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INTENSITY OF OPTICAL ABSORPTION OF Mn$^{2+}$ ION IN CUBIC ANTI-FERROMAGNET

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Résumé. — On explique l'intensité d'absorption optique des transitions $^6A_{1g} \rightarrow ^4T_{1g}$ et $^6A_{1g} \rightarrow ^4T_{2g}$ de Mn$^{2+}$ dans MnO et MnS et sa dépendance en température par les bandes associées à un magnon. L'approximation de paire est employée pour déterminer les états des spins de chaque paire d'ions.

Abstract. — The intensity and its temperature dependence of the optical absorption in MnO and MnS corresponding to the transitions $^6A_{1g} \rightarrow ^4T_{1g}$ and $^6A_{1g} \rightarrow ^4T_{2g}$ of Mn$^{2+}$ ion are explained in terms of sidebands associated with magnon. The pair approximation is used and the spin states of each ion pair are determined.

1. Introduction. — Optical absorption in MnO and MnS corresponding to transitions $^6A_{1g} \rightarrow ^4T_{1g}$ and $^6A_{1g} \rightarrow ^4T_{2g}$ of the Mn$^{2+}$ ions, measured by Huffman [1], shows an anomalously high intensity ($f \sim 10^{-3} \rightarrow 10^{-2}$) despite the parity- and spin-forbidden character of these transitions. The intensity increases with increasing temperature before reaching the Néel point but shows a tendency to saturate above the Néel point.

Provided that these transitions are the phonon-assisted transitions, the oscillator strength for non-assisted transition is expressed as $I(a \rightarrow b)$, where $I(a \rightarrow b)$ is the total spin of the ion $b$ and $I(a)$ is the spin of an electron of the ion $a$; $m$ and $m'$ represent spin and orbital states of an electron. $\epsilon_{q, m'}^{(1)}$ and $\epsilon_{q, m'}^{(2)}$ are creation and annihilation operators of an electron. Considering the Boltzmann distribution over the 36 spin states of the initial state, the intensity of the magnon sideband with respect to the pair is given by

$$I(a \rightarrow b) = \sum_{\pm} \exp \left[ - \frac{\epsilon_{q}^{(1)} T}{kT} \right] \times \sum_{m} \left| f_{m} \right|^{2} \left| f_{\pm m} \right|^{2} \exp \left[ - \frac{\epsilon_{q}^{(2)} T}{kT} \right],$$

where $\epsilon_{q}^{(1)}$, $\epsilon_{q}^{(2)}$, $f_{m}$, and $f_{\pm m}$ are spin substates of the initial and final states, and they are determined self-consistently by the pair approximation.

Since MnO and MnS have the type II antiferromagnetic spin arrangement, we take three kinds of ion pairs into account:

- pair 1: n.n.n ions with parallel spins,
- pair 2: n.n.n ions with antiparallel spins,
- pair 3: n.n.n ions with antiparallel spins.

The Hamiltonian of each pair can be written for pair 1

$$H_{1}(a, b) = - 2 J_{1} S_{a} S_{b} - B(S_{a} + S_{b})$$

for pair 2

$$H_{2}(a, b) = - 2 J_{2} S_{a} S_{b} - C(S_{a} + S_{b})$$

for pair 3

$$H_{3}(a, b) = - 2 J_{3} S_{a} S_{b} - D(S_{a} + S_{b})$$

where $J_{1}$ and $J_{2}$ are exchange constants for n.n. spins and n.n.n. spins, respectively, $B$, $C$, and $D$ are parameters representing the molecular fields arising from other spins except the pair spins under consideration. We introduce one-body Hamiltonian as

$$H(a) = - A S_{a}$$

where $A$ represents the molecular field and is related to $B$, $C$, and $D$ by $A = (B + C + D)/17$. We deter-

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mine the molecular fields, $B$, $C$ and $D$, as functions of temperature by the following equations:

$$\text{Tr} \left[ S_{\alpha \alpha} \rho(a) \right] = \text{Tr} \left[ S_{\alpha \alpha} \rho_j(a, b) \right] \quad (j = 1, 2, 3) \quad (6)$$

where $\rho(a)$ and $\rho_j(a, b)$ are the density matrices constructed from $H(a)$ and $H_j(a, b)$, respectively. In the paramagnetic region the molecular fields $B$, $C$ and $D$ vanish. Hence the spin states of the initial state are eigenstates of $-2J_1 S_x S_x$ for pairs 1 and 2 and those of $-2J_2 S_x S_y$ for pair 3. In the antiferromagnetic region the eigenstates of the pair Hamiltonians $H_1(a, b)$, $H_2(a, b)$ and $H_3(a, b)$ are solved self-consistently, together with eq. (6). Using these solutions we calculate the intensity by eq. (3) for each of three pairs. The temperature dependence of the intensity comes from the Boltzmann distribution over the 36 spin-states of the initial state and the variation of the molecular fields with temperature.

3. Results. — In figure 1 we show the calculated Néel temperature as a function of $\gamma (= J_2/J_1)$. For $\gamma < \gamma_c \approx 1.8$, the equation which determines the Néel temperature has no solution [4] and therefore the type II antiferromagnetic spin ordering is not stable in the pair approximation. For $\gamma > \gamma_c$ the upper and lower branches of the curve represent the Néel and anti-Néel temperatures. Thus, our pair approximation cannot be used to far below the Néel temperature. In our numerical calculations we fixed $\gamma = 3$ and we determined the exchange constant $J_1$ by making the calculated Néel temperature agree with the observed $T_N$. The thermal average of the spin was calculated and is shown in figure 2. It is compared with that calculated by the molecular field approximation. The intensities of magnon sidebands for pairs 1, 2 and 3 are shown by three solid curves and their average by a broken curve in figure 3. For pair 1, which has parallel spins, $P_{ab}$ at 0°K has no matrix elements between the ground spin state and the excited states, so that the intensity should decrease rapidly below the Néel temperature and tend to zero. For pairs 2 and 3, the intensity shows a gradual temperature dependence and remains finite at 0°K. The intensity at 0°K was calculated with the assumption that the molecular field for each pair arises from the perfect order of the spins. In figure 3, fine dotted curves interpolate to the values calculated at 0°K. Above the Néel point the intensity is almost temperature-independent.

References