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# DAVYDOV SPLITTING OF EXCITON LINE IN ANTIFERROMAGNETIC $RbMnF_3$


V.V. Eremenko and V.P. Novikov
Physico-technical Institute of Low Temperatures, Ukrainian Academy of Sciences


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$RbMnF_3$ is an ideal cubic two-sublattice antiferromagnet with $T_N = 82.5°K$ and with an ordering direction along a threefold axis. The unusually small anisotropy field, $H_A \simeq 4$ Oe, and the large exchange field $H_N = 8.9 \times 10^5$ Oe cause the magnetic structure of $RbMnF_3$ to be easily realigned by a weak magnetic field while remaining collinear, but requiring very strong fields for a noticeable disturbance of the collinearity. It was reported earlier [1] that application of an external magnetic field leads to a doublet splitting of the magnetic-dipole 25 144.5 $cm^{-1}$ line due to the optical transition $^6A_{1g} \rightarrow {}^4E_g(^4G)$ of the $Mn^{2+}$ ion. The effect is strongly anisotropic and is determined by the orientation of the magnetic moments of the sublattices relative to the crystallographic axis. It was shown in [2] that allowance for the magnetoelastic interaction makes it possible to explain the observed doublet splitting as being due to the lifting of double orbital degeneracy of the $^4E_g(^4G)$ state when the crystal lattice is distorted. The doublet character of the splitting is retained also in the region of strong magnetic fields applied along the directions [100] and [110]. We report here on the behavior of th 25 144.5 cm$^{-1}$ line in a strong magnetic field $\vec{H}$ || [111].

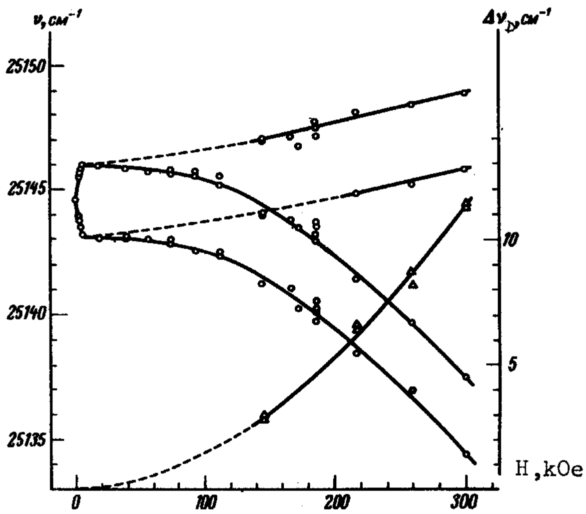


Fig. 1. Zeeman effect of exciton absorption line of the $^6A_{1g} \rightarrow {}^4E_g(^4G)$ transition of $RbMnF_3$ at 20°K in a field $\vec{H}$ || [111]. The triangles denote the field dedepdence of the additional splitting.

Figure 1 illustrates the field dependence of the observed splitting in unpolarized light at 20°K, as obtained with the aid of a pulsed solenoid. Figure 2 shows microphotometric curves of the considered region of the spectrum for certain characteristic values of the magnetic field. It is clearly seen that the doublet splitting, which appears even in a weak field, retains its value up to 300 kOe (as predicted when account is taken of the magnetoelastic splitting of the line), but is accompanied by additional doublet splitting of each of the components. The latter circumstance does not fit in the framework of the single-ion approximation, and calls for the use of exciton representations.

One characteristic feature of the spectrum of the electronic excited states of antiferromagnets is the Davydov splitting (DS) of the exciton lines, due to the interaction of the translationally-nonequivalent magnetic ions. The recently published first experimental data [3 - 6] have stimulated interest in this phenomenon, which has made it possible to assess the mechanism whereby excitation is transferred in ferromagnets.

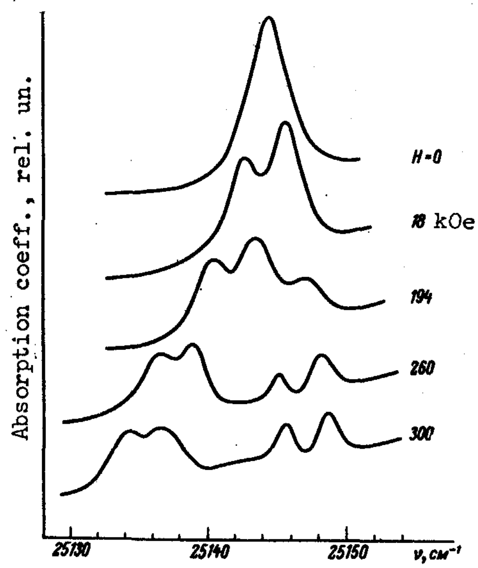

Fig. 2

As shown by Loudon, the DS of orbitally nondegenerate levels is possible if the space-group representation corresponding to the operator of the transition considered in the single-ion approximation (its symmetry is determined by the local magnetic group) is resolved into pairs of nondegenerate (in the case of two nonequivalent magnetic ions per unit cell) space-group irreducible representations determining the polarization of the Davydov components. The results of such an analysis[1] for $RbMnF_3$ and for different directions of the external magnetic field and the antiferromagnetism vector $\vec{l}$ [2] are summarized in the table, from which it follows that the DS is possible in all the listed cases. It is interesting that when $\vec{H} \parallel [111]$ any polarization of the splitting components is possible. Thus, the table does not contradict the statement that the additional splitting observed by us in DS. At the same time, since only translational spatial degeneracy remains after lifting the orbital degeneracy of the $^4E_g(^4G)$ level upon deformation of the lattice (two ions per unit cell), it follows that the interpretation of the additional splitting observed when $\vec{H} \parallel [111]$ as being DS is the only one possible.

The presently known observations of DS in optical spectra of antiferromagnets are limited only to $Cr_2O_3$ and $YCrO_3$ crystals [4 - 6]. In both cases, the exciton absorption lines pertaining to the so-called R group were investigated. It should be noted that the unit cells of $Cr_2O_3$ [6] and $YCrO_3$ [8] contain four non-equivalent ions each. This, on the one hand, greatly complicates the identification and makes it ambiguous. On the other hand, it leads to the need for considering two DS mechanisms that are connected with the

| Field direction | Ordering direct. | Local group | Trans. symmetry | Factor-group | Davydov comp. symmetry | Basis |
|---|---|---|---|---|---|---|
| $H \parallel [001]$ | $\vec{l} \parallel [110]$ | $C_{2h}(C_i)$ | $\Gamma_1^+$ | $D_{2h}(C_{2h})$ | $\Gamma_1^+$<br>$\Gamma_2^+$ | $S_z$<br>$S_x, S_y$ |
| $H \parallel [1\bar{1}0]$ | $\vec{l} \parallel [111]$ | $C_{2h}(C_i)$ | $\Gamma_1^+$ | $D_{2h}(C_{2h})$ | $\Gamma_1^+$<br>$\Gamma_2^+$ | $S_b$<br>$S_a, S_z$ (z axis $\parallel \vec{l}$) |
| | $\vec{l} \parallel [001]$ | $C_{2h}(C_i)$ | $\Gamma_1^+$ | $D_{2h}(C_{2h})$ | $\Gamma_1^+$<br>$\Gamma_2^+$ | $S_b$<br>$S_a, S_z$ (z axis $\parallel \vec{l}$) |
| $H \parallel [111]$ | $\vec{l} \parallel [1\bar{1}0]$ | $C_i$ | $\Gamma_1^+$ | $C_{2h}(C_i)$ | $2\Gamma_1^+$ | $S_x, S_y, S_z$ |
| | $\vec{l} \parallel [1\bar{1}2]$ | $C_i$ | $\Gamma_1^+$ | $C_i$ | $2\Gamma_1^+$ | $S_x, S_y, S_z$ |

[1] We confine ourselves to the point $k = 0$ of the Brillouin zone, which is the most important in exciton optical absorption. The analysis then reduces to a determination of the representations of the factor-group of the space group.

possibility of resonant transfer of excitation between the translationally nonequivalent ions which enter in either one or in different magnetic sublattices. As shown by Allen et al. [6], the results obtained with $Cr_2O_3$ show that the DS observed in it is due to exchange interaction of the nonequivalent $Cr^{3+}$ ions that enter in one sublattice. On the other hand, it is impossible to obtain unambiguous estimates of the resonant interaction between the sublattices from the available experimental data. The same pertains also to observations of DS in $YCrO_3$, since the identification given in [5] for the exciton spectrum is evidently not the only one.

We note in this connection that DS in $RbMnF_3$, which has two nonequivalent $Mn^{2+}$ ions per unit cell, is due exclusively to excitation transfer between the sublattices, and the data obtained by us yield direct information on this phenomenon for the case of optical excitation. It is interesting that, unlike $Cr_2O_3$ and $YCrO_3$, the DS in $RbMnF_3$ has no field-independent component, and is induced completely by the external magnetic field, increasing in proportion to the square of the field intensity:

$$\Delta\nu_D(\mathrm{cm}^{-1}) = 12{,}7\cdot 10^{-10}\cdot H^2\,(\mathrm{Oe}).$$

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## OBSERVATION OF A SINGLE NUCLEAR SPIN-SPIN RESERVOIR IN A CRYSTAL WITH TWO NUCLEAR SPIN TYPES

M.E. Zhabotinskii, A.E. Mefed, and M.I. Rodak
Institute of Radio Engineering and Electronics, USSR Academy of Sciences
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The question of the existence of a single reservoir of spin-spin interactions (the SS reservoir) in a crystal with several spin types is still unclear. Theoretical considerations were advanced [1 - 7], but the existence of such a reservoir has been proved experimentally only under special cross-relaxation conditions [3, 6] (when the resonant frequencies of the spins are practically equal or practically multiples of one another). Our experiments were undertaken for the purpose of clarifying this question. If a single SS reservoir does indeed exist, then saturation of one of the NMR lines on the wing should lead to a strong shift of the spin-spin temperature $T_{SS}$ [8 - 10] of all the spins, and this should result in a new effect, namely a characteristic distortion of other NMR lines that are not saturated directly.