Spin and lattice excitations of multiferroic $(Ba_{0.2}Sr_{0.8})_3Co_2Fe_{24}O_{41}$ in the THz range

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***Abstract*—THz, infrared and inelastic neutron scattering spectra of Z-hexaferrite ceramics were measured in a broad range of temperatures. A sharp electromagnon is observed near 1 THz, vanishing with increasing magnetic field until a phase transition occurs at $H \approx 2\,\mathrm{T}$. Then, another strong peak whose resonance frequency is linearly proportional to the magnetic field turns up from the sub-THz range. We assign this peak to a ferrimagnetic resonance.**

I. Introduction

THE materials with the Z-type hexaferrite structure have a general chemical formula $(Ba,Sr)_3M_2Fe_{24}O_{41}$ where M is a divalent metal atom (e.g. Co, Mg, Zn). They crystallize in the hexagonal $P6_3/mmc$ space group. These compounds belong to the rare examples of room-temperature magnetoelectric multiferroics [1]–[3] which are expected to have a great technological importance in future electronic applications. Remarkably, although the ferroelectricity is induced by spins, the Curie temperatures T_C are higher than room temperature, which is not the case for the vast majority of multiferroic compounds of this category. The advantages of Z-type hexaferrites include their high resistivity, a pronounced magnetoelectric effect, a low permittivity and a low loss tangent at room temperature. At the same time, they exhibit a transverse-conically ordered magnetic state up to ca. 400 K [4] and, most of them, a uniaxial magnetic anisotropy parallel to the c axis. Their particular feature is the absence of ferroelectric polarization at zero magnetic field. However, a polar state with space group $P6_3mmc$ can be magnetically induced via the inverse Dzyaloshinski-Moriya interaction [5]; it arises at very weak intensities of the magnetic field H and persists up to a few T.

The dynamical magnetic properties of Z-type hexaferrites are still largely unknown; they present a great interest both from the fundamental point of view and with respect to potential applications in memories, spintronics and magnonics.

II. Samples and Experimental

Our $(Ba_{0.2}Sr_{0.8})_3Co_2Fe_{24}O_{41}$ sample was synthesized as a powder which was compacted to a pellet with a diameter of 6 mm and polished down to a thickness of 0.48 mm. Infrared (IR) reflectance measurements were performed using a Bruker IFS-113v Fourier-transform IR spectrometer. THz complex transmittance was measured using a custom-made time-domain spectrometer including an Oxford Instruments Spectromag cryostat with a superconducting magnet, allowing to apply an external magnetic field of up to 7 T; the Faraday geometry (wavevector parallel to the magnetic field) was used. Inelastic neutron scattering (INS) was measured on a powder sample using a neutron time-of-flight spectrometer in ILL Grenoble, France. Further, we carried out measurements of the magnetic susceptibility, magnetization, Raman scattering, INS and magnetoelectric effect. The measurements were performed at temperatures from 5 to 900 K.

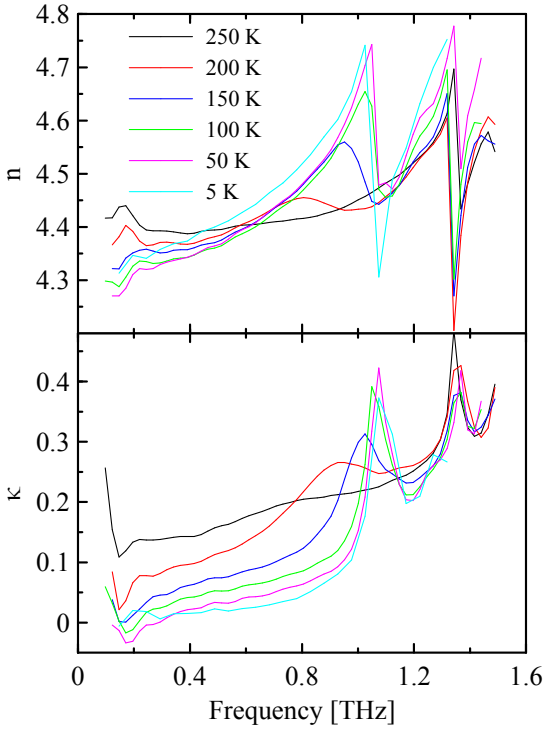

Fig. 1. Complex refractive index of the $(Ba_{0.2}Sr_{0.8})_3Co_2Fe_{24}O_{41}$ ceramics determined by THz spectroscopy as a function temperature. Above: index of refraction; below: extinction coefficient.

III. Results

The IR reflectivity spectra show 21 phonon modes—a half of the number allowed by the factor-group analysis. Upon cooling down to $T = 10\,\mathrm{K}$, the spectra change only slightly, confirming that there are no structural phase transitions.

In the THz range, at zero field, we observe a pronounced resonance near 1 THz (see Fig. 1). This peak is sharp at low temperatures and broadens upon heating; at room temperature, the excitation is overdamped but it is always present in the spectra. Upon applying magnetic field, the maximum in $\kappa(\omega)$

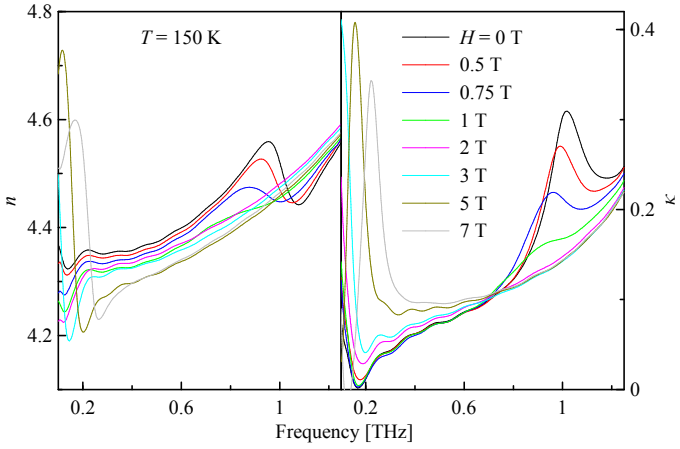

Fig. 2. THz spectra of the $(Ba_{0.2}Sr_{0.8})_3Co_2Fe_{24}O_{41}$ ceramics at a temperature of 150 K as a function of applied magnetic field. Left: refractive index; right: extinction coefficient.

shifts towards lower frequencies, it broadens and disappears near $H = 2\,T$ (see Fig. 2). Simultaneously, we observe a decrease in the extinction coefficient with H until a constant value is reached. This behavior apparently reflects a phase transition in the spin subsystem, which passes from the transverse conical to a collinear arrangement, similarly to other hexaferrite systems [6].

This magnetically active excitation was observed also in Raman spectra, which lets us assume that it corresponds to an electromagnon. This statement is based on the application of selection rules: Polar excitations (like phonons or electromagnons) are both IR- and Raman-active in non-centrosymmetric ferroelectric phases [7]. In order to confirm this hypothesis, we have performed INS experiments at a temperature of 5 K and at various energy resolutions. Fig. 3 shows a map representing the orientation-averaged scattering intensity at $T = 5$ K. Near the value of the neutron momentum transfer $Q = 1.3\,\text{Å}^{-1}$, we observe a magnetically active branch which extends in energy up to at least 20 meV. The magnon density of states (scattering intensity) does not exhibit any maximum at the energy of ca. 4 meV where the mode is observed in THz spectra. This means that the wavevector of the electromagnon is not at the Brillouin zone boundary; such magnons are usually activated by the exchange striction [8]. Instead, the observed electromagnon has a $\vec{k}$ vector in a general point of the Brillouin zone, and it is probably activated by the inverse Dzyaloshinskii-Moriya interaction. Note that, previously, an electromagnon was also detected in a hexaferrite with the Y-type structure [9].

As the magnetic field is further increased, another narrow resonance appears in the low-frequency part of the spectrum. At $H = 7$ T, its absorption peaks near 0.22 THz, a value which is temperature-independent within 5–250 K. This is probably a ferrimagnetic resonance, whose frequency is linearly proportional to the magnetic field.

IV. Conclusion

We have performed an extensive study of the magnetic properties of $(Ba_{0.2}Sr_{0.8})_3Co_2Fe_{24}O_{41}$. Two spin-wave excitations with frequencies and shapes depending on external magnetic

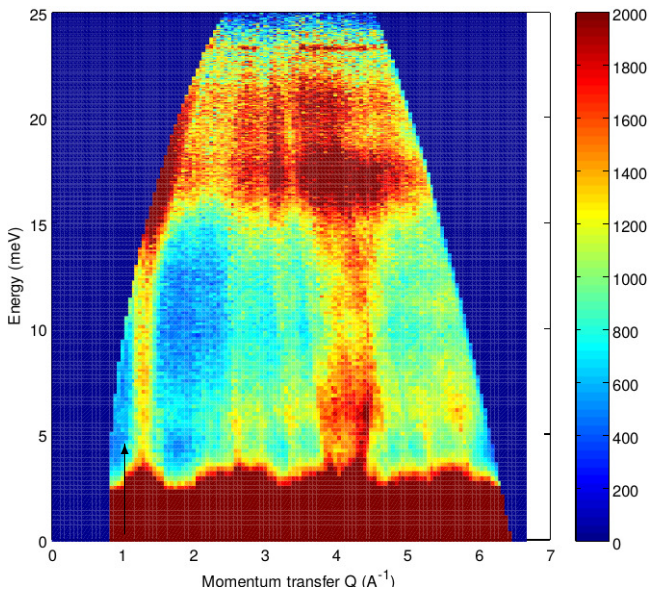

Fig. 3. INS map measured at 5 K. The arrow indicates the signal corresponding to the electromagnon branch.

field were observed in the THz range. The first one, active also in the Raman spectra and in INS, is assigned to an electromagnon due to the inverse Dzyaloshinskii-Moriya interaction. The other one, whose frequency is linearly proportional to the applied field, is interpreted as a ferrimagnetic resonance.

Acknowledgment

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