Antiferromagnetic resonance in $TmFeO_3$

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The observation of two absorption lines in submillimeter (SBMM) spectra of single crystals of $TmFeO_3$ ($\nu_1 = 13\ \text{cm}^{-1}$, $\nu_2 = 23.9\ \text{cm}^{-1}$ at $T = 300$ K) is reported. These lines are interpreted as two branches of the antiferromagnetic resonance (AFMR), which are characteristic for orthorhombic antiferromagnets.

At room temperature thulium orthoferrite is an antiferromagnet ($T_N = 632$ K) with a weak ferromagnetic moment. The theory of AFMR, based on a two-sublattice model, predicts that two magnon modes exist in orthorhombic antiferromagnets.[1,2] Experimental investigations of the Raman[3] and neutron[4] scattering spectra and infrared[5,6] spectra of orthorhombic orthoferrites ($YFeO_3$, $DyFeO_3$, $HoFeO_3$,$ErFeO_3$) confirmed these predictions. It turned out that the AFMR frequencies in these crystals at room temperature lie in the range 8–25 cm^{-1}. For $TmFeO_3$ the AFMR lines have so far been observed only in neutron spectra,[4] and with low resolution ($\nu/\Delta\nu \sim 3$) their frequencies at $T \sim 300$ K were determined as $\nu_1 = 16\ \text{cm}^{-1}$ and $\nu_2 = 30\ \text{cm}^{-1}$.

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With respect to $TmFeO_3$, which we chose for the investigations, it should also be noted that in this material at frequencies characteristic for AFMR, namely, at $\nu = 13.0$ cm^{-1}, a group of authors observed and are studying intensively[7–10] a phenomenon which they interpret as a new type of resonant absorption in orthoferrites—natural AFMR in domain walls.

The purpose of this study was to observe and analyze AFMR in $TmFeO_3$ in infrared absorption spectra (by the method of SBMM spectroscopy) and, if possible, to clarify the nature of the absorption at $\nu = 13.0$ cm^{-1}, observed in Refs. 7–10.

We investigated a single crystal of $TmFeO_3$, grown by the method of floating-zone melting with radiation heating.[11] The specimens consisted of plane-parallel plates with characteristic transverse dimensions ~ 10 mm and thicknesses in the range 0.5–3 mm. The plates were cut out from a block in such a way that the weak ferromagnetic moment was situated in the plane of the plates.

Plates of $TmFeO_3$ were transilluminated in the "Epsilon"[12] backward-wavetube spectrometer along the normal to the surface by a beam of polarized monochromatic radiation. In the course of the experiment the radiation frequency was tuned continuously and in this manner we measured the transmission spectra $T(\nu)$ of the $TmFeO_3$ specimens at room temperature at frequencies ranging from 8 to 33 cm^{-1}. The spectra $T(\nu)$ were recorded for two orientations of the high-frequency magnetic field vector **h** relative to **M**—$\mathbf{h}\|\mathbf{M}$ and $\mathbf{h}\perp\mathbf{M}$. In addition, the measurements were performed both without an external magnetic field and in the presence of a field H of up to 3.6 kOe, applied to specimens with the orientation $\mathbf{H}\|\mathbf{M}$.

Typical results of these measurements are shown in Figs. 1 and 2. In complete correspondence to the conditions of excitation of AFMR in orthorhombic antiferromagnets, a single, sharp absorption line is observed in $TmFeO_3$ in each of the orientations $\mathbf{h}\perp\mathbf{M}$ and $\mathbf{h}\|\mathbf{M}$ at the frequencies $\nu_1 = 13.0$ cm^{-1} and $\nu_2 = 23.9$ cm^{-1}, respectively. When an external magnetic field $\mathbf{H}\|\mathbf{M}$ is applied, the frequencies of both modes increase. In this case, neither the shape nor the width of the lines changes appreciably.

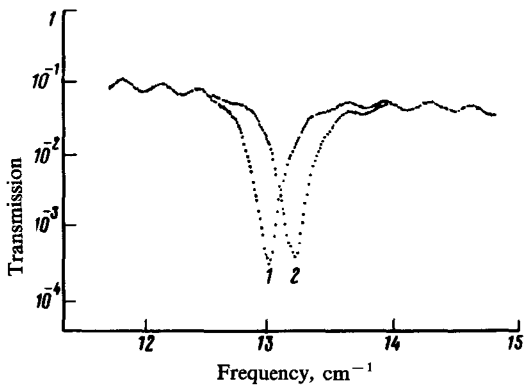

FIG. 1. Transmission spectrum $T(\nu)$ of a $TmFeO_3$ specimen with a thickness of $d = 2.703$ mm with $\mathbf{h}\perp\mathbf{M}$. The periodic oscillations in the dependence of $T(\nu)$ are a result of the interference of radiation within the specimen. The deep minimum represents the absorption line on the low-frequency branch of AFMR. 1) $H = 0$ and 2) $H = 3.6$ kOe.

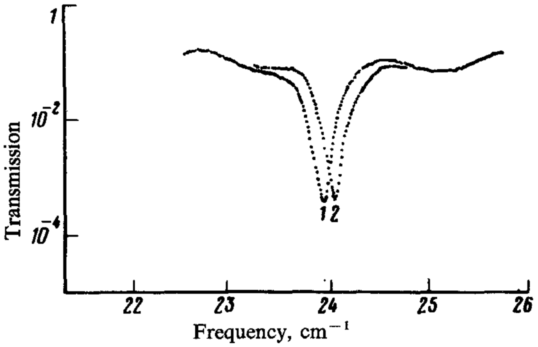

FIG. 2. Transmission spectrum $T(\nu)$ of the $TmFeO_3$ specimen with a thickness of $d = 0.883$ mm with $\mathbf{h} \| \mathbf{M}$ in the vicinity of the absorption line of the high-frequency branch of AFMR. 1) $H = 0$ and 2) $H = 3.6$ kOe.

From the well-known relation[1,2] we find

$$\nu_{1,2}^2(H) = \nu_{1,2}^2(0) + \gamma^2 H H_{1,2}$$

where γ is the gyromagnetic ratio. Under the assumption that the g factor is two ($\gamma = 0.093$ cm^{-1}/kOe), and using the measured values of ν_1 and ν_2, it is possible to calculate the effective fields in the crystal—$H_{1,2}$. For $TmFeO_3$, according to our data, $H_1 = 150 \pm 10$ kOe and $H_2 = 90 \pm 10$ kOe.

The low-frequency AFMR branch that we recorded at $\nu_1 = 13.0$ cm^{-1} coincided precisely in frequency with the resonant mode investigated in Refs. 7–10. Repeating the experiments in Refs. 7–10, and for this purpose it was necessary to measure the transmission of $TmFeO_3$ plates with a different orientation of the vector **M**—in a plane perpendicular to the plane of the plate, rather than parallel to it, we again observed a resonance at $\nu = 13.0$ cm^{-1} with all the indications of AFMR. In the same experiments it became clear that the given geometry—Faraday's geometry in which the wave propagates along **M**—is extremely unfavorable for observing AFMR. It happened that in this case the shape of the AFMR line depends on the domain structure of the specimen, its thickness, its dimensions, and the orientation of the external magnetic field. These phenomena, although present, were not taken into account in the experiments described in Refs. 7–10.

In summary, there is no doubt that the theoretically predicted AFMR occurs in $TmFeO_3$ at 13.0 cm^{-1}. However, the conclusions drawn in Refs. 7–10 concerning the observation of a fundamentally new phenomenon must be viewed as being incorrect.

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