for different incident microwave power values (P_0) at

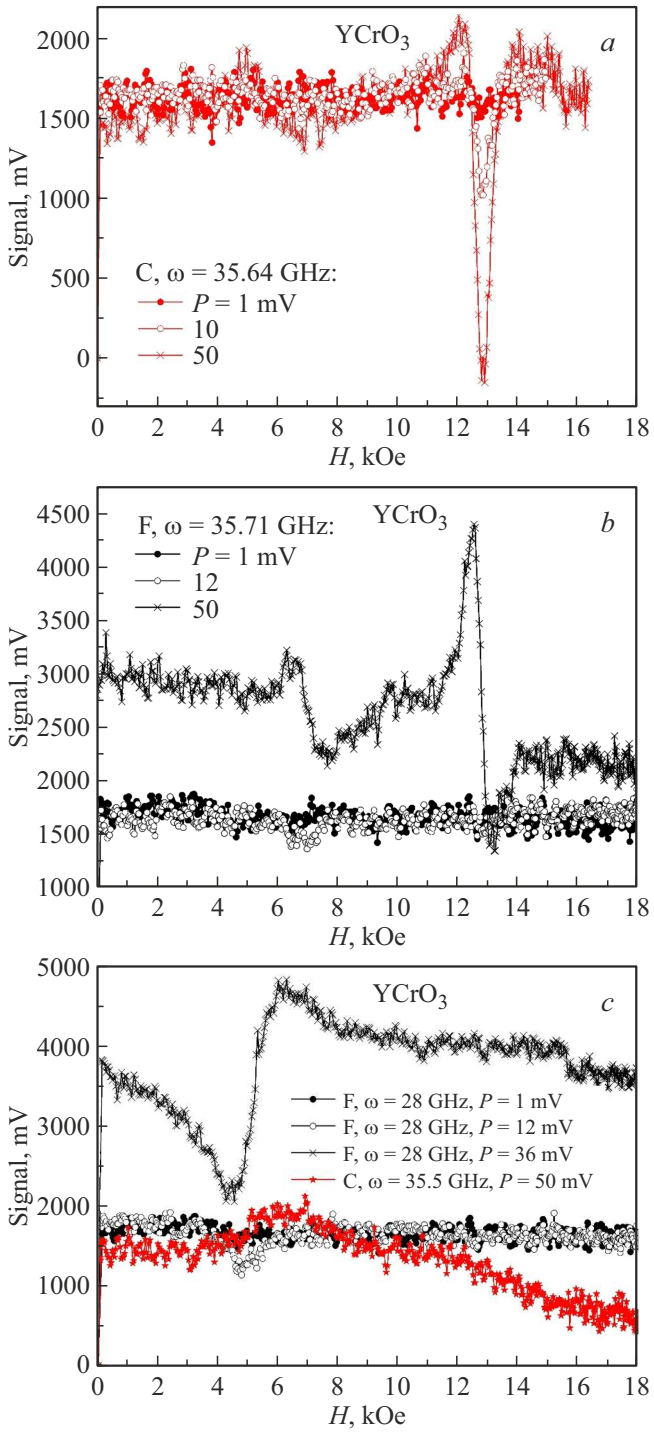

Figure 1. Dependence of the intensity of the observed signals on the magnetic field at C and F configurations and at different levels of microwave field power for $YCrO_3$ crystals. The figures (a) and (b) illustrate the dependencies at $\omega > \omega_c$ (ω_c — critical frequency), in figure (c) — for $\omega < \omega_c$.

room temperature. All graphs corresponding to the configuration C is shown in red, and F — in black. It should be emphasized, first of all, that since the crystal is in a paramagnetic state at room temperature, all the resonance peaks observed in the dependence $P(H)$ could be considered related to the PSR, and not to the main volume of the crystal. On Figure 1, a the measurement results are given in the configuration C, i. e., optimal for the excitation CR. At the lowest microwave power levels ($P_0 \sim 1$ mV), no obvious signals were observed. At $P_0 \approx 10$ mV, a resonant peak appears at a magnetic field close to $H_\omega = \omega/\gamma$ (ω — the frequency at which measurements were made, γ — gyromagnetic ratio). At a power of P_0 close to 50 mV this resonant peak increases significantly and along with it a fairly wide absorption band appears, the center of which is located near $H = H_\omega/2$. The resonant peak at $H = H_\omega$ we identified as CR. The absorption band is apparently caused by nonlinear processes in a system of ferromagnetic like ordered localized spins. The mechanism of its occurrence can be understood on the basis of ideas about the parametric excitation of spin waves in ferromagnets [8,9].

A certain background of thermally equilibrium spin waves is known to be present in all magnetically ordered materials. To a linear approximation, spin waves are independent. In the nonlinear mode, a relationship arises between them, as well as between them and homogeneous precession, which is greatly dependent on the amplitude of the exciting magnetic field **h**. Starting from a certain amplitude h_c, there is an intense oscillation in their intensity, and the thresholds for both longitudinal and transverse pumping are quite low, i. e. about $(10^{-2}–10^{-3})\Delta H$, where $\Delta H$is the width of the line of a homogeneous FMR [8]. The minimum thresholds naturally correspond to lower-order processes — excitation of pairs of spin waves with a wave vector **k** and $-$**k**. The absorption is related with this process and forms a band $1/2H_\omega$ (SW band) at H. Intensely „swung“ spin waves excite homogeneous precession, which in turn, due to the dipole-dipole interaction excites CR. The resulting system of coupled oscillators redistributes energy in such a way that less intense excitations are amplified and more intense ones are somewhat weakened. As can be seen from Figure 1, a the resonant peak corresponding to CR and the absorption in the SW band are directed in different directions. Generally speaking, in the simplest model, this should be the case, since the spin excitations correspond to right precession ($\chi > 0$), and CR is diamagnetic ($\chi < 0$), i. e., the corresponding signals have opposite phases (χ — the actual part of microwave magnetic susceptibility). However, in reality, taking into account the anisotropy leading to elliptical precession, the presence of a connection between FMR and CR, and a number of other factors, the shape and mutual orientation of the absorption lines may be different.

Figure 1, b are given in the configuration F. At low and medium microwave power levels ($P_0 < 10–12$ mV), no signals are visible. At $P_0 = 50$ mV, an intense spin-wave

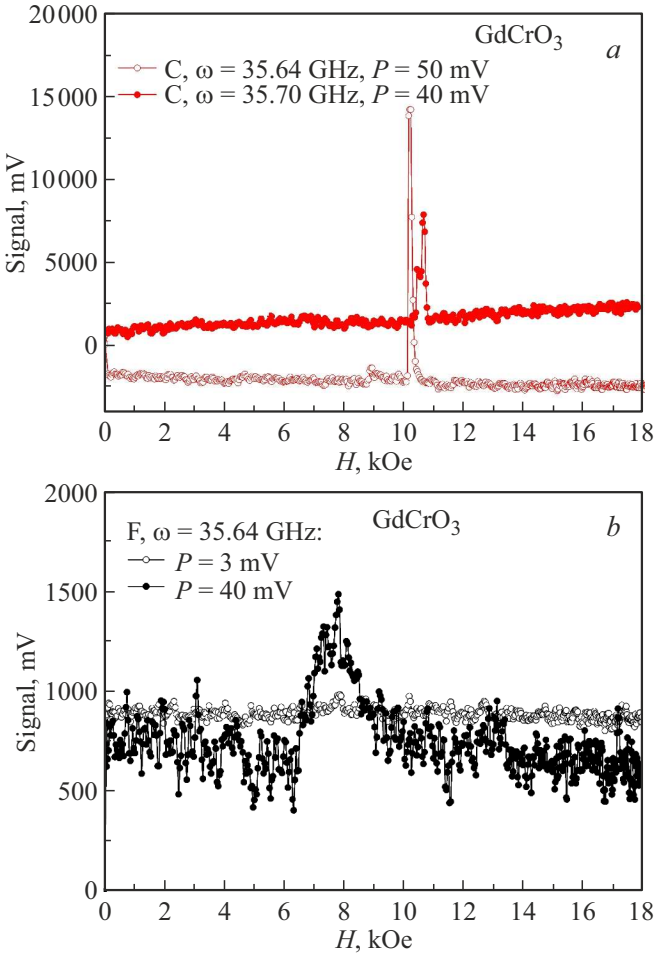

Figure 2. Intensity of the observed signals versus magnetic field at configurations C and F (figures (a) and (b), respectively) and at different power levels of the microwave field for $GdCrO_3$ crystals.

band (SW band) occurs and a resonant peak near $H = H_\omega$. The observed pattern corresponds to the above-mentioned scenario of parametric excitation of spin waves. With a pretty intense microwave pumping, $h > h_c$ (h_c — threshold amplitude) spin waves are excited, which form a SW band, and then a homogeneous precession. Further, CR is excited due to dipole-dipole bond. Thus, in the nonlinear threshold mode, a system of coupled oscillators arises independently of the polarization of the microwave excitation field. It should be stressed that results illustrated in Figure 1, a and b were obtained when measured at frequencies $\omega > \omega_c$, higher than some critical value ω_c, which for $YCrO_3$ is equal 32 GHz. When measured at frequencies $\omega < \omega_c$ at any P_0 a SW-band occurs, but no any resonance signals are observed (Figure 1, c). As can be seen from Figure 1, c and in the C configuration, even at maximum P_0, there is no resonant CR signal. The most likely explanation for these results is that at low frequencies, the diameter of the electron orbit at CR becomes larger than the size of the PSR in the corresponding direction. In this case, effective electron scattering occurs at the boundaries of PSR, and CR disappears. We do not have independent data on the size of PSR in $YCrO_3$, but we can make an order-of-magnitude estimate based on the ratio:

$$R_C = \frac{v^\perp}{\omega}, \quad v^\perp \sim v^{\mathrm{F}} = \frac{\hbar}{m_c (2\pi^2 N/V)^{1/3}} \qquad (1)$$

here R_c — cyclotron radius, ω — frequency of exciting microwave field, $v^\perp$ — orbital speed of electron, v^{F} — velocity of electrons on the Fermi surface, $\hbar$ — Planck constant, m_c — cyclotron mass of electron, N/V — concentration of electrons. If we assume that the cyclotron mass is approximately equal to the mass of a free electron, and the electron concentration is $\sim 10^{13-14}\,\mathrm{cm}^{-3}$, as in YIG [1.10], then we get the value $R_c \sim 10^{-5}\,\mathrm{cm}$, which is close to the estimate of PSR size for manganites made based on X-ray data [10,11]. It should be noted that the critical frequency ω_c is not a well-defined value for different crystals and even for crystals of the same composition, due to the dispersion of PSR in size, magnetic history and other factors. It should also be noted that the above considerations about the nature of the SW band and other manifestations of nonlinearity are purely qualitative in nature. A well-founded analysis of the mechanisms of their occurrence requires the theory of magnetic linear and nonlinear dynamics of PSR, which is currently missing.

Let us now consider the results for other crystals that have been studied in less detail than $YCrO_3$. Figure 2 illustrates the data for $GdCrO_3$. The crystalline structure of this crystal is similar to $YCrO_3$, the temperature is not higher than $T_N = 170\,\mathrm{K}$, so it is also in a paramagnetic state at room temperature. In configuration C (Figure 2, a), a clear signal of CR is observed. In contrast to the previously discussed cases, it is slightly more offset from the value $H_\omega = \omega/\gamma$. Perhaps this is due to the inaccuracy of the crystal orientation, since repeated measurements at a close but slightly different frequency gave a similar result. In F configuration (Figure 2, b), the crystal exhibits behavior similar to $YCrO_3$-no signals at small P_0, SW band at large P_0, and the center of the SW band is also slightly shifted relative to the position $H = 1/2H_\omega$.

Figure 3 illustrates the results for $EuCrO_3$ crystal. It can be seen that as in C configuration (Figure 3, a), and in F configuration (Figure 3, b), the dependencies $P(H)$ are similar to the GdCrO crystal$_3$, with at the same time, the amplitude of the resonant peak CR is much smaller and for their excitation they require high power P_0. Apparently, this is due to the greater conductivity of this crystal at room temperature.

Figure 4 illustrates the results for $YFeO_3$ crystal. This crystal has the same crystal and magnetic structure as the orthochromites discussed above, but, unlike them, it has a stronger exchange and, accordingly, a higher Néel temperature ($T_N = 648$,K). At room temperature, it is in a magnetically ordered state, namely, it is an antiferromagnet with weak ferromagnetism. Eigen antiferromagnetic excitations have a large energy slit ($\sim 300\,\mathrm{GHz}$) and

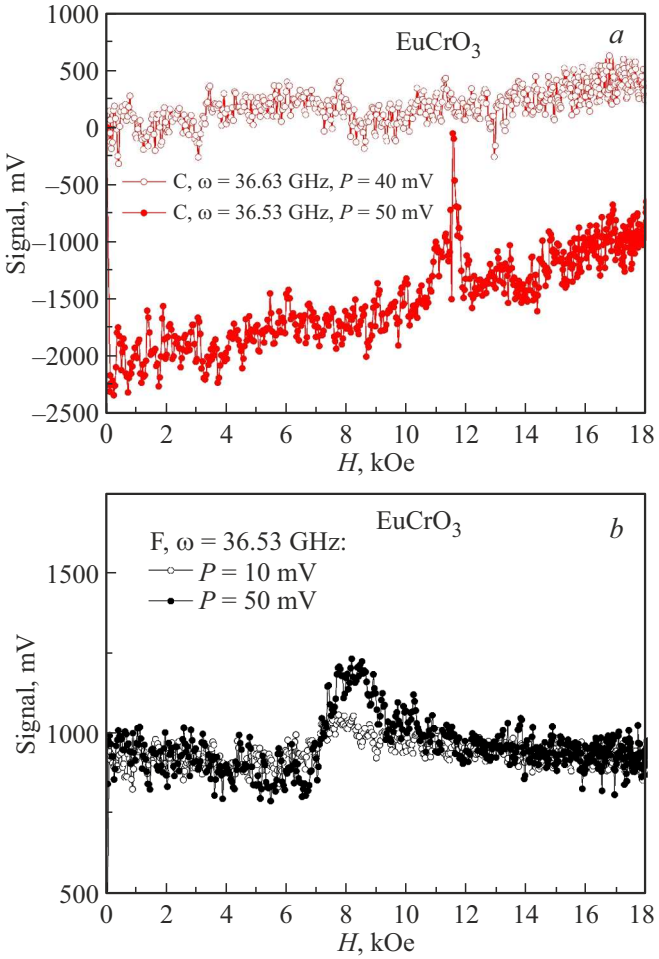

Figure 3. Intensity of the observed signals versus magnetic field at configurations C and F (figures (*a*) and (*b*), respectively) and at different power levels of the microwave field for $EuCrO_3$ crystals.

should not noticeably affect magnetic oscillations in PSR in 8-mm range. The weakly ferromagnetic moment is small in magnitude and rigidly fixed in direction by Dzyaloshinsky interaction, so the presence of this moment should also not affect the magnetic dynamics of PSR. However, in reality, the magnetic dynamics of PSR in $YFeO_3$ differs somewhat from the dynamics of orthochromites in the paramagnetic state. Perhaps this is due to the presence of a magnetically ordered matrix. In C configuration there are no significant differences: starting from $P_0 = 4$ a resonant peak appears near $H_\omega = \omega/\gamma$mV, which increases with the growing microwave power. At $P_0 = 50$ mV two close intense peaks are observed, and their position in the field is slightly shifted from the value of H_ω. We interpret these peaks as interconnected CR and homogeneous precession of localized spins (FMR). In F configuration, with an increase in microwave power, a SW band appears and then a resonant signal, also slightly shifted in the field from H_ω. This signal corresponds to excitation in the nonlinear regime of homogeneous spin precession (FMR). When measured at a low frequency (28.35 GHz) in C configuration no any resonant signals are observed, and in F configuration at $P_0 \approx 50$ mV there is a resonant peak, and its position in the field practically corresponds to the value of H_ω, i. e., a shift associated apparently with the presence of a connection between FMR and CR is missing. This indicates that in $YFeO_3$, as in orthochromites at these frequencies, CR is not excited due to scattering at boundaries.

As already mentioned, we also measured $NaNiF_3$ crystal in order to find out how the replacement of the oxygen environment of magnetic ions affects the environment of fluorine ions. $NaNiF_3$ crystals are close to orthochromites both crystallographically and magnetically. They also have the structure of rhombically distorted perovskite (spatial group $D_{16}^{2h}-Pbnm$). Parameters *a, b* and *c* of elementary cells $NaNiF_3$ and $YCrO_3$ are close (5.36, 5.52, 7.69 Å and 5.24, 5.18, 7.69 Å, respectively). Figure 5 shows the results of measurements. In C configuration under no P_0 the resonant signals were observed. In F configuration, when magnified P_0, starting approximately from 15–20 mV, a SW

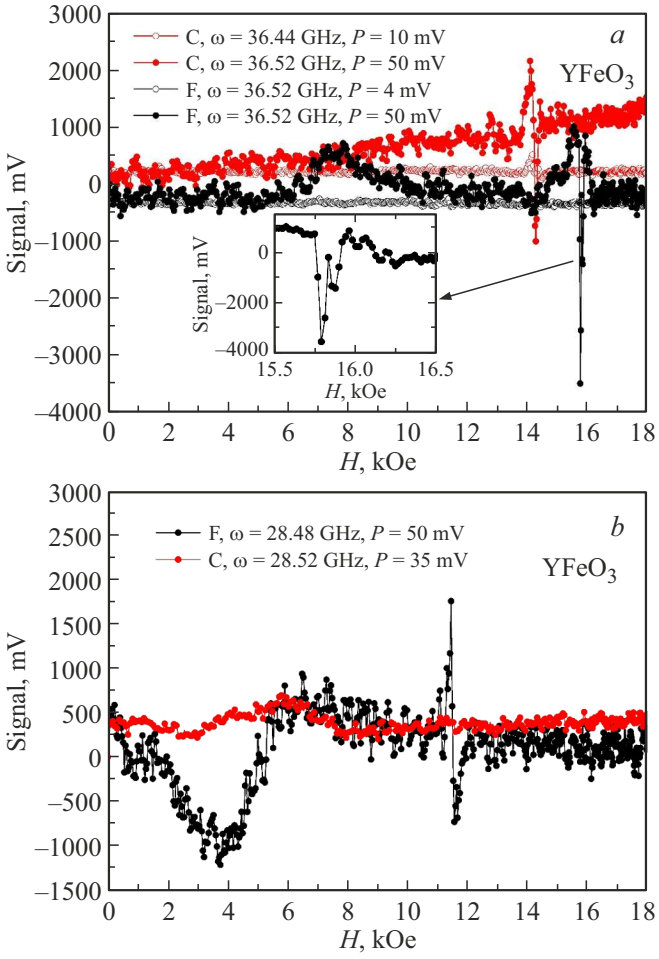

Figure 4. Dependence of the intensity of the observed signals on the magnetic field at C and F configurations and at different levels of microwave field power for $YFeO_3$ crystals. The figures (*a*) and (*b*) show the dependencies at different frequencies. In the insert — zoomed in for F configuration at $\omega = 36.52$ GHz, $P = 50$ mV.

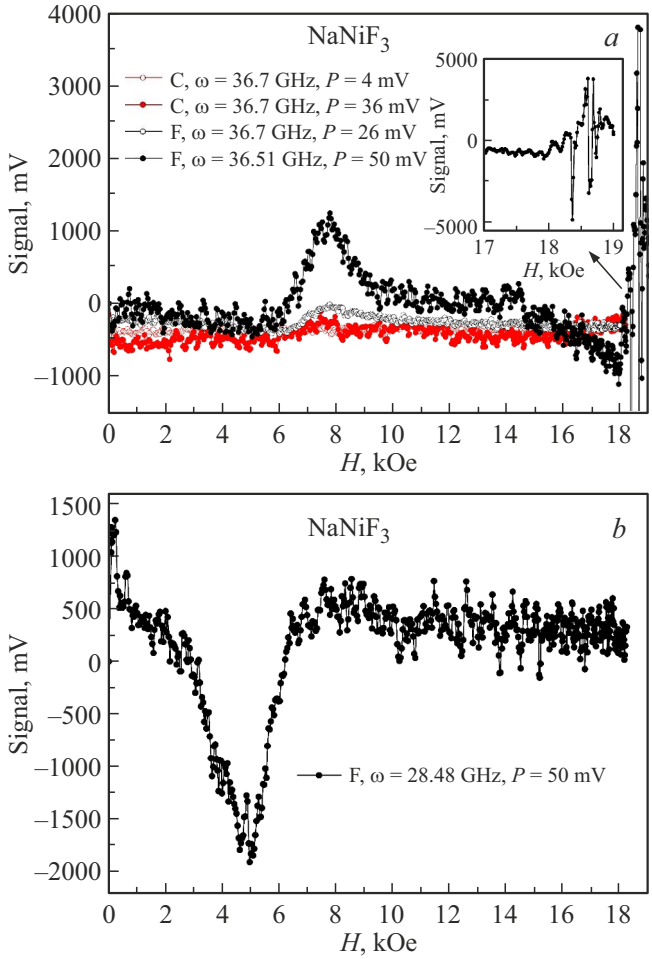

Figure 5. Intensity of observed signals versus magnetic field at C and F configurations and at different levels of microwave field power for $NaNiF_3$ crystals. In the insert window — enlarged scale 17–19 kOe for F configuration at $f = 36.51$ GHz, $P = 50$ mV.

band appears and its center, as in previously measured cases, is close to $H = \omega/2\gamma$. At $P_0 \approx 50$ mV, an intense resonant signal appears, more precisely, two close signals, and their position in the field is radically different from $H_\omega = \omega/\gamma$. When measuring at low frequencies (28.48GHz) in F configuration at maximum $P_0 = 50$,mV, there are no signals except for the intense SW band. The situation with $NaNiF_3$ may have different explanations. It is possible, for example, that the resonance field of CR is slightly larger than the maximum magnetic field for our spectrometer, and therefore CR is not observed in linear and weakly nonlinear modes. At $P_0 \geq 50$ mV, i. e. with a sufficiently high amplitude of the microwave field, in F configuration, as the coupling with excitations in the spin system rises, CR shifts towards smaller fields approaching FMR, which, in turn, shifts towards larger fields, approaching with CR. As a result, both signals appear in magnetic fields close to the maximum. It is also possible that CR at the fundamental frequency is not excited due to scattering at PSR boundaries, and the resonant signals that we observe at $P_0 = 50$ mV, under conditions of highly developed nonlinearity, are a bundle of FMR and a high-frequency harmonic of the fundamental CR, which fits into a PSR of a given size. Of course, both versions need to be verified, which requires additional experimental studies, which have not been conducted within this study.

4. Conclusion

Thus, the results of the conducted studies have shown that cyclotron resonance exists at room temperature in a large group of real magnetic crystals, in which, for various reasons, the phase separation regions (PSR) have arisen and exist at room temperature. Despite the small volume of these regions, relative to the volume of the main crystal, due to the electro-dipole nature of cyclotron resonance, its intensity is high, and it can significantly affect the magnetic dynamics in the corresponding frequency range. The possibility of cyclotron resonance at high temperatures is provided by the presence of potential barriers at PSR boundaries, which effectively shield the internal volume of the PSR from excitations in the bulk of the crystal.

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Conflict of interest

The authors of this paper declare that they have no conflict of interest.

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