Absorption and magneto-optical properties of the $^3A_{2g} \rightarrow {}^1T^a_{2g}$ transition in $CsNiF_3$. II. Pure-exciton and hot-magnon bands in a one-dimensional ferromagnet

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Absorption spectra associated with the lowest-energy region of the spin-forbidden transition $^3A_{2g} \rightarrow {}^1T^a_{2g}$ in $CsNiF_3$ are investigated in the one-dimensional ferromagnetic phase in magnetic fields up to 4.5 T. Pure-exciton bands are found to have a linear blue shift with a field applied in the easy plane, whereas the hot-magnon band shifts slightly to lower energies. When the field is increased the intensity of the exciton bands increases and the absorption due to the hot-magnon band decreases. An anomalous behavior of the band intensities and the half-widths is observed in fields below 1 T. The structure of the exciton spectra and their field dependence are discussed by using an energy-level scheme obtained by a single-ion approximation. The properties of the hot-magnon sideband are explained by the exciton creation accompanied by the annihilation of a thermally excited magnon at the Brillouin-zone edge. The anomalies of the exciton and the hot-magnon bands observed in low fields may be understood by the reduction of spin fluctuations in a magnetic field applied in the easy plane of magnetization.

I. INTRODUCTION

The properties of one-dimensional (1D) magnets are of current interest, especially as concerns the problem of magnetic solitons.[1] Although the existence of magnetic solitons has not been confirmed by optical methods, investigations of pure-exciton as well as phonon and magnon sidebands provide important information about spin dynamics in low-symmetry magnetic materials. Up to now various 1D magnetic insulators have been investigated by means of optical spectroscopy, e.g., 1D antiferromagnets tetramethyl ammonium trichloride (TMMC) (Refs. 2 and 3), $CsMnCl_3 \cdot 2H_2O$ (Refs. 2 and 4), and $CsMnBr_3 \cdot 2H_2O$ (Ref. 2), and 1D ferromagnets $A FeCl_3$ (A=K,Rb,Cs) (Ref. 5) and $CsNif_3$ (Refs. 6–10).

The spin-allowed $^3A_{2g} \rightarrow {}^3T_{2g}, {}^3T_{1g}$ transitions of Ni^{2+} ions in $CsNiF_3$ have been investigated extensively by using both optical-absorption and Raman scattering spectroscopy.[6–9] Recently we studied the absorption, magnetic circular dichroism, and the Faraday rotation spectra of the spin-forbidden transition $^3A_{2g} \rightarrow {}^1T^a_{2g}$ in this material and found a hot-magnon band in addition to pure-exciton transitions[10] (Ref. 10 is denoted paper I hereafter). The specific temperature dependence observed for the hot-magnon band was connected to the presence of 1D ferromagnetic order in the crystal. In the present paper a detailed level assignment is given for the pure-exciton and the hot-magnon bands. We also discuss the contribution of the exciton dispersion to the structure of the B and C bands in relation to the crystal-field splitting of the $^1T_{2g}$ level and give further evidence for the earlier interpretation of the A band as a hot-magnon band.[10]

In $CsNiF_3$, which has a hexagonal crystal structure, the Ni^{2+} ions are located in the center of fluorine octahedra (see Fig. 1), which are slightly distorted along the c axis so that the site symmetry of Ni^{2+} is D_{3d}. When the spin orientation is taken into account, the symmetry is further reduced to C_{2h}. Below $T_N = 2.61$ K $CsNiF_3$ is a 3D antiferromagnet. Above this temperature the exchange coupling between the Ni^{2+} ions is much stronger along the c axis than along the a and b axes, leading to the appearance of 1D Heisenberg ferromagnetism. The magnetic moments are bound to the a-b plane by the strong single-site anisotropy. Because the in-plane anisotropy is comparatively small, magnetic-field-induced effects can be expected in the behavior of the A, B, and C bands when the field is applied along the easy plane.[5] Cibert and Merle d'Aubigne[8] have observed a drastic reduction of the linewidth of the $^3A_{2g} \rightarrow {}^3T_{2g}$ exciton band in the field region of 0.3–1.2 T. In this paper we (1) investigate wheth-

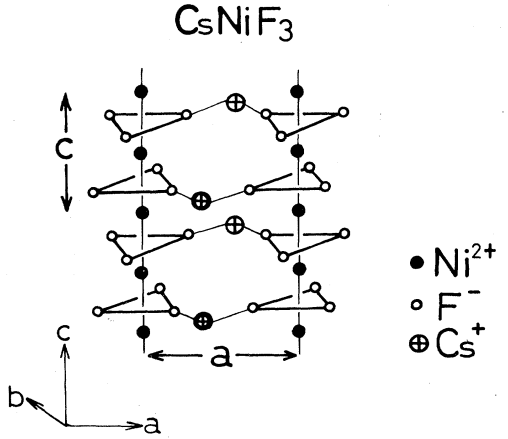

FIG. 1. Crystal structure of $CsNiF_3$.

er such a narrowing can be also seen in the hot-magnon band, (2) determine the magnetic field dependence of the peak position, intensity, and the half-width of the exciton lines of the $^3A_{2g} \rightarrow {}^1T_{2g}$ transition, and (3) compare the behavior of the A band with the properties of a hot-magnon band observed in a 2D ferromagnet.[12] The same experimental methods and the crystals were used as in paper I.

II. EXPERIMENTAL RESULTS

Figure 2 shows the π- ($\mathbf{k} \perp c$, $\mathbf{E} \| c$) and the σ- ($\mathbf{k} \perp c$, $\mathbf{E} \perp c$) polarized absorption spectra in the lowest-energy region of the $^3A_{2g} \rightarrow {}^1T^a_{2g}$ band at 7 K. All the A, B, and C bands appear in the σ polarization, but the A band does not appear in the π polarization. The intensity of the B and C bands is higher in the σ than in the π polarization. It is observed that the intensity ratio of the B band to the C band is different in the spectra corresponding to the two polarizations; the area of the B band is almost the same as that of the C band in the σ polarization, whereas the former is slightly smaller than the latter in the π polarization. A doublet structure (peaks C_1 and C_2) is observed under the C band although the components are not well resolved in zero magnetic field.

Figure 3 shows an unpolarized spectrum measured under an external magnetic field parallel to the a axis at 7 K. Resulting from the application of the magnetic field, it is observed that (1) the B and C bands are shifted towards the high-energy side, whereas the A band is slightly shifted to lower energies, (2) the intensity of the A band is decreased, as has been reported for the hot-magnon band in the 1D ferromagnet $CsFeCl_3$,[5] but the B and C bands are enhanced, and (3) the half-widths of all the A, B, and C bands are decreased. Due to the narrowing of the bands in the magnetic field the doublet structure of the C band can be resolved. The separation between the components C_1 and C_2 appears to be independent of the magnetic field and has the value of 8.3 cm^{-1}. As can be seen from the bottom of Fig. 3 the A band has an asymmetric line shape with a long tail on the low-energy side. In addition, a very weak absorption band which is called the X band was found on the high-energy side of the A band. It could be resolved in magnetic fields above 2 T and at temperatures below 10 K. The separation between the A and X bands is about 13 cm^{-1} at 4.4 T.

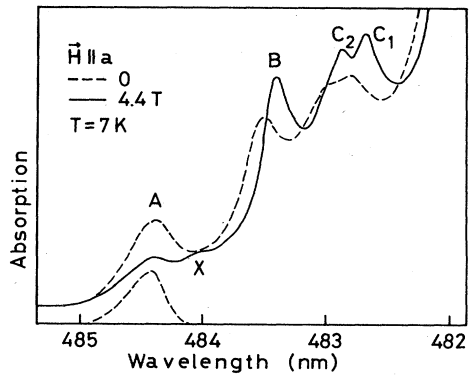

FIG. 3. Absorption spectra of the A, B, and C bands in a magnetic field along the easy plane (H=4.4 T, $\mathbf{H} \perp c$) at 7 K. The bottom curve is the A-band line shape obtained by subtracting the background when H=0.

In Fig. 4 the peak positions of the A and B bands are plotted against the magnetic field. Both bands are shifted linearly with the field. The slope of the shift is $+25.7\mu_B$ $cm^{-1}T^{-1}$ (μ_B is the Bohr magneton) for the B band and $-6.4\mu_B$ $cm^{-1}T^{-1}$ for the A band. The slopes for the B, C_1, and C_2 bands are found to be the same and agree well with the value observed previously for the G band.[10] Besides the positions of the hot-magnon band (A band) and the exciton bands (B or C), their intensities also behave in an opposite way when the magnetic field is applied in the easy plane: the intensity of the B and C bands is increased with the increasing magnetic field, whereas the intensity of the A band is decreased, as shown in Fig. 5. The B and C bands exhibit the same dependence on the magnetic field. A feature common to all the bands is that their intensity changes rapidly in the magnetic field region from 0.3 to 1 T. The intensity of the B and C bands is found to increase almost linearly with the magnetic field above 1 T, but as shown in Fig. 6 the intensity of the A band follows an exponential law in high magnetic fields. This law can be expressed as

$$I(H) = I(0)\exp(-1.2H/k_BT)\,, \tag{1}$$

where H is given in T and k_B is the Boltzmann constant.

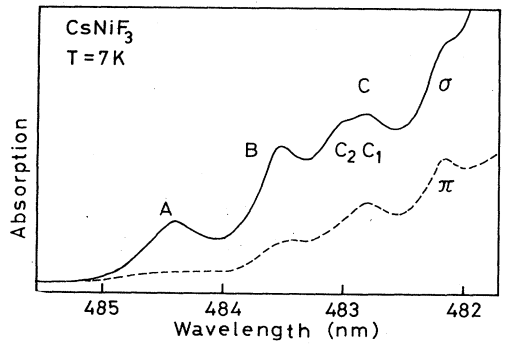

FIG. 2. Absorption spectra of $CsNiF_3$ observed on the low-energy tail of the $^3A_{2g} \rightarrow {}^1T^a_{2g}$ band with the light polarized with $\mathbf{E} \perp c$ ($\mathbf{H} \| c$, σ polarized) and $\mathbf{E} \| c$ ($\mathbf{H} \perp c$, π polarized) at 7 K.

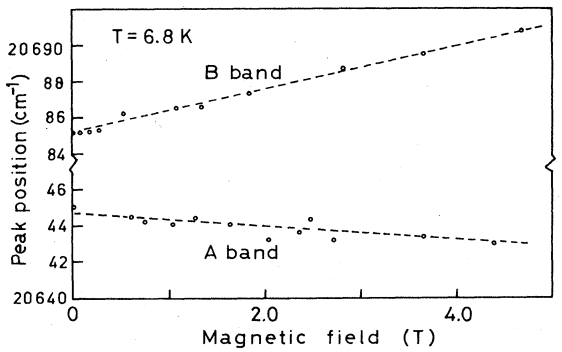

FIG. 4. Peak positions of the A and B bands plotted against the strength of the magnetic field H applied in the easy plane.

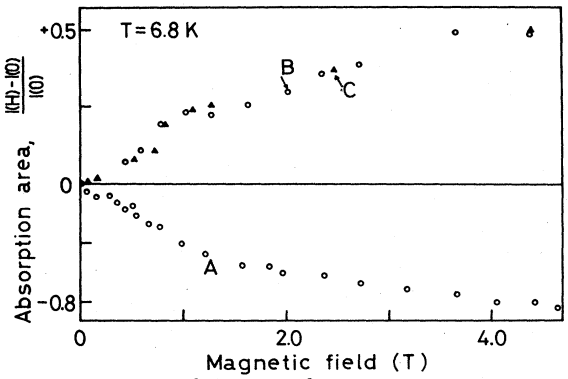

FIG. 5. Variation of the area of the A, B, and C bands under the magnetic field $\mathbf{H}\perp c$. The area is plotted for the difference from the area at $H=0$ divided by that observed at $H=0$.

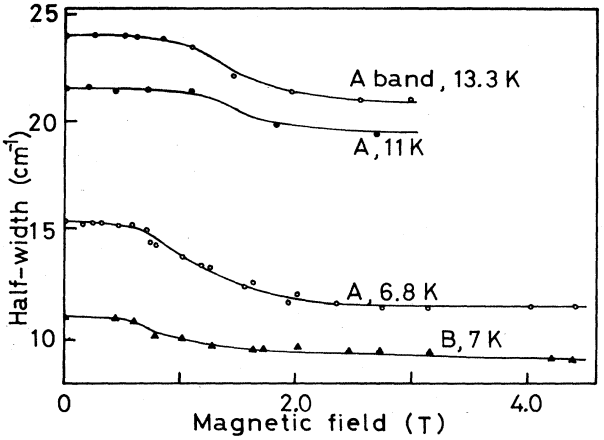

FIG. 7. Variation of the half-width of the A and B bands with the magnetic field applied in the easy plane.

Figure 7 shows the field dependence of the half-width of the A band at different temperatures and that of the B band at 7 K. A rapid narrowing of the bands is observed in the fields below 1 T. This is in accordance with the narrowing reported earlier for the pure-exciton line of the ${}^3A_{2g}\rightarrow{}^3T_{2g}$ transition.[8]

III. DISCUSSION

A. B and C bands

The dipole nature of an optical process giving rise to an absorption band can be determined from the anisotropy of the absorption, i.e., by measuring the σ-, π-, and α-polarized spectra.[13] The α spectrum ($\mathbf{k}||c$, $\mathbf{E}\perp c$) can be seen from Fig. 3(a) of paper I. The A band can be easily assigned to an electric dipole transition since it appears in the σ and α polarizations but not in the π polarization. This supports our earlier interpretation of the origin of the A band because the electric dipole nature is expected for a magnon sideband. The B and C bands are predominantly σ polarized although they appear weakly also in the π spectrum. The magnetic dipole character is expected for zero-phonon, zero-magnon (pure-exciton) lines associated with the spin- and parity-forbidden ${}^3A_{2g}\rightarrow{}^1T_{2g}$ transition of the Ni^{2+} ion with D_{3d} site symmetry, but on the basis of the experimental spectra we cannot exclude the possibility that both the magnetic and electric dipole transitions are responsible for the B and C bands.

Because there is no long-range magnetic order in a 1D ferromagnet above 0 K, the concept of exciton is properly defined only when the crystal is in an external magnetic field, which increases the spin-correlation length. To clarify the origin of the B and C bands, an energy-level scheme of isolated Ni^{2+} ions in $CsNiF_3$ was constructed by considering first the cubic part of the crystal field, then the spin-orbit interaction, the trigonal D_{3d} part of the crystal field, and finally the reduction of the space-group symmetry to C_{2h} when the spin is taken into account. The components arising from the ${}^3A_{2g}\rightarrow{}^1T_{2g}$ transition in this level scheme are shown in Fig. 8. The transitions from the A_g ground-state level into the three levels of the ${}^1T_{2g}$ state are magnetic dipole allowed. The transition to the A_g component of the excited state is σ-polarized, whereas the transition to the B_g component is both σ and π polarized. Taking into account that (1) all the B, C_1, and C_2 bands appear in the σ polarization, (2) the C_1 and C_2 bands are overlapped with each other, well separated

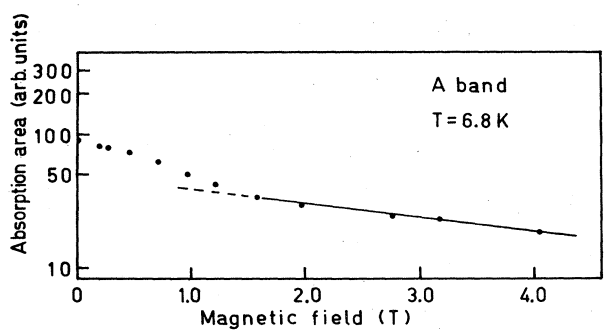

FIG. 6. Semilog plot of the A-band area against the magnetic field in the easy plane.

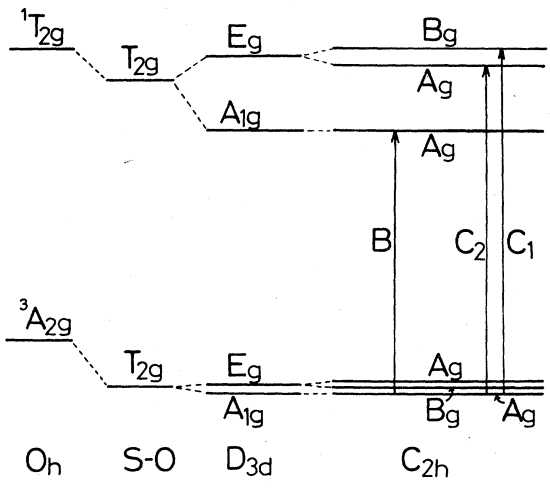

FIG. 8. Single-ion level scheme of Ni^{2+} under O_h crystal-field symmetry, spin-orbit interaction, and D_{3d} and C_{2h} symmetry. The arrows indicate magnetic dipole transitions.

from the B band, and (3) that the C_2 band is not very clearly observed in the π spectrum, we assign these bands as shown in Fig. 8. Hence the B band is associated with the $A_g(^1T_{2g})$ state and the C_1 and C_2 bands with the B_g and A_g states, which arise from the splitting of the doubly-degenerate $E_g(^1T_{2g})$ state in D_{3d} symmetry. The observation of the B band also in the π polarization, however, does not agree with the above assignment. This discrepancy can be explained by the closeness of the ground-state components $B_g(^3A_{2g})$ and $A_g(^3A_{2g})$ and by taking into account that the $B_g(^3A_{2g})\rightarrow A_g(^1T_{2g})$ transition involves the π polarization.

The A_g component of the ground state is believed to correspond to the $S^y=-1$ spin state in the exchange field,[7] where the spin is assumed to be oriented along the y axis in the easy plane. When the magnetic field is applied in the direction of the spin, the energy of the pure-exciton transitions from the $S^y=-1$ ground state into the $^1T_{2g}$ state should be shifted to higher energies by an amount $g\mu_B H$. This agrees with the experimental results which show that (1) the B, C_1, and C_2 bands exhibit the same blue shift in proportion to H, and (2) the g value estimated from the slope of the blue shift is 2.57, which is close to $g=2.4$ obtained from magnetic susceptibility measurements.[14] The above level assignment for the B and C bands is consistent with the magnetic circular dichroism (MCD) spectra reported for these bands in paper I. As can be expected for transitions form the $A_g(S^y=-1)$ ground state to a nonmagnetic excited state, the MCD line shapes of these bands are quite similar to their absorption line shapes.

According to Fig. 1 of paper I, the $^1T_{2g}^a$ band is partially overlapped by a strong $^3T_{2g}^b$ band which is spin allowed. Therefore the spin-orbit interaction between these states cannot be neglected. When the magnetic field is applied, the mixing of the two states is further enhanced by the Zeeman interaction of the spins and we can expect that the transition moment of the $^3A_{2g}\rightarrow{}^1T_{2g}^a$ exciton bands will grow in proportion to H^2. The observed enhancement of the B and C bands in magnetic fields below 1 T is consistent with this expectation (Fig. 5). However, it is observed that the band intensities deviate from the H^2 law above 1 T, where they increase almost linearly with the field. This behavior can be qualitatively understood as follows. The absorption intensity is proportional not only to the square of the transition moment but also to the population of the ground state. Taking into account that the ground-state energy decreases with the increasing magnetic field by the Zeeman effect, a monotonic increase, approximately proportional to H, can be expected in the absorption strength.[15] Therefore it seems likely that the transition-moment part would be predominantly responsible for the rapid enhancement of the B and C bands in the low-field region, whereas the ground-state—population part becomes more important in the high-field region. The anomalous increase of the intensity of the B and C bands occurs in the same region of magnetic fields where the half-widths of the bands and the intensity of the hot-magnon band decrease rapidly. This suggests that the reduction of spin fluctuations by the magnetic field contributes to the increase of the intensity of the B and C bands.

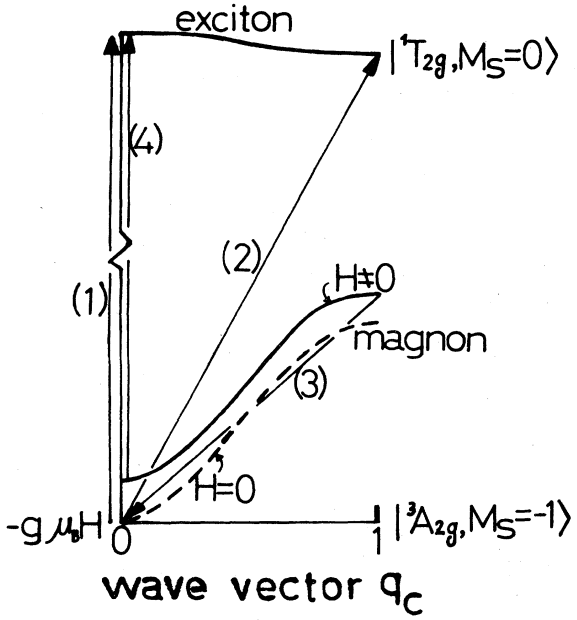

FIG. 9. Schematic dispersion curves of excitons and magnons in $CsNiF_3$ along the reduced wave vector q_c. The energy of the ground state $^3A_{2g}(M_s=-1)$ is taken to be $-g\mu_B H$. The explanation of the arrows from (1) to (4) is given in the text.

B. Hot-magnon band

Various processes relevant to the absorption of light in a ferromagnet are shown in Fig. 9, where the magnon dispersion, obtained from neutron scattering measurements,[11] is included. The hot-magnon band arises from an exciton-magnon coupling which induces the creation of an exciton [process (2) of Fig. 9] accompanied by an annihilation of a thermally excited magnon [process (3)]. Hence, the hot-magnon band is due to processes (2) + (3), whereas the pure-exciton absorption corresponds to process (1). In the exciton-magnon coupling the Brillouin-zone-edge magnon ($q_c=1$ in Fig. 9) is known to be responsible for the peak position of the absorption band,[16] as marked by arrow (3). Thus the peak energy of the hot-magnon band in a magnetic field is given by

$$E=W^e(q_c=-1)-W^m(q_c=+1)+g\mu_B H-\Delta W^m(H)\ , \tag{2}$$

where W^e and W^m are the exciton and magnon energies at the zone edge[17] at $H=0$, and $\Delta W^m(H)$ means the magnon energy shift at the zone edge when the magnetic field is applied. According to the neutron scattering measurement[11] the energy shift is linear in the field and independent of the reduced wave vector q_c, except near the zone center. The actual values are $\Delta W^m=5.65\ \text{cm}^{-1}$ (0.70 meV) at $H=4.1$ T and $q_c=0.35$ and $\Delta W^m=8\ \text{cm}^{-1}$ at $H=4.1$ T and $q_c=0$. This indicates, assuming that the contribution of the nonmagnetic $^1T_{2g}$ state to W^e is independent of magnetic field, that the position of the hot-magnon band is shifted in proportion to H. This is consistent with our experimental results, which were analyzed as follows. The value of $g\mu_B H$ was obtained from the

shift of the peak of a pure-exciton band (Fig. 4), which is 4.8 cm^{-1} at 4.1 T. From the neutron scattering results[11] it was estimated that the shift of the A band should be -0.9 cm^{-1} in a field of 4.1 T. This is close to the experimental value, -1.2 cm^{-1}, observed in the same field (Fig. 4), and gives support to the suggestion made in paper I that the A band is attributable to zone-edge magnons.

From process (4) in Fig. 9 we can expect the observation of a hot-magnon band which involves a zone-center magnon. In zero magnetic field the energy gap between the ground state and the zone-center magnon state is considered to be so small[10,11] that it is impossible to separate the transitions from the ground state and magnon states by using optical methods. In a strong field the gap energy is larger, and the conditions for the observation of zone-center magnons become more favorable. Indeed, such a state has been observed by Raman scattering in a field of 4.7 T.[7] We tried to observe a zone-center magnon band under the tail of the B band but without success. The X band shown in Fig. 3 cannot be assigned to a zone-center magnon because its separation from the B band, about 37 cm^{-1}, is too large. Cibert and Merle d'Aubigne have observed a weak band on the low-energy side of the ${}^3A_{2g}\rightarrow{}^3T_{2g}$ pure-exciton line.[8] This band is resolved from the background under a magnetic field, and as the field is increased the band shifts to lower energies and its intensity decreases. When we plot the separation between the pure-exciton band and the sideband against H by using the data of Ref. 8, we find that it resembles the magnon-gap energy at the zone center,[7,14] $[g\mu_B H(A+g\mu_B)]^{1/2}$, quite well. Taking into account that the intensity of this band decreases with increasing magnetic field, as was the case for the A band, this sideband can presumably be attributed to the hot-magnon band involving the zone-center magnon.

Let us return to the decrease of the intensity of the A band in a magnetic field. Because the transverse field increases the energy of magnons by an amount $\Delta W^m(H)$, their population is decreased at a finite temperature by a factor $\exp[-\Delta W^m(H)/k_B T]$, giving rise to a reduction of the intensity of the hot-magnon band. As can be calculated from the values given above, $\Delta W^m(H)$ is equal to $1.38H$ cm^{-1}, where H is given in T. Therefore, if the variation of the population is assumed to be predominant for the reduction of the absorption intensity, we can expect that the absorption of the hot-magnon band should be proportional to $\exp(-1.38H/k_B T)$. It was found that above 1.5 T (Fig. 6) the intensity of the A band is proportional to $\exp(-1.2H/k_B T)$, agreeing rather well with the above expectation. This suggests that the reduction of the intensity of the A band results from the reduction of the population of magnons due to their energy shift. A similar interpretation has been proposed also for the field dependence of a hot-magnon band in a 2D ferromagnet.[12] However, contrary to this example, the exponential law does not fit in the present case in fields below 1.2 T, where a more rapid reduction of the intensity of the A band was observed.

An anomalous narrowing of the pure-exciton line associated with the ${}^3A_{2g}\rightarrow{}^3T_{2g}$ transition has been found in magnetic fields below 1 T.[8] We observed a very similar narrowing of the pure-exciton bands and the hot-magnon band of the ${}^3A_{2g}\rightarrow{}^1T_{2g}$ transition. This indicates that the narrowing is a characteristic of the ground state rather than of the excited state. So far, three processes have been proposed to contribute to the bandwidths of the exciton lines in $CsNiF_3$: appearance of magnetic solitons, multimagnon states, and spin fluctuations.[8,18] Unfortunately, there are no theoretical predictions available about the field dependence of these three contributions to the half-width of the exciton and hot-magnon bands in a 1D ferromagnet. However, taking into account that (1) the anomaly has been observed both in the half-widths and the absorption intensities of the exciton bands and of the hot-magnon band, and (2) that these anomalies correspond to the field dependence of the magnetization,[11] the spin fluctuations seem to be a conceivable mechanism. We also observed that the half-width of the hot-magnon band is proportional to the temperature in a similar way to the half-width of the magnon peak observed by neutron scattering[11] and interpreted by the temperature dependence of the spin-correlation length. The magnetic-field-induced narrowing of the A band is clearly temperature dependent. In the field of 3.5 T the half-width of the band is reduced by 23% at 6.8 K but only by 12% at 13.3 K. The reduction of the band intensity is about 75% at 6.8 K and about 40% at 14 K in the same field. This indicates that thermal fluctuations of spins are responsible for the anomalies of the A band as well as of the B and C bands, especially when the external field is not strong enough to align the spins. The contribution of solitons[19] or multimagnon states to the linewidths of the exciton bands is still an open question. Within the accuracy of the existing experimental data, a conclusive analysis of this question is hardly possible.

Finally we discuss the origin of the X band shown in Fig. 3. We have already pointed out the difficulty of assigning this band to a zone-center magnon sideband of the B band. Here we temporarily propose the following assignment. As mentioned above, the $B_g({}^3A_{2g})$ level is located close to the ground-state level $A_g({}^3A_{2g})$. Therefore it is impossible to separate by optical methods the transitions into the A_g level and the B_g level in process (3) in Fig. 9. In a strong magnetic field the separation between the A_g and the B_g levels becomes large since the B_g state has $M_s=0$ (Ref. 7) and the A_g state has $M_s=-1$, so that a hot-magnon band may be detected when the magnons are annihilated to the B_g state and the excitons are created by process (2) in transitions from the A_g ground state. In this case the new hot band is located on the high-energy side of the A band, at the position $\Delta E_X = W(A_g)+g\mu_B H$. If we assume that the X band corresponds to this type of hot band, the value of $W(B_g)-W(A_g)$ is estimated to be about 8 cm^{-1} from $\Delta E_X = 13$ cm^{-1} measured in the field of 4.4 T. This energy difference is not unreasonable because it is expected to be of the same order with the exchange energy $J=7.99$ cm^{-1}.[7,11] When the magnetic field is increased, such a hot band is expected to be shifted to higher energies and its intensity should be decreased. This is qualitatively consistent with observation. The disappearance of the X band at high temperatures can be understood because under the conditions $k_B T > W(B_g)$

$-W(A_g)$ the splitting between the A_g and B_g states becomes meaningless in the transition process.

IV. CONCLUSIONS

The no-phonon, no-magnon exciton absorption bands B, C_1, and C_2, observed in the 1D ferromagnetic and 3D antiferromagnetic phases of $CsNiF_3$, are attributable to transitions to the components of the ${}^1T_{2g}^a$ state in C_{2h} symmetry. The pure-exciton bands have both electric dipole and magnetic dipole character. The hot-magnon band (A band) observed in the 1D ferromagnetic phase is due to electric dipole transitions. The dependence of the position and the intensity of the hot-magnon band on a magnetic field applied in the easy plane is understood by the energy shift of Brillouin-zone-edge magnons due to the field. The magnetic-field-dependent anomalous behavior of the half-width and the absorption intensity of both the exciton and the hot-magnon bands in fields below 1 T is suggested to be caused by the reduction of the in-plane spin fluctuations.

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