Unidirectionality of the saturating action of an external magnetic field on resonant absorption in thulium and dysprosium orthoferrites at submillimeter wavelengths

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A unidirectional hysteresis-independent saturating action of an external magnetic field H_0 on the natural antiferromagnetic resonance in the domain walls of the orthoferrites $TmFeO_3$ and $DyFeO_3$ was observed. The gist of the effect is that application of a positive field $H_0^+ \approx +100$ Oe parallel to the c axis causes the resonant absorption to vanish together with the domains, whereas in a field H_0 of opposite sign the resonant absorption is preserved when the domains vanish. When the samples are rotated 180° (so that the front and rear faces change place relative to the H_0^+ direction) the resonance vanishes when the sign of H_0 is reversed. Observations of the domain structure by magneto-optics and powder-pattern methods shows that the domains (the magnetization of the samples) vanish at $H_0 \approx \pm$ (50–100) Oe independently of the sign of H_0. The hypothesis is advanced that the unidirectional action of H_0 is due to the onset, in the orthoferrites, of a state similar to unilateral (exchange) anisotropy, causing the domains and the domain walls to vanish in one direction of H_0, whereas in a field of opposite sign the domain walls are preserved, when the domains vanish, in the form of 360-degree walls that are observable by resonant absorption.

Observation and study of a new type of resonant absorption in the orthoferrites $TmFeO_3$ and $DyFeO_3$, namely natural antiferromagnetic resonance in the domain walls, was reported in Refs. 1–3. Resonant absorption of this type is due to interaction of submillimeter radiation with high-frequency oscillations that can exist in domain walls of certain two-sublattice magnetic systems alongside the translational low-frequency oscillations of the domain walls. The possibility of existence of two resonance branches for a transition layer was demonstrated theoretically in the case of compensated antiferromagnets.[4]

It was observed in the present study that in $TmFeO_3$ and $DyFeO_3$ the weakening of the resonant absorption in the domain walls, down to complete vanishing under the action of H_0, depends on the sign of H_0. Data were obtained on the weakening of the resonant absorption as a function of the polarization and of the direction of propagation of the microwave, of the sample orientation in the magnetic field, and of the sample thickness; the effect of magnetization reversal on the characteristics of the resonant absorption was also investigated. To determine the polarization transformation contribution due to the Faraday effect, we investigated the Faraday rotation of the polarization plane at millimeter wavelengths as a function of the wavelength. The domain structure was observed by magneto-optic and powder-pattern methods. The experiments led to the conclusion that in $TmFeO_3$ and $DyFeO_3$ the saturating action of the external magnetic field on the resonant-absorption intensity is unidirectional because of the crystal properties. It will be shown that this effect is not connected with: 1) wave interference, 2) the action of different electromagnetic waves and conversion of one normal wave into another, or 3) Faraday rotation of the polarization planes. Experiments were performed aimed at observing the practically symmetric action of fields H_0^+ and H_0^- on the vanishing of the domains (sample magnetization). It is shown that the effect observed is not connected with the asymmetry of the hysteresis loop.

1. PROCEDURE

We used in the experiments $TmFeO_3$ and $DyFeO_3$ samples grown by crucibleless zone melting with radiant heating.[5] The samples were plates 0.5–2 mm thick and with transverse dimensions from 5 to 10 mm. The single crystals were cut perpendicular to the c axis, which is the easy-magnetization axis in orthoferrites.

The experiments were performed with a quasioptic spectrometer with the samples placed in a horn-waveguide system, and were next repeated with the samples placed in diaphragms. A plane wave propagating along the c axis was normally incident on the sample. The change of the microwave polarization (rotation through an angle φ) was produced by a frequent-period grating or by rotating the samples about the c axis. The receiver was an n-InSb crystal at liquid-helium temperature, equally sensitive to any polarization.

The results that follow were obtaining by reducing the plots of the sample transmission vs the wavelength λ in the 1.2–0.7 mm range in the absence of H_0 and with H_0 applied parallel to the c axis, and for different polarizations of the microwave. Typical plots of these dependences for the resonant region are shown in Fig. 1 (T is the sample transmission in the absence of H_0, and T_H^+ and T_H^- are the transmissions upon application of positive and negative fields). The transmission-measurement accuracy was about 5%.

The curves of Fig. 1 reflect the jaggedness of the frequency characteristic of the oscillator and the interference of the waves reflected from the front and back faces of the sample. It is nonetheless clearly seen that application of an external constant field H_0 affects only the magnitude of the trans-

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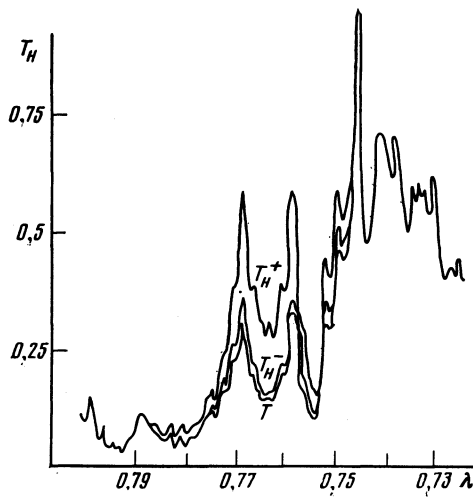

FIG. 1. Relative transmission for $TmFeO_3$ sample 0.76 mm thick vs the wavelength λ (T—at $H_0 = 0$, T_H^-—when a negative magnetic field H_0^- is applied, T_H^+—when a positive magnetic field H_0^+ is applied).

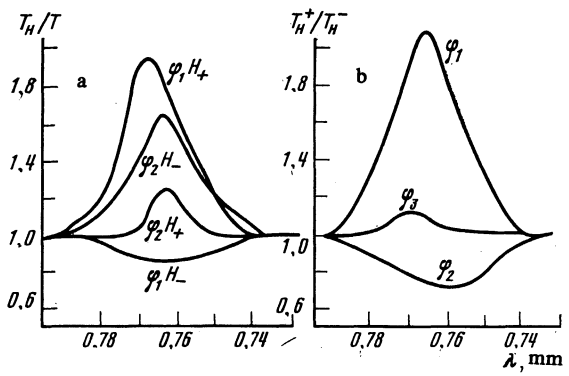

FIG. 2. Wavelength dependences for a $TmFeO_3$ sample 0.76 mm thick: a) of the weakening of the resonant absorption upon application of H_0^+ and H_0^- for different polarizations φ of the microwave, $|H_0| = 600$ Oe; b) of the ratio T_H^+/T_H^- for different φ, $|H_0| = 600$ Oe.

mission and only in the resonance region. The amplification of the signal passing through the sample (the weakening of the resonant absorption) is not the same for the positive and negative fields H_0^+ and H_0^-. A study of this circumstance is of great interest since, as will be shown below, the field action that causes the domains to vanish is almost symmetric, and the hysteresis loops are also symmetric. It can also be seen from Fig. 1 that the resonant wavelength remains unchanged (within 0.5%) and the interference pattern for the transmission is preserved when the magnetic field is applied (proof of the latter is that the frequency peaks are not shifted).

To eliminate the influence of the frequency characteristic of the oscillator and of the interference beats of the waves in the sample, we transform below from curves of the type shown in Fig. 1 to plots of the ratios T_H/T which describe the degree of weakening of the resonant absorption, following application of H_0, on the parameters that vary in the experiment.

2. EXPERIMENT RESULTS

1. We present below a number of plots for $TmFeO_3$. It can be seen from Fig. 2a that in the case of the polarization φ_1 the action of the negative field H_0^- does not weaken the resonant absorption ($\varphi_1 H_-$ curve, the ratio T_H/T does not exceed 1), whereas application of the positive field H_0^+ weakens the resonant absorption greatly ($\varphi_1 H_+$ curve). The $\varphi_2 H_-$ and $\varphi_2 H_+$ curves correspond to the polarization φ_2, at which a weakening of the resonant absorption[1)] is observed at H_0^+ and H_0^-. Figure 2b shows the dependence of T_H^+/T_H^- on λ. The values $T_H^+/T_H^- = 1$ correspond to a polarization angle φ_3 at which the resonant absorption is equally weakend by the action of H_0^+ and H_0^-. The values $T_H^+/T_H^- > 1$ correspond to φ_1, at which H_0^+ weakens the resonant absorption more strongly than H_0^-, while the values $T_H^+/T_H^- < 1$ mean that at the corresponding φ_2 the resonant absorption is more weakened by the negative field H_0^-.

The field strengths sufficient to saturate the samples were $\pm$ 600 Oe, and in stronger fields no further change of the transmission was observed.

Figure 3 shows a plot of T_H^+/T_H^- vs φ at the resonant wavelength $\lambda \approx 0.77$ mm. The maximum value of T_H^+/T_H^- corresponds to the angle φ at which H_0^+ leads to maximum weakening of the resonant absorption while H_0^- practically does not weaken the resonance. The minimum value of T_H^+/T_H^- corresponds to φ at which the maximum action on the resonant absorption is exerted by the field H_0^-. It can be seen from Fig. 3 that the dependence of T_H^+/T_H^- is periodic, with the values of φ corresponding to the maximum and minimum T_H^+/T_H^- separated by $\approx 90°$.

Figure 4 shows a plot of the weakening T_H/T of the resonant absorption, following application of H_0, on the magnitude and the sign of H_0 at different angles φ of the sample rotation about the c axis (T and T_H are the transmissions of the sample at the resonance wavelength in the absence and presence of H_0, respectively). It can be seen from curve 1 of Fig. 4 that at the polarization φ_1 the resonant absorption is noticeably weakened (the transmission T_H is increased) already in a magnetic field $H_0^+ = 200$ Oe, the absorption practically vanishes in fields $H_0^+ = +500$ Oe,

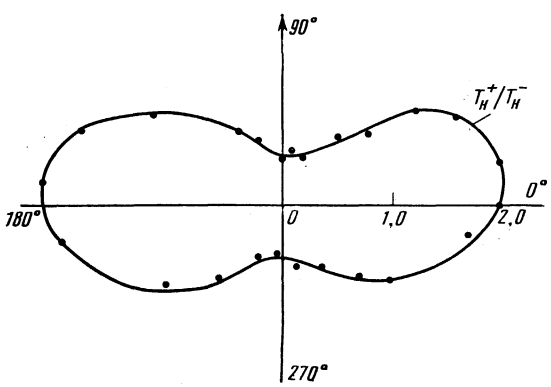

FIG. 3. Transmission ratio after application of $H_0^+ = +600$ Oe and $H_0^- = -600$ Oe for $TmFeO_3$ vs the rotation angle φ of the microwave polarization angle ($\lambda = 0.77$ mm, sample thickness $d = 0.76$ mm).

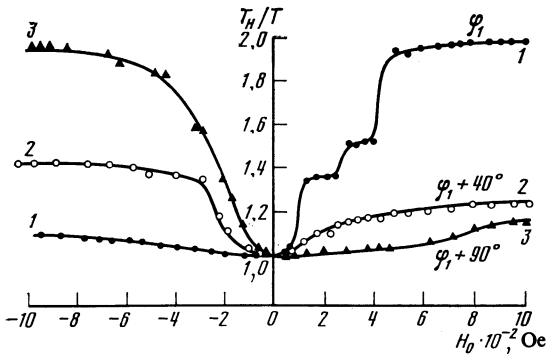

FIG. 4. Weakening of resonant absorption for $TmFeO_3$ as a function of the magnitude and sign of H_0 for different φ ($\lambda = 0.77$ mm, $d = 0.76$ mm).

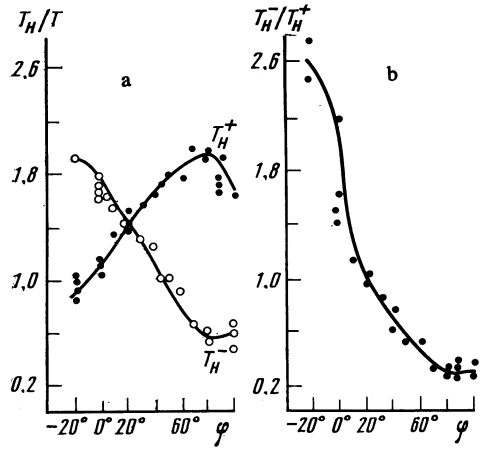

FIG. 5. Variation, with φ, in $DyFeO_3$ ($\lambda = 0.79$ mm, $d = 2$ mm): a) of the weakening of the resonant absorption under the action of $H_0^+ = +600$ Oe (T_H^+ curve) and $H_0^- = -600$ Oe (T_H^- curve), b) of the ratio T_H^-/T_H^+, $|H_0| = 600$ Oe.

whereas a field of opposite sign, $H_0^- = -1000$ Oe, has practically no effect on the resonant absorption. When the polarization is changed ($\varphi_2 = \varphi_1 + 40°$) the weakening T_H/T of the resonant absorption decreases when H_0^+ is applied, but an increase of T_H/T is correspondingly observed when H_0^- is applied. When the sample is rotated 90° about the c axis (curve 3, $\varphi_3 = \varphi_1 + 90°$), the magnetic field H_0^- begins to exert an action analogous to that of H_0^+ at φ_1, but the field H_0^+ hardly acts on the resonance at φ_3, just as the field H_0^- at φ_1. It can be seen from Fig. 4 that at certain polarizations φ_1 and $\varphi_1 + 90°$ at a maximum weakening T_H/T of the resonant absorption is observed, equal to the value T_0/T of the resonant absorption. This is the case of total vanishing of the resonant absorption upon application of H_0^+ at φ_1 and of H_0^- at φ_3. At intermediate values of φ the resonant absorption weakens partially and is observed when either H_0^+ or H_0^- acts.

Similar plots were observed for other samples with different transverse dimensions (the samples were placed then in different diaphragms) and thicknesses.

2. The dependence of the weakening of the resonant absorption on the sign of the magnetic field was investigated also in the orthoferrite $DyFeO_3$. Figures 5a and 5b show plots of the weakening of the resonant absorption, T_H/T, vs φ at $\lambda_{res} = 0.79$ mm under the action of H_0^+ and H_0^-, and plots of T_H^-/T_H^+ vs φ. It can be seen from Fig. 5a that at a certain value of φ there is observed a maximum of T_H^+/T (maximum weakening of the resonant absorption) when H_0^+ is applied; a minimum of T_H^-/T is observed at the same value of φ in the case of H_0^-.

Just as with the thulium orthoferrites, experiments were performed on $DyFeO_3$ for the purpose of observing the dependence of T_H/T on the magnitude and sign of H_0 at different φ. Strong weakening of the resonant absorption was observed already in fields $H_0^- = -50$ Oe (for φ_1), while at $H_0^- = -100$ Oe saturation of the sample set in and the resonant absorption vanished (in this case T_H/T is practically equal to T_0/T), but a field $H_0^+ = +600$ Oe had practically no effect on the resonant absorption (T_H/T was close to unity); at $\varphi_2, \varphi_3, \varphi_4$, etc., the field H_0^+ began to influence the resonance while the action of H_0^- was preserved, with T_H/T decreasing for H_0^- and increasing for H_0^+. At $\varphi_5 = \varphi_1 + 90°$ a field $H_0^+ = +50$ Oe lead to a weakening of the resonant absorption, which vanished at $H_0^+ = +100$ Oe, but a field $H_0^- = -600$ Oe had no influence on the weakening of the resonant absorption.

When all the cited dependences on H_0 were measured in fields 200–400 Oe, the samples became magnetized and remained so (retaining correspondingly their transmissivity) after H_0 was turned off. Each new curve was plotted after demagnetizing the samples by cooling from the Curie point. The dependences of T_H/T on the magnitude and sign of H_0 were reproducible for each value of φ.

3. The domain structure in the investigated samples was observed by magneto-optic means using the Faraday effect (for thin samples) and by powder patterns (for thick samples). These were in the main samples with a through labirynth-like domain structure with antiparallel orientation of the magnetizations.

We observed rather well-known patterns of variation of the domain structure: when an external magnetic field was applied along the c axis the domains with unfavorable magnetization orientations decreased in size at the expanse of growth of domains with favorable orientations. Experiments performed with different samples have shown that in fields 50–200 Oe the samples become magnetized (one domain with favorable magnetization orientation is observed) independently of the sign of H_0 in practically equal fields (with slight deviation, 20–30 Oe, from symmetry). Variation of φ does not influence the magnetization process.

4. The change of the resonant absorption when the magnetization is reversed can be seen from Figs. 6a and 6b. The point O ($H_0 = 0$) with $T_H/T = 1$ corresponds to the resonance state for a demagnetized sample. With increasing positive magnetic field H_0^+ (Fig. 6a for $\varphi = 90°$) the resonant absorption weakens (the transmission T_H increases), in fields + (100–150) Oe the resonance vanishes and saturation of the sample sets in, and further increase of the field H_0^+ produces practically no change in the transmission (curve OBC). As can be seen from observation of the domain struc-

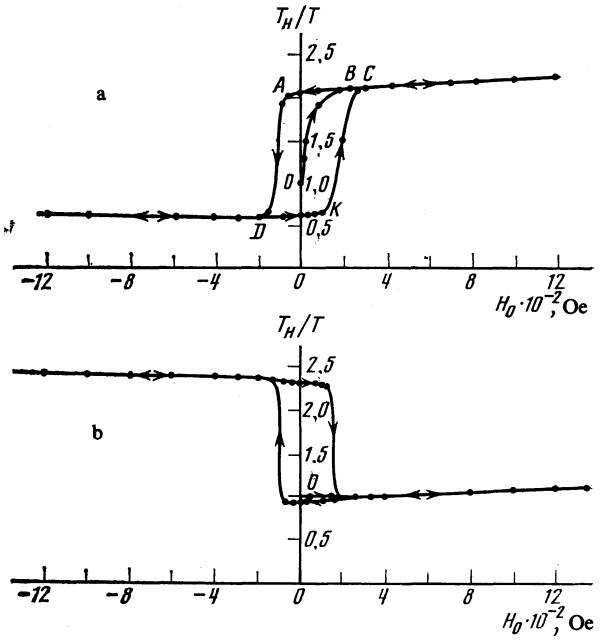

FIG. 6. Weakening of resonant absorption for $DyFeO_3$ following reversal of magnetization ($\lambda = 0.79$ mm, $d = 2$ mm): a) $\varphi = 90°$, b) $\varphi = 0°$.

ture by magneto-optics methods and by powder patterns, at these field values the domains vanish and the samples become magnetized. As the value of H_0^+ that saturated the sample is lowered, starting with a certain value of H_0^+, the change of T_H/T will no longer follow OB and will not reach unity at $H_0^+ = 0$, i.e., no resonant absorption will be observed in a field $H_0^+ = 0$. We are dealing here with a hysteresis phenomenon wherein the domain structure is not restored in zero field after magnetization of the sample, and the resonant absorption is accordingly likewise not restored. For T_H/T to reach the resonant value it is necessary to apply a reverse magnetic field $H_0^- = -100$ Oe. Further increase of the negative field will saturate the sample in the opposite direction, but this direction corresponds to a resonance state in the sample, even though there are no domains and the sample is in a magnetized state. On returning to $H_0^- = 0$ (decreasing the negative field), this state is preserved to fields $H_0^+ = +100$ Oe, and then T_H/T changes jumpwise, followed again by vanishing of the resonance and saturation of the sample.

There are thus two states of saturation, with applied fields $H_0^+ \geqslant +100$ Oe and $H_0^- \leqslant -100$ Oe, in which there are no domains but for one direction of H_0 the resonance is preserved, whereas a field of opposite direction causes the resonance to vanish simultaneously with the domains. The transition from one state to the other follows an almost symmetrical loop (counterclockwise for $\varphi = 90°$). When the polarization of the microwave is changed by 90° (Fig. 6b, $\varphi = 0°$), analogous saturation states are observed when the sign of H_0 is changed (in accordance with Fig. 5): a transition from one state to the other also follows a loop, but in the opposite direction (clockwise for $\varphi = 0°$).

Figure 7 shows the traditional hysteresis loop for a $DyFeO_3$ sample, obtained using the optical Faraday effect.

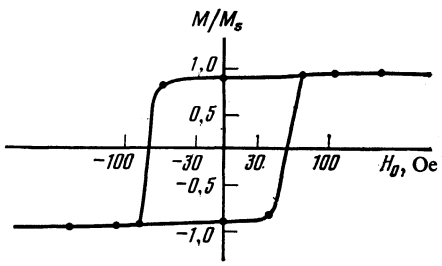

FIG. 7. Hysteresis loop plotted using the Faraday effect in light for $DyFeO_3$, $d = 100\,\mu$m.

The values of the fields in which sample saturation sets in (Fig. 7) coincide with the fields in which the domains vanish in the same sample when visually observed by magneto-optics methods. The fields in which the saturation of the resonant absorption sets in (Fig. 6) also correspond to fields in which the domains vanish in the same samples upon visual observation by the powder-pattern method. The cause of the small difference between the saturation fields (Figs. 6 and 7) is that the hysteresis loops were obtained by the Faraday effect for thin samples transparent to light, while the resonant-absorption loops were plotted for thicker samples (to obtain a larger resonant absorption and hence higher sensitivity). Regardless, however of the samples and of the methods, the observed hysteresis loops were practically symmetrical.

5. We were unfortunately unable to track the behavior of T_H/T following application of a much stronger magnetic field directly in the experimental apparatus. We therefore investigated samples that were magnetized beforehand by cooling from the Curie point in fields up to 11 kOe. This method causes practically complete vanishing of the domains that have unfavorable orientation, including the nuclei of former domains. In a $DyFeO_3$ sample magnetized beforehand in a positive field $H_0^+ = +11$ kOe, the residual resonant absorption at $\varphi = 0°$ differed little from the absorption in the demagnetized sample. When φ was varied the transmission of the sample increases gradually and reached maximum saturation at $\varphi = 90°$. After magnetization in a negative field $H_0^- = -11$ kOe the resonant absorption was preserved at $\varphi = 90°$ and vanished at $\varphi = 0°$ in accord with Figs. 6a and 6b. Application of a weak field (100–200 Oe) of opposite sign changed the transmission jumpwise from minimum to maximum or vice versa, depending on φ.

6. We investigated the effect of rotating the sample in a solenoid around an axis perpendicular to c, through an angle $\theta = 180°$ (so that the front and back faces of the sample changed place relative to the H_0^+ direction), as shown in Fig. 8a. Figure 9 shows plots of T_H/T against H_0 for $DyFeO_3$ at $\theta = 0°$ and $\theta = 180°$ (φ remains unchanged). At $\theta = 0°$ (curves 1 and 1′ for thicknesses $d_1 = 2$ mm and $d_2 = 1$ mm, respectively) the weakening and vanishing of the resonant absorption are effected by a negative magnetic field $H_0^- = -100$ Oe; H_0^+ alters the resonant absorption little, whereas at $\theta = -180°$ an analogous action is exerted by the magnetic field of opposite sign (curves 2 and 2′).

7. Curves 1 and 1′, 2 and 2′ in Fig. 9 pertain to two

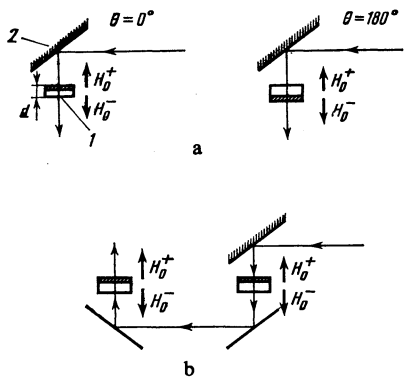

FIG. 8. Diagram of setup for the observation of resonant absorption a) for samples rotated 180° relative to the H_0^+ direction, b) for 180° change of the direction of the microwave propagation.

samples of different thickness, cut alongside the others from one and the same rod of 8 mm diameter and 30 mm length. A triangle scrateched on the lateral surface of the rod made it possible to "fix" the angles φ and θ of all the samples in the same positions as in the uncut material. A dependence of T_H/T on the sign of H_0 was observed in all samples. The resonant absorption vanished in all samples in fields of one and the same sign. At $\theta = 180°$ reversal of the sign of H_0 (in analogy with Fig. 9) was also observed in all the samples. Similar relations for crystals of other batches likewise differ little qualitatively from those given here. The value of T_H/T varies with thickness. Different magnetic fields are needed for resonant absorption to vanish in the different samples, but the variation range is small: from 100 to 150 Oe in $DyFeO_3$ and from 200 to 350 Oe in $TmFeO_3$.

8. A set of experiments was devoted to the influence of reversal of the wave vector, with the sample orientation with respect to φ and θ unchanged. This was realized in experiment by using a periscopic system of three mirrors (Fig. 8b). Parallel displacement of the solenoid together with the sample from one arm to the other was equivalent to reversing the propagation direction of the microwave. The behavior of T_H/T with changing magnitude and direction of H_0 was

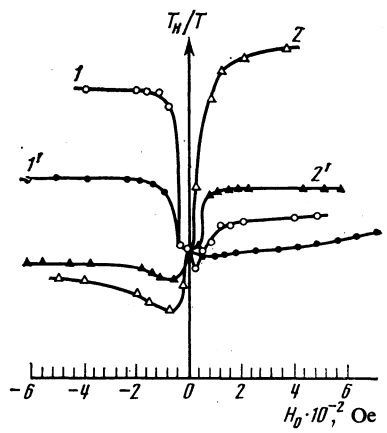

FIG. 9. Weakening of resonant absorption for $DyFeO_3$ vs H_0 at $\theta = 0°$ (curves 1 and 1') and at $\theta = 180°$ (curves 2 and 2'), $\lambda = 0.79$ mm, $d_1 = 2$ mm, $d_2 = 1$ mm.

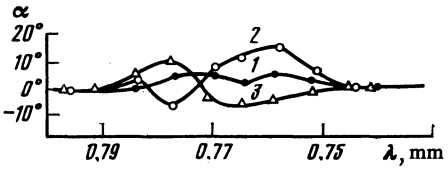

FIG. 10. Dependence of the angle of the Faraday rotation on the wavelength for $TmFeO_3$ (1—$H_0 = 0$, 2—$H_0^+ = +1000$ Oe, 3—$H_0^- = -1000$ Oe).

independent of the arm in which the sample was placed, i.e., of the microwave propagation direction.

9. Figure 10 shows the dependence of the angle α of the Faraday rotation of the polarization plane on λ for a $TmFeO_3$ sample 1 mm thick. With change of φ, the form of this dependence did not change substantially. Off resonance we have $\alpha = 0°$, in the resonance region a small rotation of the polarization plane is observed, and at certain λ the value of α reaches 10°, with α reversing sign and going through zero at the center of the resonance curve.

3. DISCUSSION OF EXPERIMENTAL RESULTS

We regard the most important facts to be the most important.

1. The weakening of the resonant absorption upon application of a magnetic field depends on the sign of the latter. At a certain polarization of the microwave, the vanishing of the domains is accompanied by vanishing of the resonant absorption when a positive magnetic field $H_0^+ \approx +100$ Oe is applied, whereas in a field of opposite sign the resonant absorption is preserved when the domains in the saturated samples vanish. The resonant absorption is preserved if the samples are magnetized beforehand by cooling from the Curie point in a negative magnetic field -11 kOe.

2. When the microwave polarization is changed by 90°, a similar action is exerted on the resonant absorption by a magnetic field of opposite sign.

3. When the sample is rotated through 180° (so that the front and back faces change places relative to the direction of the field H_0^+), the same action of H_0 as before is exerted on the resonant absorption when the sign of H_0 is reversed.

4. The character of the cited dependences is the same for samples of different thickness. These relations differ mainly in the value of $(T_H/T)_{max} = T_0/T$.

5. The behavior of the resonant absorption under the action of H_0 is not changed when the microwave propagation direction is reversed.

6. Resonant absorption is observed in magnetized samples when no domains or domain walls are revealed by investigation with powder patterns or with magneto-optic methods.

The saturating action of an external magnetic field on the intensity of the resonant absorption is thus unidirectional. This effect is not caused by any of the following:

1) Wave interference. This is evidenced by the reproducibility of the results for different electrodynamic systems, by the fact that the interference pattern is not changed by application of a magnetic field, and by the similarity of

the dependences of T_H/T on φ, H_0, and θ plotted for samples with different thicknesses.

2) Action of different electromagnetic waves and conversion of one wave into another. This is attested by the experimental results 3 and 5.

3) Faraday rotation of the polarization place, inasmuch as a) the propagation and reception of waves of different polarization are on a par in the experimental apparatus; b) the Faraday rotation angle is small and passes through zero at the center of the resonance curve.

The effect discussed cannot be attributed to asymmetry of the hysteresis loop. Numerous experiments on observation of symmetric hysteresis loops (Figs. 6a,b; Fig. 7) and on visual observation of the domain structures indicate that the domains vanish and the samples are magnetized in practically symmetric fields independently of the sign of H_0.

We propose that the experimentally observed unidirectional action of H_0 on the resonant absorption (independently of the nature of this absorption) is due to the fact that there exist in the crystal two directions, along the same axis, which are magnetically not equivalent, i.e., that a state similar to exchange (unilateral) anisotropy EA is realized in orthoferrites.

The effects observed do not contradict the explanation of the absorption as being due to resonance in the domain wall. The existence of resonant absorption in magnetized samples can be attributed to the fact that domain walls can still be preserved in this case, for example in the form of 360-degree walls. Such walls were predicted theoretically[6] and were observed experimentally[7] in iron-garnet films. Observation of 360-degree walls in orthoferrites meets with considerable difficulties because the thicknesses of the domain walls in orthoferrites are much smaller than in iron garnets and are beyond the resolving powers of presently existing methods. According to a calculation carried out in Ref. 8 using parameters from Refs. 9 and 10, the domain-wall thickness in orthoferrites is of the order of $10^{-2}\,\mu$m.

For resonant absorption in domain walls, the unidirectional character of the action of H_0 is a manifestation of the unidirectional action of H_0 on the change of the domain structure in the course of magnetization: one direction of the magnetic field leads to the vanishing (e.g., collapse) of the domain walls, while in the field in the opposite direction the domain walls do not vanish with the domains, but are preserved (e.g., in the form of 360-degree ones, which manifest themselves via resonant absorption). Since the EA depends on the directin of the antiferromagnetism vector,[11] different sections of the domain walls can have different signs of EA and accordingly different signs of the field H_0 that exerts identical action. In experiment one observes in fact a reversal of the sign of H_0 following a change of the polarization of the microwave, when resonance is excited in those domain-wall sections in which the rotation of the ferromagnetism and antiferromagnetism vectors on going through the wall takes place in another plane.

In conclusion, we thank A. S. Borovik-Romanov, Ya. A. Monosov, and the staff of the Magnetism Problems Laboratory of the Moscow State University headed by A. M. Kadomtseva for helpful discussions.

[1)]The intensity of the resonant absorption in the investigated samples is practically the same for different φ in the absence of H_0.

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