Measurement of the magnetic spectrum of $YFeO_3$ by the method of submillimeter dielectric spectroscopy

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The spectrum of the complex magnetic permeability of antiferromagnetic material $YFeO_3$ is measured in the frequency range $\nu = 6$–$20\ cm^{-1}$ with a resolution of $0.001\ cm^{-1}$ in a zero external magnetic field at $T = 78$–600 K. The behavior of two-magnon modes, which at $T = 300$ K are situated at 10 and $17.6\ cm^{-1}$ and have a width of 0.1 and $0.05\ cm^{-1}$, respectively, is studied.

The submillimeter wavelength range ($\lambda \sim 3$–0.3 mm), which includes the resonance absorption lines of many antiferromagnets, is considered to be a difficult range in which to perform spectroscopic studies. This also pertains to IR absorption spectroscopy, although it is valid only for conventional methods of performing measurements, which are used in the microwave and IR regions. If, however, we take into account the rapidly improving technique of submillimeter (SBMM) spectroscopy, which is based on the use of tunable backward-wave monochromatic oscillators like the backward-wave tubes (BWT spectroscopy) as sources of radiation, then the situation, in our opinion, is reversed. In the methodological sense, the SBMM range is becoming the most convenient one for performing spectroscopic work. It turned out that here the basic advantages of the techniques of microwave and IR spectroscopy can be combined. On the one hand, the high "quality" of the working radiation—its high intensity (~ 10 mW), high monochromaticity ($\Delta\nu/\nu \sim 10^{-5}$), and high degree of polarization ($\sim 99.99\%$)—permits performing measurements with high resolution and high signal-to-noise ratio. On the other hand, as in IR spectroscopy, measurements are performed in free space with the help of simple measuring schemes. This makes it possible, first of all, to tune the frequency of the working radiation over a wide range and, as a result, to record the measured quantities in the form of spectra, rather than in the form of separate frequency points. Secondly, the simple distribution of the field in the working wave greatly simplifies the organization of the measurements and all subsequent calculations. Spectra can be recorded on a real time scale.

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The SBMM spectroscopy method has already been used to some extent to study the properties of magnets.[1-3] The Epsilon-type SBMM BWT spectrometers now have all of the indicated advantages.[4] These devices were initially designed for dielectric measurements on SBBM waves, i.e., for recording dielectric-permittivity spectra of materials.[5] Now, however, we can extend this experimental technique to the case of materials that exhibit magnetic properties ($\mu^* \neq 1$).

We investigated the antiferromagnetic crystal $YFeO_3$ in the temperature range 78–600 K and in the frequency range 6–20 cm^{-1}, using the Epsilon-2 BWT spectrometer. Plane-parallel *a*-section $YFeO_3$ plates were transilluminated by radiation with two orientations of the magnetic-field vector **h** relative to the magnetization vector of the specimen **M** (the cases **h**⊥**M** and **h**||**M**). The spectra of the energy coefficients of

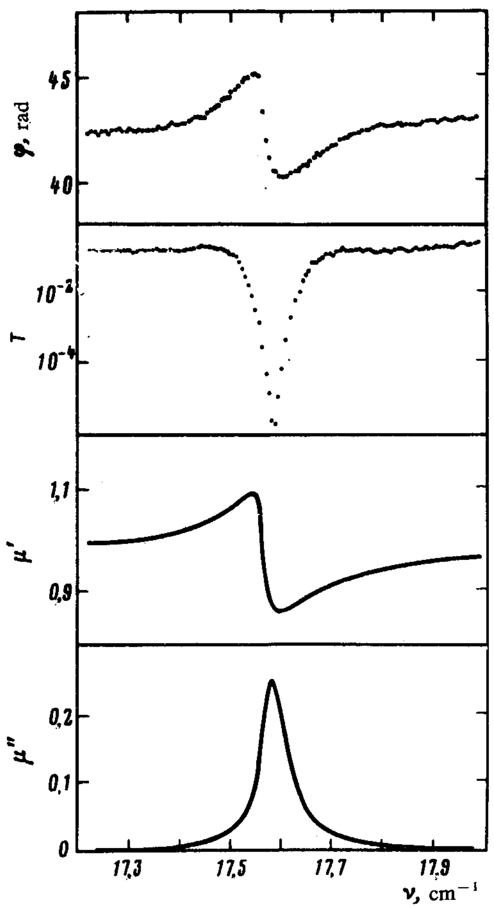

FIG. 1. a, b) Submillimeter spectra of the phase shift of the transmitted wave $\varphi(\nu)$ and of the energy transmission coefficient $T(\nu)$ of a 0.818-mm-thick $YFeO_3$ plate measured with **h**||**M** in the region of the high-frequency branch of the antiferromagnetic resonance. There is no external magnetic field and the temperature is the room temperature. c, d) Magnetic-permittivity spectra $\mu'(\nu)$ and $\mu''(\nu)$, calculated from the spectra $T(\nu)$ and $\varphi(\nu)$.

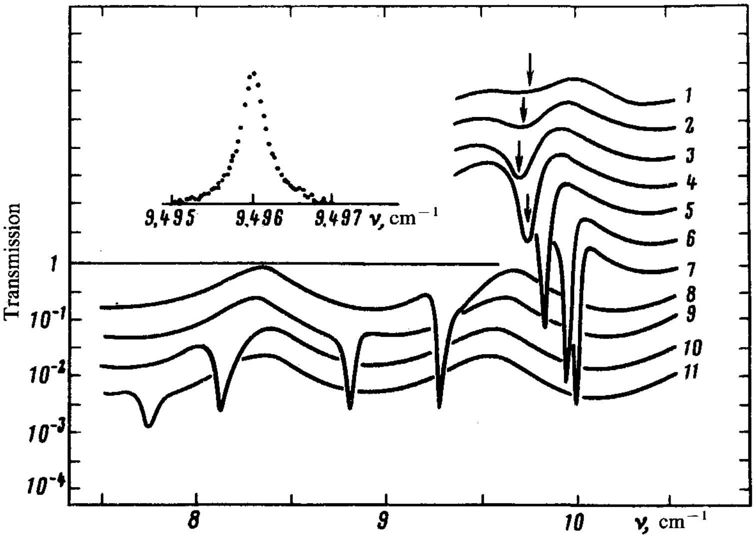

FIG. 2. Transmission spectra $T(\nu)$ of a 0.818-mm-thick $YFeO_3$ plate, measured at different temperatures (1—78, 2—111, 3—137, 4—175, 5—210, 6—256, 7—296, 8—437, 9—473, 10—506, 11—523K) in the orientation **h**⊥**M** in the region of the low-frequency branch of the antiferromagnetic resonance. There is no external magnetic field. The smooth oscillations on the curves are due to interference of waves within the specimen. The trace of the transmission of an open Fabry-Perot resonator, demonstrating the resolution of the spectrometer (line width ~0.001 cm^{-1}, transmission at maximum 2%), is shown in the upper lefthand corner.

transmission T and the corresponding phase shifts of waves φ were measured for several specimens of different thicknesses in the absence of an external magnetic field. The typical results of these measurements are shown in Figs. 1 (a and b) and 2.

Analysis of the experimental data obtained shows the following:

1) In both orientations (**h**⊥**M** and **h**||**M**), modes of the antiferromagnetic resonance are observed in the SBMM spectra of $YFeO_3$ (Figs. 1 and 2). In the first case, this corresponds to the well-known line at 10 cm^{-1} (at room temperature)[6] and in the second case it corresponds to the higher-frequency line (at 17 cm^{-1}), which has not been observed in IR spectra until now. This line, as well as the line at 10 cm^{-1}, can be seen in Raman light scattering spectra[7]; in this connection, it is interesting to note that the appearance of Raman scattering of active modes in the IR spectra of a crystal with an inversion center, which $YFeO_3$ is, occurs in spite of the rule of alternative forbiddenness, which is valid for electrical interactions. It can be shown that the observed modes have a magnetic nature.

2) The magnetic-permittivity spectra $\mu'(\nu)$ and $\mu''(\nu)$ can be calculated from the spectra $T(\nu)$ and $\varphi(\nu)$ by using the optical equations (Fig. 2). The value of the dielectric constant (in this case $\epsilon' = 22.6$, $\epsilon'' = 0.08$) required for these calculations is determined from these spectra $T(\nu)$ and $\varphi(\nu)$ outside the magnetic-resonance line. With appropriate programming of the spectrometer, the spectra $\mu'(\nu)$ and $\mu''(\nu)$ can be recorded on a real time scale. Until now, measurements of this type have not been performed, as far as we know, by optical methods.

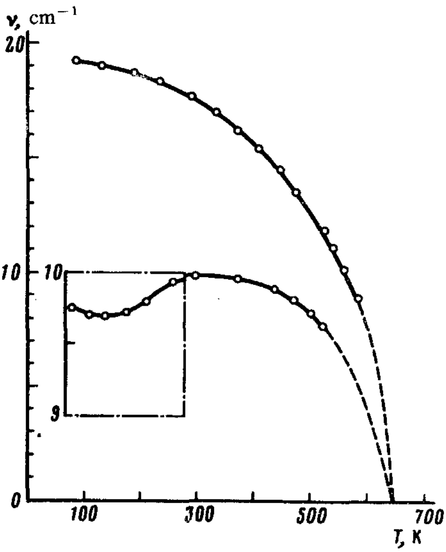

FIG. 3. Temperature dependences of the frequencies of two magnetic modes in $YFeO_3$ ($T_N = 648$ K).

3) An important aspect of this study is the high resolution of this method 0.001 cm^{-1}, whereas the preceding recording of the frequency spectra of magnetic materials in the SBMM wavelength range was limited to resolutions not exceeding 0.1 cm^{-1}.[6,7] This permitted us to observe, apparently for the first time at such high frequencies, the real contour of an antiferromagnetic resonance line and to follow its variation as a function of temperature (Fig. 2). We note, in particular, that the widths of the lines of the antiferromagnetic modes at 10 and 17 cm^{-1} at room temperature were, respectively, 0.1 and 0.05 cm^{-1}. When $YFeO_3$ is cooled to liquid-nitrogen temperature, the high-frequency mode contracts an additional severalfold. A very fine effect is observed in this case in the behavior of the low-frequency mode: nonmonotonic temperature dependences of its frequency, width, and intensity (marked by arrows in Fig. 2 and shown on an enlarged scale in Fig. 3). The entire effect unfolds in frequency within 0.3 cm^{-1}.

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