THE ROLE OF PHONONS AND MAGNONS IN FORMATION OF OPTICAL ABSORPTION FINE STRUCTURE IN MnO

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Manganese oxide MnO is a type-II easy-plane antiferromagnet which undergoes a paramagnetic to antiferromagnetic transition at the Néel temperature T_N = 118 K [1]. Optical absorption in MnO has been studied previously [2-8] and is interpreted using the energy level diagram for a free Mn^{2+} ($3d^5$) ion in a cubic crystal field. The optical absorption spectrum consists of six absorption bands due to spin and parity forbidden *d-d* transitions from $^6A_{1g}(S)$ ground state to $^4T_{1g}(G)$, $^4T_{2g}(G)$, $^4E_g(G)$+$^4A_{1g}$(G), $^4T_{2g}(D)$, $^4E_g(D)$ and $^4T_{1g}(D)$ excited states. In the present work we will discuss the fine structure of the $^6A_{1g}(S) \rightarrow {}^4T_{1g}(G)$ band, which appears at low temperatures (<60 K) and has not been observed in earlier works.

Single-crystal MnO(100) were grown epitaxially on freshly cutted and polished single-crystal MgO(100) substrates by chemical transport reactions method (the "sandwich" technique) using HCl gas [5]. Thus obtained MnO crystals have green colour and retain orientation of the MgO substrate. Optical absorption spectra were recorded using the split-beam Jasco spectrophotometer (Model V-570) with the tungsten iodine lamp used as a source. PbS photoconductive cell was used as a detector. A liquid helium cryostat was used to control the temperature of the samples down to 5 K with an accuracy ±1 K.

Temperature dependence of the low resolution spectra in the range of the first four absorption bands in MnO is shown in Fig. 1. The observed spectra are in agreement with previous data [2-8]. However, we would like to note some evidence of the fine structure, visible at 10 K, at low-energy side of the $^4T_{1g}$ band.

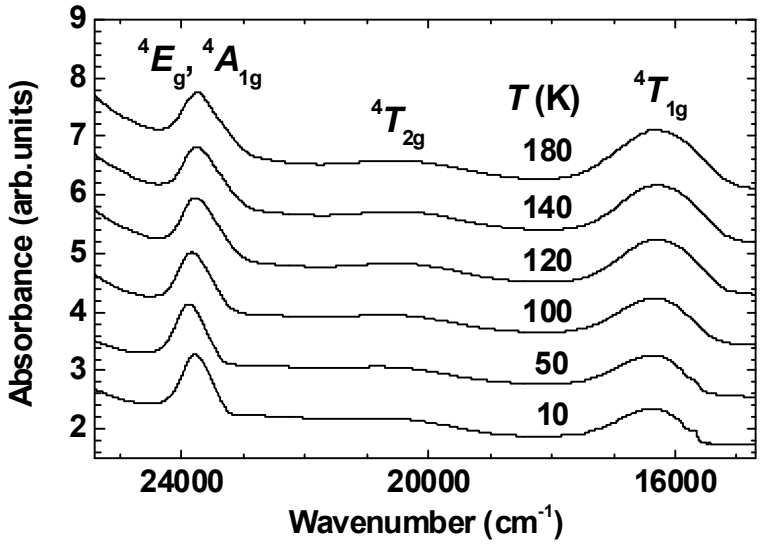

Fig. 1. Temperature dependence of the low resolution optical absorption spectra in single-crystal MnO(100). Three bands, at 16400 cm^{-1}, 20400 cm^{-1} and 23700 cm^{-1}, are visible.

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In Fig. 2 high-resolution optical absorption spectra in MnO are shown for the temperature range from 6 K to 60 K, that corresponds to antiferromagnetic state region. The fine structure consisting of three main peaks at 15680, 15950 and 16150 cm^{-1} is well visible at 6 K. Moreover, the first peak at 15680 cm^{-1} is splitted into two components, separated by about 34 cm^{-1} at 5 K (Fig. 3). The splitting decreases rapidly with temperature increase and becomes not detectable above 40 K (Fig. 2). Such behaviour allows us to suggest a magnetic origin of the observed splitting.

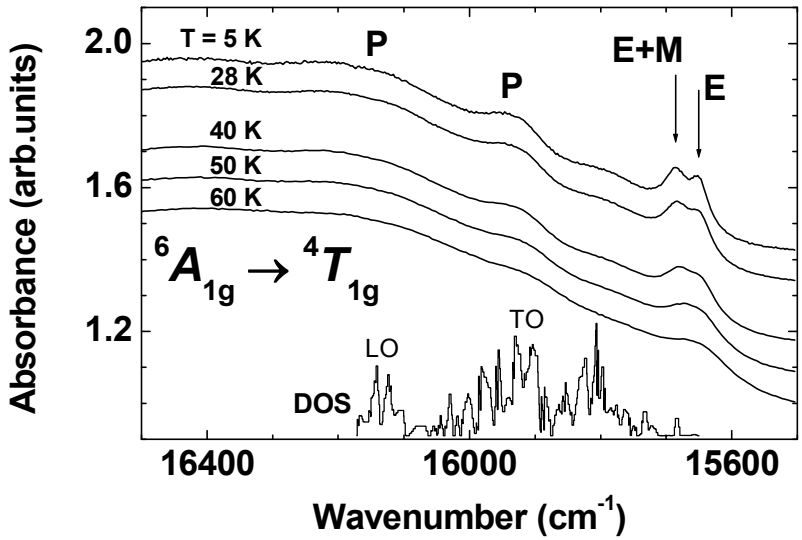

Fig. 2. Temperature dependence of the high-resolution optical absorption spectrum in the region of the $^6A_{1g} \rightarrow {}^4T_{1g}$ transition in single-crystal MnO(100). Theoretical MnO phonon density of states (DOS) (taken from [11]) is shown to indicate the phonon sidebands (P). Pure exciton (E) and exciton–one-magnon (E+M) transitions are marked by arrows.

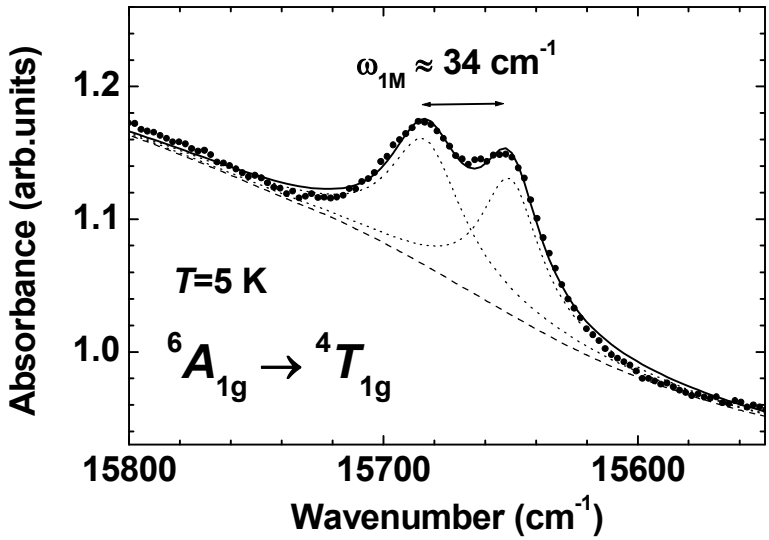

Fig. 3. Enlarged low-energy part (circles) of the high-resolution optical absorption spectrum in the region of the $^6A_{1g} \rightarrow {}^4T_{1g}$ transition in single-crystal MnO(100). Two peaks, due to pure exciton at 15650 cm^{-1} and exciton–one-magnon at 15684 cm^{-1} transitions, are well visible. Decomposition of the experimental signal into two Lorentzian lines (dotted curves) and background (dashed curve) is also shown.

Inelastic neutron scattering measurements of spin-wave dispersion curves in MnO at 4.2 K indicate that there are two magnon modes at 14.5±2.4 and 31.5±1.6 cm^{-1} [9] at the Brillouin zone-center. Note that the high-frequency mode was also detected at 27.5±0.3 cm^{-1} in far infrared measurements of the antiferromagnetic resonanse (AFMR) in MnO at 2 K [10]. Therefore, we attribute the two peaks at 15680 cm^{-1} in Figs. 2 and 3 to pure exciton transition (E) and exciton–one-magnon excitation (E+M). The separation between two peaks gives the one-magnon frequency of about 34 cm^{-1}: this value is in good agreement with high-frequency magnon mode found by neutron scattering [9].

The fine structure at higher energies is attributed to the phonon side-bands. In Fig. 2 we present the theoretically calculated phonon density of states (DOS) for MnO from [11]: it indicates the location of the TO and LO phonon modes exactly under the two observed peaks at 15950 and 16150 cm^{-1}. Moreover, recent high-resolution inelastic neutron scattering study [12] of the TO mode in the temperature interval from 4.3 to 300 K found the TO mode splitting by about 25 cm^{-1} below 100 K due to the magnetic-order induced phonon anisotropy. Therefore, this fact could explain large sensitivity of the peak at 15950 cm^{-1} to the temperature change.

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