## SUBMILLIMETER INTRINSIC DOMAIN-WALL RESONANCE IN ORTHOFERRITES

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A hitherto unknown resonance absorption was observed in thulium and dysprosium orthoferrites in the submillimeter-wave range. The dependences of the resonance-absorption frequency and intensity on the external static magnetic field, the thickness of a sample, its temperature, and the polarization of the microwave field were investigated. This was done in order to determine the nature of the resonance process. A study was also made of the influence of the static magnetic field, the thickness of a sample, and its temperature on the domain structure and on the diffraction of light by domains. The experimental results were analyzed. It was found that the results could be explained on the assumption that the observed resonance was due to high-frequency oscillations of the domain walls.

## INTRODUCTION

T HE present investigation was carried out in order to study the transmission of orthoferrites in the submillimeter-wave range. The intention was to measure the transmission coefficient in a certain range of frequencies and temperatures. The need to carry out such investigations arose from the almost complete absence of the theoretical and experimental data on this subject.

We discovered<sup>[1]</sup> a resonance absorption at  $\lambda \sim 0.77$  mm in a sample of thulium orthoferrite<sup>1)</sup> at room temperature in the absence of a magnetic field. Thulium orthoferrite is one of the rare-earth compounds whose magnetic properties have been the subject of detailed experimental<sup>[2-4]</sup> and theoretical<sup>[5-7]</sup> investigations. The crystal lattice of these compounds has the space group  $D_{2h}^{16}$ . All such compounds exhibit weak ferromagnetism. At room temperature the spontaneous magnetization is always, with the exception of  $SmFeO_3$ , directed along the C axis. The domain structure in these compounds has been investigated by the magnetooptic method.<sup>[8,9]</sup> The domain boundaries are parallel to the C axis. These boundaries are shifted significantly by fields  $H_0 \sim 1$  Oe and the domain structure disappears in fields of 50-100 Oe.

Theoretical investigations of elementary excitations in a system exhibiting weak ferromagnetism predict that in the absence of a domain structure there should be two types of antiferromagnetic resonance at low and high frequencies. We shall show that the resonance absorption discovered by us has properties different from either of these two resonances. No theoretical treatments have yet been published on elementary excitations in the presence of a domain structure in a system exhibiting weak ferromagnetism.

There have been experimental observations of resonances in TmFeO<sub>3</sub> frequencies  $f = 57.7 \text{ GHz}^{[10]}$  and f = 1 MHz.<sup>[11]</sup> Hagedorn et al.<sup>[10]</sup> observed the resonance absorption in TmFeO<sub>3</sub> near the reorientation temperature (~ 81°K) at f = 57.7 GHz in magnetic fields H<sub>0</sub> = 6 kOe and H<sub>0</sub> = 8.5 kOe. Rossol<sup>[11]</sup> found low-frequency oscillations of domain boundaries in  $TmFeO_3$ , which were in the form of translational displacements.

The resonance described in the present paper differs from the phenomena reported in<sup>[10,11]</sup>. It is suggested that this resonance is due to the interaction of submillimeter radiation with high-frequency oscillations in domain boundaries which can occur in two-sublattice systems alongside translational displacements of domain boundaries. Resonance absorption of this type has not yet been observed experimentally in any of the three classes of magnetically ordered materials (ferromagnets, ferrimagnets, and antiferromagnets).

## EXPERIMENTAL RESULTS

1. We determined the transmission coefficient of thulium orthoferrite<sup>2)</sup> in the wavelength range 1.2-0.7 mm. The measurements were carried out using a quasioptical spectrometer.<sup>[13]</sup> A sample of TmFeO<sub>3</sub> was placed in a waveguide system matched to the quasioptical line by two horns. This was done in order to avoid diffraction losses in the quasioptical line because the dimensions of the sample were smaller than those of the quasioptical beam. The sample was a plate of  $5 \times 5 \times 1.2$  mm dimensions cut at right-angles to the C axis (the dimension along the C axis was 1.2 mm). The sample was placed in the horn-waveguide system in such a way that the electromagnetic beam was propagated along the C axis.

Our investigation of the dependences of the transmission coefficient of TmFeO<sub>3</sub> on the wavelength showed that a resonance absorption line was located at  $\lambda \sim 0.77$  mm. As shown in<sup>[1]</sup>, the resonance wavelength was practically independent of the static magnetic field H<sub>0</sub>. The influence of H<sub>0</sub> was observed only in the resonance absorption region ( $\lambda \sim 0.79-0.75$  mm) and this influence was manifested simply by weakening of the absorption. The transverse magnetic field had no influence on the resonance.

Figure 1 shows the temperature dependence of the resonance wavelength. This dependence enabled us to plot the resonance curves with the aid of an XY auto-

 $<sup>^{1)}</sup>$  Similar results were also obtained for dysprosium orthoferrite DyFeO<sub>3</sub>.

<sup>&</sup>lt;sup>2)</sup> A TmFeO<sub>3</sub> single crystal was grown by the floating zone method (see  $[1^2]$ ).



FIG. 1. Temperature dependence of the resonance wavelength for a  $TmFeO_3$  sample d = 1.2 mm thick.

FIG. 2. Resonance curves for TmFeO<sub>3</sub> samples of different thickness d: 1-1.2 mm;  $2-360\mu$ ;  $3-230\mu$ ;  $4-150\mu$ .



FIG. 3. Temperature dependences of the transmission coefficient T at the resonance frequency of a TmFeO<sub>3</sub> sample d = 1.2 mm thick:  $1-H_0 = 0$ ;  $2-H_0 = 50$  Oe.

matic recorder of the PDS-0.21M type keeping the wavelength constant (we used the x axis for the temperature and y axis for the change in the transmission through the sample). It was quite difficult to record the resonance curves as a function of the wavelength because the incident power was a function of the wavelength (the frequency characteristic of the oscillator power was not flat).

Figure 2 shows the temperature dependences of the transmission coefficients at  $\lambda = 0.9$  mm in the resonance region. These dependences are plotted for Tm FeO<sub>3</sub> samples of thickness d = 1200, 360, 230, and 150  $\mu$ . The dependences were deduced from the curves plotted by the X-Y recorder. The transmission outside the resonance line was assumed to be unity. It is evident from Fig. 2 that the resonance wavelength was independent of the thickness and that only the intensity of the absorption varied with the thickness (which was fully expected).

When the sample was heated above room temperature, the resonance wavelength remained constant (Fig. 1). However, the resonance intensity decreased with increasing temperature. Figure 3 shows the temperature dependences of the transmission of an TmFeO<sub>3</sub> sample 1.2 mm (1200  $\mu$ ) thick. The resonance disappeared practically completely at t ~ 130° C and the application of a longitudinal magnetic field had little effect on the value of the transmission coefficient. Thus, the resonance absorption began to disappear at temperatures considerably lower than the Curie point t<sub>C</sub> ~ 400° C.

The dependence of the resonance absorption on the orientation of a sample (i.e., the influence of the polarization of the incident radiation) was determined by recording the resonance curves at different temperatures. The results indicated that the resonance wavelength did not vary with the polarization.

2. The presence of a small net magnetic moment in antiferromagnets exhibiting weak ferromagnetism can be utilized in a study of domains by the same methods which are used for ferromagnets. A method particularly convenient in the case of antiferromagnets is that based on the Faraday effect. We investigated the domain structure of TmFeO<sub>3</sub> by this method. Our samples were plates of  $\sim 5$  mm radii, which were 230, 150, and 75  $\mu$  thick; they were cut at right-angles to the C axis in plane-parallel form and were polished. Figure 4 shows photographs of domain structures observed in crystals of different thickness: we found that the dimensions of the domains increased with the thickness.

The patterns resulting from the diffraction of light by domains in crystals of various thicknesses are shown in Fig. 5. The ring nature of the diffraction pattern corresponded to the labyrinth-like domain structure of orthoferrites. The radii of the rings decreased with increasing thickness because of the increase in the domain size, i.e., because of the increase in the grating period.

We investigated changes in the domain structure of  $Tm FeO_3$  above room temperature. It was known that the domain structure of orthoferrites varied with temperature. The dependence of the domain structure in a single crystal of thulium orthoferrite on the degree of cooling below room temperature was studied in<sup>[14]</sup> and it was found that cooling increased the number of domains but reduced their dimensions, as well as the thickness and energy of the domain walls. Heating reduced the number of domains (increased their dimensions) so that at t ~ 150°C the number of domains was approximately 2-3 times smaller than at room temperature.

We studied changes in the domain structure under the influence of a longitudinal magnetic field, parallel to the C axis, and during illumination of a sample with radiation of the resonance frequency. The magnetic field reduced the number of domains (increased their dimensions) and in fields  $H_0 \sim 100$  Oe the domain structure disappeared. Illumination of a sample with radiation of the resonance frequency did not blur the diffraction pattern or the domain structure.<sup>3)</sup>

## DISCUSSION OF RESULTS

The most important observations can be summarized as follows.

1. When the external static magnetic field, applied parallel to the C axis, is increased, the number of domains decreases (their dimensions increase) and the resonance absorption is weakened but the resonance frequency is unaffected. When the magnetic field is increased sufficiently ( $H_0 \sim 100$  Oe), the domain structure and the resonance absorption disappear simultaneously.

2. When a sample is heated above room temperature, the number of domains decreases (their dimensions increase) and the resonance absorption is reduced, but the frequency is not affected.

<sup>&</sup>lt;sup>3)</sup>This was true within the limits of resolution of our microscope  $(\sim 1\mu)$ .



FIG. 4. Photographs of domain structures in TmFeO<sub>3</sub> samples of different thicknesses d:  $a-230 \mu$ ;  $b-150 \mu$ ;  $c-75 \mu$ .

3. When the thickness of a sample is varied, the domain dimensions change but the resonance frequency is not affected.

The first of these observations shows that the observed resonance is not the conventional antiferromagnetic resonance and it is not identical with that observed experimentally in  $\text{Tm} \text{FeO}_3$  by Hagedorn et al.<sup>[10]</sup>

It also follows from our observations that the resonance absorption occurs only in the presence of a domain structure and that its characteristics are related to the parameters of the structure. We must reject the hypothesis that the cause of the resonance is a threedimensional diffraction grating formed by the domains because the resonance absorption frequency is independent of the domain dimensions (irrespective of the method used to vary these dimensions). Moreover, we cannot attribute the observed resonance to the interaction between the incident radiation and translational oscillations of the domain walls. This is supported by theoretical and experimental investigations [11] in which resonance absorption of this type occurs at low frequencies. We shall assume that the observed resonance absorption is due to the interaction of the submillimeter radiation with high-frequency oscillations in domain walls. These oscillations may coexist with low-frequency translational oscillations of the domain walls in orthoferrites.

FIG. 5. Diffraction of light by domains in TmFeO<sub>3</sub> samples of different thicknesses d:  $a-230 \mu$ ;  $b-150 \mu$ ;  $c-75 \mu$ .

The absence of a theory (because of the complexity of the problem) does not allow us to draw final conclusions. However, the suggested mechanism of resonance absorption in orthoferrites is supported by the analogy with compensated antiferromagnets. Thus, it is shown theoretically in<sup>[15]</sup> that for some antiferromagnets (for example,  $Cr_2O_3$  and  $MnF_2$ ) two types of motion of the

ferromagnetic M and the antiferromagnetic L vectors can give rise to three types of elementary excitations, one of which (the one with the smallest energy gap) is observed only in transition layers (antiferromagnetic resonance of domain walls). The low-frequency branch of the antiferromagnetic resonance of transition layers is in the form of translational oscillations of the domain walls. The high-frequency branch of the resonance of transition layers is due to the difference between the anisotropy constants of the sublattices. In the case of antiferromagnets such as NiO and MnO, for which the anisotropy constants of the two sublattices are the same, the low-frequency branch of the resonance of transition layers is absent.<sup>[16]</sup> The high-frequency antiferromagnetic resonance of domain walls has not yet been observed experimentally in compensated antiferromagnets. This is evidently due to the fact that in an antiferromagnet in which domains can exist at all they are observed only in a very narrow range of temperatures near the Néel point.[15]

We are of the opinion that in antiferromagnets with weak ferromagnetism the high-frequency oscillations are due to oscillations in transition layers of the type just described. The experimental data are not in conflict with this assumption.

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