I. INTRODUCTION

Spin waves (magnons) in magnetically ordered materials can be excited by the magnetic component $\mathbf{H}(\omega)$ of the electromagnetic radiation, giving rise to a resonant dispersion of magnetic permeability in the microwave or terahertz (THz) frequency region. Recently, new coupled spin-lattice excitations named electromagnons have been discovered in multiferroics, where the magnetic order coexists with the ferroelectric one.\(^1,2\) Electromagnons are excited by the electric component $\mathbf{E}(\omega)$ of the electromagnetic radiation, therefore they can be detected in the THz dielectric permittivity spectra. Though they were theoretically predicted in 1970,\(^3\) they can be detected in the THz dielectric permittivity $\varepsilon_c$ within the same frequency range. This excitation contributes to the dielectric spectra in both antiferromagnetic and paramagnetic phases. Its oscillator strength significantly increases upon heating toward room temperature, thus providing evidence of piezomagnetic or higher-order couplings to polar phonons. Other heavily-damped dielectric excitations are detected near 100 cm\(^{-1}\) in the paramagnetic phase in both $\varepsilon_c$ and $\varepsilon_a$ spectra, and they exhibit similar temperature behavior. These excitations appearing in the frequency range of magnon branches well below polar phonons could remind electromagnons, however their temperature dependence is quite different. We have used density functional theory for calculating phonon dispersion branches in the whole Brillouin zone. A detailed analysis of these results and of previously published magnon dispersion branches brought us to the conclusion that the observed absorption bands stem from phonon-phonon and phonon-paramagnon differential absorption processes. The latter is enabled by strong short-range in-plane spin correlations in the paramagnetic phase.

DOI: 10.1103/PhysRevB.84.174120 PACS number(s): 75.85.-t, 78.30.—j, 63.20.D—, 75.30.Ds
magnetoelastic coupling. Nevertheless, electromagnons were recently observed also in BiFeO$_3$. In particular, the hexagonal YMnO$_3$ is ferroelectric below $\approx 1250$ K and the antiferromagnetic (AFM) ordering sets only below $T_N \approx 70$ K. The magnetic symmetry is $P_{6_3}cmm$ and therefore the linear magnetoelastic coupling is forbidden. However, piezomagnetic and magnetoelastic couplings and higher-order magnetoelastic couplings are allowed. The piezomagnetic coupling is characterized by a bilinear interaction between the magnetic order parameter and strain, in contrast to the magnetoelastic coupling which is proportional to the product of squared order parameter and strain. By using the method of optical second harmonic generation, the piezomagnetic coupling was observed owing to the interaction between AFM and FE domain walls in YMnO$_3$. Switching of the FE polarization triggers a reversal of the AFM order parameter. Higher order magnetoelastic coupling in YMnO$_3$ has been observed in several works. Exceptionally large atomic displacements at $T_N$ were observed in structural studies, and they demonstrate unusually strong magnetoelastic coupling. The large spin–polar-phonon coupling manifests itself by a decrease of the low-frequency permittivity near $T_H$, which is probably caused by anomalous hardening of several infrared-active phonons. Similar phonon anomalies were observed near $T_N$ also in the Raman spectra. Ultrasound measurements on a single crystal of the hexagonal YMnO$_3$ showed anomalous behavior of the elastic moduli $C_{11}$ and $C_{66}$ due to a strong coupling of the lattice with the in-plane exchange interaction. The AFM resonance in hexagonal YMnO$_3$ crystal was first reported in Ref. 33. More detailed THz studies of YMnO$_3$ ceramics were recently published in Ref. 34. The AFM resonance lies near 43 cm$^{-1}$ at 4 K, and its frequency softens upon heating toward $T_N$, where it disappears. Three magnon branches were discovered below $T_N$ using inelastic neutron scattering (INS). Two of them are degenerated near the BZ center, and their frequencies correspond to the above-mentioned AFM resonance. Moreover, a possible existence of magnons and short-range correlations between spins at Mn sites in paramagnetic phase were indicated by INS. The magnetoelastic coupling manifests itself also by a strong mixing of magnons with acoustic phonons; this leads to a gap in the transverse acoustic (TA) phonon branch occurring at the frequencies and wave vectors where the uncoupled magnon and TA branches would intersect. Recent polarized INS measurements revealed that the excitation detected at liquid helium temperatures near 43 cm$^{-1}$ has a mixed character of magnetic spin wave and lattice vibration, i.e., its contribution to both the magnetic permeability and the dielectric permittivity is possible. The reported piezomagnetic, magnetoelastic, and higher-order magnetoelastic couplings in optical, acoustic, and mainly INS data stimulated our spectroscopic study of hexagonal single crystals of YMnO$_3$. In this paper, we present results on far-infrared (FIR) and THz polarized spectra in this material emphasizing interaction between magnetic, electric, and phonon subsystems. We demonstrate that strongly underdamped AFM resonance observed near $\approx 40$ cm$^{-1}$ contributes only to the magnetic permeability spectra below $T_N$. An additional broad and weak absorption band was observed in the same frequency range in the dielectric spectra both below and above $T_N$. In contrast to electromagnons which are typically observed only below 50 K, the oscillator strength of this excitation significantly increases upon heating when room temperature is approached. This indicates that the feature must be related to the occupation number of magnons and/or phonons. An additional absorption band with similar temperature behavior was observed also near 100 cm$^{-1}$. We will show that both these excitations can be explained by differential multiphonon and magnon-phonon processes.

II. EXPERIMENTAL DETAILS

The experiments were performed using a Fourier-transform infrared (FTIR) spectrometer Bruker IFS 113v and a custom-made THz time-domain spectrometer. In both experiments, Optistat CF cryostats (Oxford Instruments) with polyethylene (FIR) or Mylar (THz) windows were used for measurements between 10 and 300 K. A helium-cooled bolometer operating at 1.6 K was used as a detector in the FTIR spectrometer. Principles of THz time-domain spectroscopy are explained in Ref. 42. The output of a femtosecond Ti:sapphire laser oscillator (Coherent, Mira) excites an interdigitated photoconducting switch TeraSED (Giga-Optics) to generate linearly polarized broadband THz probing pulses. A gated detection scheme based on an electro-optic sampling with a 1-mm-thick [110] ZnTe crystal permits to measure the profile of the electric field of the transmitted THz pulse (see Ref. 41 for further details).

Hexagonal YMnO$_3$ single crystals were grown by the floating zone method. Two crystal plates with lateral dimensions of $\approx 4.5 \times 5$ mm$^2$ and with the c axis oriented either in-plane or out-of-plane along its normal, were cut and polished to obtain highly plane-parallel samples (within $\pm 1 \mu$m) with thicknesses of 1100 and 348 $\mu$m for each orientation, respectively. These crystal plates were probed using the THz and FIR beam in all possible geometries: $E(\omega) \perp c$, $H(\omega) \perp c$, $E(\omega) \perp c$, $H(\omega)|c|$, and $E(\omega)||c|$. It enabled us to get access to the complex spectra of the products $\varepsilon_a\mu_a$, $\varepsilon_a\mu_c$, and $\varepsilon_c\mu_a$ as shown in Figs. 1(a), 1(b), and 1(c), respectively.

III. RESULTS

At low temperatures, the peak around 40 cm$^{-1}$ seen in the spectra of $\varepsilon_a\mu_a$ and $\varepsilon_c\mu_a$ [Figs. 1(a) and 1(c)] but not in those of $\varepsilon_a\mu_c$ [Fig. 1(b)] is definitely due to the AFM resonance as it contributes only to the magnetic permeability $\mu_a$. The AFM resonance vanishes above $T_N \approx 70$ K. The data shown in Fig. 1(b) allow us to assume that $\mu_c = 1$ in the THz range. This is in agreement with the magnetic order of YMnO$_3$ in the AFM phase: The spins are ordered in adjacent layers in the hexagonal plane in such a way that the magnetic resonances are not expected to be excited with $H|c|$. Based on this assumption, we are able to retrieve the complex values of the permeability $\mu_a$ and of the permittivity $\varepsilon_c$ (see Fig. 2).
FIG. 1. (Color online) Complex THz spectra of YMnO$_3$ taken at various temperatures. The polarization of the THz beam is indicated above the plots. The resonance feature near $\sim 40$ cm$^{-1}$ corresponds to the doubly-degenerated AFM mode contributing to the magnetic permeability $\mu_a$ spectra. The spectra of $\mu_a$ were fitted by a damped harmonic oscillator, and the resulting AFM resonance frequency is plotted in Fig. 3; a strong softening is observed upon heating toward $T_N$. Similar temperature dependence was briefly published earlier, with the magnon frequency higher by approximately 2 cm$^{-1}$. Besides the sharp AFM resonance line in the low-temperature $\mu_a$ spectra, one can observe a broad dielectric absorption band around 40 cm$^{-1}$ in the $\varepsilon_c$ spectra. This feature is detected even above $T_N$, where its strength remarkably increases with temperature. The presence of such a resonance in $\varepsilon_c$ is qualitatively expected from a simple comparison of the raw data in Figs. 1(a) and 1(c). The accessible spectral range of the THz measurements for our sample is limited to $\sim 60$ cm$^{-1}$, therefore we have performed also FTIR transmission (up to 100 cm$^{-1}$) and reflectivity (up to 650 cm$^{-1}$) measurements for all polarizations.

An example of FTIR experimental transmittance and reflectivity spectra obtained at 120 K and their various

FIG. 2. (Color online) Temperature dependence of the complex permittivity $\varepsilon_c$ and permeability $\mu_a$ spectra calculated from data plotted in Fig. 1. The solid $\varepsilon_c$ curves at 100 and 140 K result from the oscillator fit.

FIG. 3. (Color online) Temperature dependences of parameters of the resonances observed in magnetic $\mu_a$ and dielectric $\varepsilon_c$ spectra. Closed circles: frequency of the AFM resonance. Solid squares and open triangles: eigenfrequency $\omega_{\text{diele}}$ and oscillator strength $\Delta\varepsilon_0\omega_{\text{diele}}^2$, respectively, of the mode observed in the dielectric spectra in Fig. 2. The dotted line shows the population increase of an energy level at 66 cm$^{-1}$ following the Bose-Einstein statistics.
fits are shown in Fig. 4. Regular oscillations observed in the transmittance spectrum are due to Fabry-Pérot interferences in the plane-parallel sample; a weak minimum near 40 cm$^{-1}$ corresponds to the broad absorption band detected in the THz dielectric spectra (see Fig. 2). According to Ref. 30 as well as according to our FTIR reflectivity (see e.g. Fig. 4), the lowest frequency polar phonons lie above 150 cm$^{-1}$ in both polarized $E \parallel c$ and $E \perp c$ spectra. Nevertheless, our simultaneous fits of the THz complex permittivity and FTIR transmittance and reflectivity data reveal several additional modes below these phonon frequencies. The relevant spectra are plotted in Fig. 5. Besides the sharp magnon line at 40 cm$^{-1}$, three other broad modes at roughly 10, 40, and 100 cm$^{-1}$ were used in the fitting procedure in order to account for the measured shape of the $E \parallel c$ spectra at 10 K [see Fig. 5(a)]. The additional modes remain in the spectra up to room temperature and their strength increases upon heating. Also in $E \perp c$ polarized spectra, two broad modes observed near 10 and 90 cm$^{-1}$ were used for the fits above 50 K.

The feature observed near 10 cm$^{-1}$ in both polarized spectra could be related to low-frequency magnons (cf., the low-frequency magnon branches shown in Fig. 7). However, the sensitivity and accuracy of our THz spectra below 20 cm$^{-1}$ is limited; therefore we cannot exclude that it is only an artifact. For this reason, we will not speculate about the origin of this excitation. All other modes appearing below 150 cm$^{-1}$ are clearly observed in the THz and/or FTIR transmittance spectra, while the FTIR reflectivity measurements are not sensitive enough to detect and resolve these weak and broad spectral features (see Fig. 4). Their origin will be discussed in the next section.

The temperature dependence of the sub-THz complex dielectric permittivity $\varepsilon'_a$ plotted in Fig. 6 for 20 cm$^{-1}$ exhibits a pronounced drop below $T_N$. Such an anomaly is a typical feature of large spin-phonon coupling which occurs only in hexagonal planes of YMnO$_3$, where the spins are ordered. For that reason the anomaly is not observed in $\varepsilon'_c$ ($T$). The AFM phase transition is accompanied by unusually large atomic displacements, which were detected by neutron diffraction; for this reason the phonon frequencies change below $T_N$. The decrease in $\Delta \varepsilon_a$ and $\Delta \varepsilon'_a$ is mainly caused by hardening of the $E_1$ symmetry polar mode seen near 250 cm$^{-1}$ in the IR reflectivity spectra with polarization $E \perp c$. Fits of our IR reflectivity spectra show that the mode near 250 cm$^{-1}$ hardens from 246 cm$^{-1}$ (at 300 K) to 256 cm$^{-1}$ (at 10 K) and therefore its dielectric contribution $\Delta \varepsilon_a$ is reduced from 9.1 (300 K) to 7.6 (10 K). This decrease of $\Delta \varepsilon_a$ is mainly responsible for the
change of the permittivity $\varepsilon'_a(T)$ seen in Fig. 6. Hardening of other modes brings a minor contribution to the decrease of $\varepsilon'_a(T)$ upon cooling. Similar temperature dependence of $\varepsilon'_a$ was observed also in the radio-frequency region providing evidence of the absence of dielectric dispersion below 100 GHz. Gradual decrease of $\varepsilon'_a$ and $\varepsilon'_c$ upon cooling from 300 to 100 K is a usual behavior caused by a small phonon stiffening as a consequence of thermal contraction.

IV. DISCUSSION

The question arises about the origin of the absorption bands appearing below phonon resonances in Fig. 5. They are much weaker and significantly broader than those of polar phonons, and their strength increases when the temperature is increased, i.e., the strength is high in the paramagnetic phase. Their frequencies lying in the range of 40–100 cm$^{-1}$ coincide with those of the magnon branch observed by INS at 7 K over the BZ (see Fig. 7). In the following text we discuss whether these features can be related to the magnon dispersion branches.

Could a spin wave still exist in hexagonal YMnO$_3$ at room temperature? It is well established that Mn spins exhibit a strong short-range correlation in hexagonal YMnO$_3$ far above $T_N$. This was proved by an anomalous behavior of the thermal conductivity, elastic moduli, as well as by neutron scattering experiments. Nevertheless, due to the short-range correlation of the spins in the hexagonal plane of YMnO$_3$, one can expect the existence of only short-wavelength paramagnons, i.e., magnons with large wave vectors near the $M$ point of the BZ. A part of such a paramagnon branch is schematically plotted in Fig. 7. Note that its frequency is lower than that of the magnon branch at 7 K, as the magnon frequency decreases by almost 10 cm$^{-1}$ on heating toward $T_N$ (see Fig. 2).

Electromagnons are excitations with frequencies close to those of spin waves, which, due to specific couplings, are activated in the dielectric spectra. In perovskite manganites, the parts of magnon branches exhibiting a high density of states are mainly involved in these interactions (at BZ edge or close to the spin modulation wave vector). However, these electromagnons were observed only at very low temperatures (typically less than 50 K). Their strength dramatically decreases upon heating and they usually disappear from the spectra at $T_N$ or close above $T_N$. This is in contradiction with our observations in YMnO$_3$.

We came to the conclusion that the broad absorption bands we observe in the dielectric spectra reflect excitations which must be coupled to phonons. Let us discuss in brief which types of interaction between the magnetic subsystem and other degrees of freedom might be expected on the basis of the point group crystallographic symmetry $6mm$ and the magnetic symmetry $6mm$. The magnetic order parameter of

FIG. 6. (Color online) Temperature dependence of the (a) permittivity and (b) dielectric loss measured at 20 cm$^{-1}$ with polarization $E \perp c$ (red solid lines) and $E \parallel c$ (black dashed lines).

FIG. 7. (Color online) Dispersion branches of phonons (theoretical; black solid lines) and magnons (experimental at 7 K; red dashed lines). The red-dotted line indicates the presumable dispersion of the paramagnon near the $M$ point. The symbols shown at the BZ edges indicate the polarization of the phonons at the BZ boundary: $a$ and $c$ stand for phonons polarized within the hexagonal plane and in the perpendicular direction, respectively. In the $\Gamma$-point, the $E_1$ and $A_1$ phonons observed experimentally are marked by green and blue points, respectively; other modes are silent. Blue arrows with assignment $\omega_{\text{diel1}}$ and $\omega_{\text{diel2}}$ indicate phonon-paramagnon excitations observed in the dielectric loss spectra of $\varepsilon''_a$. Green arrow marked as $\omega_{\text{diel3}}$ indicates a broad multiphonon absorption observed in the $\varepsilon''_a$ loss spectra (see Fig. 5).
YMnO$_3$ was analyzed in several publications and it was shown to transform following $B_1$ ($\Gamma_3$) irreducible representation of the $6mm$ group. The $6mm$ symmetry strictly forbids the linear magnetoelectric effect, i.e., bilinear terms $a_{ij}H_iE_j$, where $H_i$ and $E_j$ are components of the magnetic and electric field, respectively, are not allowed in the thermodynamic potential. However, a higher order magnetoelectric effect (called sometimes the magnetodielectric effect), accounted for by the $\beta_{ijk}H_iH_jE_k$ terms in the thermodynamic potential, is allowed. This effect manifests itself in our measurements as a kink near $T_N$ in the temperature dependence of $\varepsilon''_p$ (see Fig. 6).

The magnetic symmetry of YMnO$_3$ allows the piezomagnetic contribution to the thermodynamic potential described by the terms $p_{ijk}H_i\sigma_{jk}$, where $\sigma_{jk}$ is a stress component and $p_{ijk}$ denotes components of the piezomagnetic tensor.

We believe that this type of bilinear coupling must play an important role in the interaction between the magnetic subsystem and the lattice. Usually, the piezomagnetic effect is allowed thanks to the relativistic part of spin-lattice and spin-spin interactions, provided the symmetry restrictions are met. However, in YMnO$_3$, which is a noncollinear antiferromagnet, the exchange (Coulomb) interactions may be by several orders of magnitude stronger than the relativistic ones and, therefore, they can be the origin of piezomagnetism. For example, extraordinary spin-phonon interactions were shown to contribute to the thermal conductivity of YMnO$_3$ below $T_N$. Higher order effects such as $p_{ijk}H_iH_j\sigma_{kl}$ are naturally also allowed in YMnO$_3$.

In order to provide a more quantitative explanation of the interaction between magnetic subsystem and phonons, we calculated the phonon spectrum from first principles within the spin-polarized local density approximation. We used projector augmented-wave potentials as implemented in Vienna Ab Initio Simulation Package (VASP). The following valence-electron configurations were considered: $4s^24p^65s^24d^1$ for Y, $3p^64s^22d^5$ for Mn, and $2s^22p^4$ for oxygen. To account for the strong electron correlation effects, exchange (Coulomb) interactions may be by several orders of magnitude stronger than the relativistic ones and, therefore, they can be the origin of piezomagnetism. For example, extraordinary spin-phonon interactions were shown to contribute to the thermal conductivity of YMnO$_3$ below $T_N$. Higher order effects such as $p_{ijk}H_iH_j\sigma_{kl}$ are naturally also allowed in YMnO$_3$.

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nature, which exist below polar phonon frequencies. The sharp AFM resonance band observed near 40 cm\(^{-1}\) at low temperatures broadens upon heating and disappears close to \(T_N\). This resonance is the main contributor to the magnetic permeability \(\mu_m\). Additional broad excitations were observed in the frequency range 40–100 cm\(^{-1}\) in the dielectric permittivity spectra in both the AFM and paramagnetic phases. Our theoretical explanation of the activation of these excitations in the THz dielectric spectra is based on two-particle differential processes schematically shown in Fig. 7. The resonance observed in \(\varepsilon_{\parallel}\) spectra is caused by differential phonon absorption in the A point of the BZ. The two broad absorption bands in \(\varepsilon_{\perp}\) spectra were described as differential phonon-paramagnon processes. The absorption strength of these excitations in the THz spectra increases upon heating due to the growing popular of magnons and phonons with temperature. This is possible in the paramagnetic phase owing to strong short-range spin correlations within hexagonal planes of YMnO\(_3\). The processes we observe in YMnO\(_3\), where the linear magnetoelcetric coupling is forbidden, are clearly different from the one responsible for the appearance of electromagnons in multiferroics with spin-induced ferroelectricity.\(^{1,2,45}\) The multiphonon absorptions are allowed by symmetry in all dielectric systems, while paramagnon-phonon absorptions can be expected only in paramagnetic systems with a strong short-range magnetic order (e.g., in hexagonal manganites). Magnon-phonon absorption should be also detectable in all magnetically ordered systems (FM, AFM, ferrimagnets, etc.) with relatively high critical temperatures. In such conditions, the magnons at the Brillouin zone edge may become sufficiently populated to allow multiparticle effects in the spectra. This may stimulate further THz and FIR studies of other magnetically polarizable systems.

ACKNOWLEDGMENTS

The authors thank M. Mostovoy for valuable discussions. This work was supported by the Czech Science Foundation (Project No. 202/09/0682), by AVOZ1010520, and by the Young Investigators Group Program of the Helmholtz Association (Contract VH-NG-409). The contribution of Ph.D. student V.G. has been supported by Project Nos. 202/09/H041 and SVV-2011-263303. R.V.P. acknowledges the support by the RFBR (Project No. 09-02-00070). The support of the Julich Supercomputing Center is gratefully acknowledged.
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