Unusual behavior of the magnetic resonance frequencies of $HoFeO_3$ in the spin-flip region

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Two antiferromagnetic resonance modes have been found in the spectra of $HoFeO_3$ measured over the frequency range $2 < \nu < 30$ cm^{-1}. The temperature dependence of the antiferromagnetic-resonance frequencies in the spin-flip region differs from that observed in other rare-earth orthoferrites. The difference is attributed to a strong interaction of the magnetic excitations in the iron and holmium subsystems in $HoFeO_3$.

Among the rare-earth orthoferrites, the $HoFeO_3$ crystals occupy a special position. According to optical data,[1–3] the energy of the splitting of the ground quasidoublet of the rare-earth ion falls directly in the region of antiferromagnetic-resonance frequencies in the Fe subsystem.

We have now studied the spectra of $HoFeO_3$ at submillimeter wavelengths, $2 < \nu < 30$ cm^{-1}, in the temperature interval 4.2–300 K. As a result, we have observed magnetic excitations of both $HoFeO_3$ subsystems, and we have found that their interaction has an important effect on the dynamics of the phase transitions $\Gamma_4 \rightarrow \Gamma_{42} \rightarrow \Gamma_2$, which result in a flip of the weak-ferromagnetic moment **m** from the c axis (the Γ_4 phase) to the a axis (Γ_2) through the Γ_{42} angular phase.

As in our previous studies of the magnetic spectra of orthoferrites, the measurements are taken on an Épsilon submillimeter backward-wave tube[4] with $HoFeO_3$ single crystals grown by float zoning with radiative heating.[5] The samples are a-cut parallel plates with transverse dimensions ~ 10 mm and a thickness ~ 1 mm.

In the transmission spectra of the $HoFeO_3$ we find two narrow ($\Delta\nu/\nu \sim 10^{-2}$) absorption lines and one broad ($\Delta\nu/\nu \sim 1$) line, which we interpret as antiferromagnetic-resonance modes $\nu_{1,2}^{Fe}$ and an ESR mode ν_R, respectively. As in other orthoferrites, the antiferromagnetic-resonance modes are observed over a broad temperature range: ν_1^{Fe} when the rf field is in the orientation $\mathbf{h} \perp \mathbf{m}$, and ν_2^{Fe} in the case $\mathbf{h} \| \mathbf{m}$.

The broad ν_R line is seen in the $HoFeO_3$ spectra only at low temperatures, $T < 40$ K (Fig. 1). From the spectral position, the excitation condition, and the temperature dependence of the intensity, we identify this excitation with an electronic transition between energy levels of the ground quasidoublet of the Ho^{3+} ion, which is split in the crystal and exchange fields. The frequency ν_R corresponds to a splitting of Ho^{3+} quasidoublet in the Γ_2 phase, found from the optical data[1–3] ($\Delta = 6$–7.5 cm^{-1}). The condition for its excitation ($\mathbf{h} \perp \mathbf{c}$) agrees with the selection rules for the magnetic moment of the Ho^{3+} ion,[2] and the increase in the intensity with decreasing tempera-

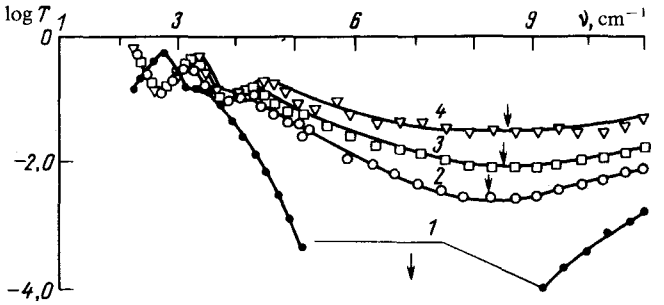

FIG. 1. Transmission spectra of an $HoFeO_3$ parallel plate with a thickness of 1.182 mm in the orientation **h**||**b** axis. 1—$T = 8$ K; 2—17 K; 3—21.5 K; 4—40 K.

ture corresponds to an increase in the population of the lower level of the quasidoublet of the Ho^{3+} ion.

We turn now to the temperature dependence of the antiferromagnetic resonance frequencies $\nu_{1,2}^{Fe}$ and the rare-earth mode ν_R in the region of orientational phase transitions T_{R1} and T_{R2} (Fig. 2). According to the model of Ref. 6, which incorporates only the dynamics of the Fe sublattice, the frequency of the mode ν_2^{Fe} remains essentially constant upon a spontaneous flip in the *ac* plane, while the softening of the quasiferromagnetic mode ν_1^{Fe} occurs both at the beginning (T_{R1}) and at the end (T_{R2}) of the flip process. A similar behavior of antiferromagnetic-resonance frequencies has been observed,[7] in particular, in $TmFeO_3$. In the case at hand, $HoFeO_3$, the picture is more complicated: As we approach the upper point of the spin flip, T_{R1}, the frequency ν_1^{Fe} does not exhibit any radical change, and its softening is observed only at

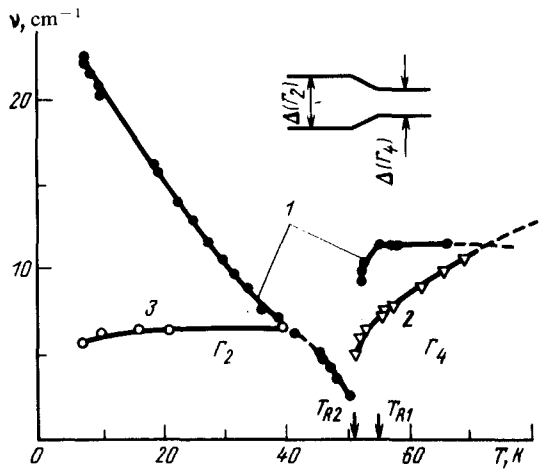

FIG. 2. Temperature dependence of the antiferromagnetic-resonance frequencies and of the frequency of the rare-earth mode in $HoFeO_3$. 1—ν_1^{Fe}; 2—ν_2^{Fe}; 3—ν_R. The inset shows the behavior of the low-lying energy levels of the Ho^{3+} ion due to a flip.

the lower boundary, T_{R2}. The frequency ν_2^{Fe}, on the other hand, falls off markedly near the transitions.

We believe that this unusual behavior of the frequency of the antiferromagnetic-resonance mode ν_1^{Fe} is due to an interaction of this mode with the low-lying rare-earth mode ν_R. In the Γ_4 phase, the frequency ν_R, determined by the splitting of the ground quasidoublet of the Ho^{3+} ion only in the crystal field, is 100 GHz, according to Ref. 3. In the Γ_2 phase, ν_R increases to 180–230 GHz because of an additional exchange splitting. In the spin-flip region, there is a repulsion of the frequencies of the antiferromagnetic-resonance mode and the rare-earth mode, with the result that ν_R softens at the phase-transition points T_{R1} and T_{R2}, while ν_1^{Fe} remains finite. This behavior of the ν_R and ν_1^{Fe} modes is in fact predicted by our calculations of the coupled vibrations of the magnetic moments of the Fe and Ho subsystems in the single-doublet approximation for Ho^{3+}.

The pronounced softening of the ν_2^{Fe} mode observed in $HoFeO_3$ in the vicinity of the phase transitions, with the result that the frequency ν_2^{Fe} becomes lower than ν_1^{Fe}, is typical of a Morin transition, in which the flip of the spins of the Fe^{3+} ions occurs in the *ab* plane; it is observed in $DyFeO_3$, for example.[8] This is also evidence of a tendency toward such a transition in $HoFeO_3$, i.e., a decrease in the anisotropy energy in the *ab* plane, as has been mentioned previously.[9] The softening of the ν_2^{Fe} mode of $HoFeO_3$ is apparently caused by the same factor as in $DyFeO_3$: a renormalization of the anisotropy energy of the Fe subsystem in the *ab* plane caused by an *R*-Fe exchange interaction (*R* is a rare-earth ion). A contribution to the anisotropy energy is made by the matrix elements of the Hamiltonian of this interaction between the ground quasidoublet and the excited states of the Ho^{3+} ion. An important point here is that the frequency of the corresponding transitions is substantially higher than ν_2^{Fe}, so that its softening is not disturbed, in contrast with the ν_1^{Fe} case.

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