High-Frequency Electromagnon in GdMnO$_3$

A. M. Shuvaev,$^1$ F. Mayr,$^2$ A. Loidl,$^2$ A. A. Mukhin,$^3$ and A. Pimenov$^1$

$^1$Experimentelle Physik IV, Universität Würzburg, 97074 Würzburg, Germany
$^2$Experimentalphysik V, EKM, Universität Augsburg, 86135 Augsburg, Germany
$^3$General Physics Institute, Russian Acad. Sci., 119991 Moscow, Russia

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We present the results of transmittance experiments on GdMnO$_3$ multiferroic manganite in the far infrared frequency range. The spectra allow to obtain the positions and the intensity of the high frequency electromagnon (\textasciitilde 75 cm$^{-1}$) in this compound. We present the comparative analysis of the high and low frequency electromagnons across the phase diagram of GdMnO$_3$. The traces of the electromagnon excitation can be detected even at room temperature, i.e. deeply in the paramagnetic state.

Multiferroics are materials which simultaneously reveal magnetic and electric order [1,2]. Enormous recent theoretical and experimental interest to these systems is due to new interesting physics and possibilities for applications especially in memory devices. Multiferroics are often characterized by strong coupling between magnetism and electricity. This can lead to such effects like switching of the electric polarization in external magnetic fields or modulation of the magnetization by electric field.

Dynamic properties of the multiferroics are quite rich as well. In addition to classical modes of the ordered magnetic structure, multiferroics show the existence of new excitations of magnetolectric nature [3, 5]. These excitations have been called electromagnons and represent magnetic modes which can be excited by electric component of the radiation. A very instructive example is represented by the dynamics of TbMnO$_3$, because optical experiments in this compound [6, 8] can be combined with the results by inelastic neutron scattering [9, 10]. Closely similar systems, like GdMnO$_3$ or (Eu:Y)MnO$_3$ reveal in many aspects the same dynamic properties. The electromagnon dynamics in these composition seem to consist of two modes [3]. A strong high frequency electromagnon is observed in the frequency range 60-90 cm$^{-1}$. The frequency of this mode corresponds to a zone edge magnon as experimentally proved in inelastic neutron scattering experiments [10]. The spectral weight and excitation conditions for this mode can well be explained [8, 11] by the Heisenberg exchange mechanism combined with a modulation of a magnetic structure. In addition, another electromagnons can be observed in the frequency range of about 23 cm$^{-1}$. The origin of this mode, which is often split into two modes, is not fully understood. The characteristic positions of the low frequency electromagnon seems to correspond to a magnon at the center of the magnetic Brillouin zone [10, 12, 13]. In these scenario, the zone center magnons are electrically active modes of the cycloidal magnetic structure and receive electric dipole activity due to Dzyaloshinskii-Moriya mechanism [3,14]. As this mechanism cannot explain the intensity of the modes, the model including the magnetic anisotropy has been recently suggested [13, 16].

A comparison of the model predictions with the experimental parameters may help to resolve the problems with the origin of electromagnons. Therefore, investigations of dynamical properties of different multiferroics remain an actual task.

In this work we have carried out infrared transmittance experiments on multiferroic GdMnO$_3$. The combination of these data with the results by terahertz experiments [6] allowed to carry out a comparative analysis of low an high frequency electromagnons in this material.

Single crystals of GdMnO$_3$ have been prepared using the floating-zone method with radiation heating [17]. The samples have been characterized using X-ray, magnetic and dielectric measurement [18]. The basic properties of our samples agree well with the results obtained by other groups [19, 20]. Transmittance spectra in the infrared frequency range have been obtained using a Bruker IFS-113 Fourier-transform spectrometer. For this purpose the ab-oriented cut of the crystal was polished down to the thickness of 220 \textmu m. Reflectance spectra in the infrared frequency range have been measured on a thick sample from the same batch and were published elsewhere [21]. We note at this point that in spite of relatively large electromagnon intensity, these modes are hardly seen in reflectance spectra. Although the reflectance derived spectra [21] do show some characteristic feature around 75 cm$^{-1}$, without transmittance experiments the high frequency electromagnon could not be observed unambiguously. In present work, in order to obtain the full picture of the electromagnons, the results by infrared transmittance are combined with the terahertz spectra as obtained previously using the BWO-type technique [6].

Figure 1 shows transmittance spectra of a thin GdMnO$_3$ sample in the far infrared frequency range. The strongest absorption is observed close to 120 cm$^{-1}$, which corresponds to the lowest phonon in GdMnO$_3$. Close to this phonon the transmittance is below the sensitivity level of the spectrometer. Most importantly, a broad minimum in transmittance can be seen around 75 cm$^{-1}$ corresponding to the high frequency electromagnon. Already at this point we may state that with increasing
temperature the intensity of the electromagnon gradually decreases which corresponds to the increase of the transmission level. Even at room temperature, a weak local minimum in transmittance close to 75 cm$^{-1}$ can be seen indicating nonzero electromagnon intensity at this temperature. The presented data are sufficient to fill in the frequency gap between previous experiments [6, 21] in GdMnO$_3$, which gave reliable spectra above 100 cm$^{-1}$ and below 40 cm$^{-1}$.

The measured transmittance spectra have been transformed to the dielectric permittivity by inverting the Fresnel optical equations for transmittance and reflectivity which neglect the interferences within the sample. These interferences are seen as a Fabry-Pérot type modulation of the transmittance spectra in Fig. 1 and they are especially clear at room temperature and between 20 cm$^{-1}$ and 60 cm$^{-1}$. An attempt to take into account the interferences did not improve the quality of the solution probably due to imperfections of the sample surface. Figure 2 represent the far infrared spectra of the dielectric permittivity of GdMnO$_3$ in the frequency range relevant for electromagnons. The results by the infrared transmittance rapidly loose the accuracy below 40 cm$^{-1}$. Therefore, the data by BWO spectroscopy [3, 6] are plotted as closed symbols in this frequency range.

Two strong electromagnons can be well observed in the spectra of the dielectric permittivity close to 23 cm$^{-1}$ and 75 cm$^{-1}$. These modes are seen most clearly in the imaginary part of the dielectric permittivity (lower panel of Fig. 2). In the spectra of the $\varepsilon_1$ (upper panel) only the low frequency electromagnon can be detected. The reason of this effect is small dielectric contribution ($\Delta\varepsilon \sim 0.5$) of the electromagnons compared to the contributions of the phonons ($\Delta\varepsilon \sim 20$). In order to obtain the parameters of both electromagnons, the dielectric spectra in the far infrared frequency range were fitted using the sum of several Lorentzians. Two low frequency Lorentzians are responsible for the electromagnons and additional higher frequency modes represent the contribution of the phonons. In Fig. 2 the effect of only two lowest phonons is important.

The dielectric parameters of both electromagnons are shown in Fig. 3 as function of temperature. The res-
onance positions as represented in the left panel reveal only weak temperature dependence. As already mentioned, the high frequency electromagnon can be observed even at room temperature, i.e. far above the Néel phase transition. The eigenfrequency of this mode decreases with the increasing temperature in the magnetically ordered state and then slightly increases again towards room temperature. The position of the low frequency electromagnon remains roughly temperature independent. This mode cannot be detected in the spectra above 50 K. We note that the eigenfrequency especially of the high frequency electromagnon in Fig. 3 is slightly higher than the observed maximum in $\varepsilon_2$ (Fig. 2). This is due to the overdamped character of both electromagnons. Especially the width of the high frequency electromagnon is quite large $g = 125\pm5$ cm$^{-1}$ and, therefore, substantially shifts the position of the maximum in $\varepsilon_2$.

Contrary to the rough temperature independence of the eigenfrequencies, the spectral weight of the electromagnons in GdMnO$_3$ is strongly temperature dependent. On heating the sample into the paramagnetic state, the spectral weight of both electromagnons decreases by about a factor of three. However, the spectral weight of both electromagnons seem to remain finite even in the paramagnetic state, which could probably be related to the antiferromagnetic fluctuations.

At present, the nature of the high frequency electromagnon in orthorhombic manganites seem to be settled [8, 11]. The frequency of this mode corresponds to the zone edge magnon, the excitation conditions and the spectral weight are well explained on the basis of the Heisenberg exchange mechanism of the spin coupling. In order to provide an explanation for the low frequency mode, a magnetic anisotropy [12] and higher harmonics of the spin cycloid [10] within the Heisenberg exchange model have been suggested. The ratio of the frequency positions of both electromagnons $\nu_{0.1}/\nu_{0.2} = 3.7 \pm 0.5$ approximately agrees with the values in the (Gd:Tb)MnO$_3$ system [22] and can be roughly accounted for by the anisotropic model. On the contrary, the ratio of the spectral weights for GdMnO$_3$ can be estimated as 0.011 $\pm$ 0.002. Such small values agree with the tendency, observed in the (Gd:Tb)MnO$_3$ [22], but differ by about an order of magnitude from the theoretical estimates [13]. A possible reason for this discrepancy is the absence of the true cycloidal structure in GdMnO$_3$. In contrast to other orthorhombic manganite multiferroics like TbMnO$_3$ or DyMnO$_3$, most probably no cycloidal magnetic structure exists in the ordered state of GdMnO$_3$. Another possibility can be suggested in analogy with the discussion of the electromagnon mechanisms in TbMnO$_3$. In this case the low frequency electromagnon simply corresponds to the zone center magnon of the magnetic Brillouin zone [11, 12, 13].

In conclusion, using far infrared transmission spectroscopy, the high frequency part of the electromagnon spectrum in GdMnO$_3$ has been investigated. The spectral weight of the high frequency electromagnon is roughly two orders of magnitude higher than that of the low frequency mode. This ratio is the largest among other orthorhombic manganite multiferroics and cannot be explained within the existing models. The high frequency electromagnon can be seen in the spectra even at room temperature, i.e. deep in the paramagnetic state.

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