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Investigations of the magnetic and lattice vibrations in CoBr_2 using neutron scattering

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Résumé. — Les phonons acoustiques et les magnons ont été étudiés dans CoBr_2 par la diffusion inélastique des neutrons. Les courbes de dispersion des phonons acoustiques ont été mesurées dans les trois principales directions de symétrie, à la température ambiante. Les vitesses du son et les constantes élastiques en ont été déduites. Le caractère bidimensionnel de la structure cristallographique est mis en évidence par la différence entre les vitesses du son se propageant parallèlement et perpendiculairement aux plans. La valeur obtenue pour les paramètres d'ordre du réseau magnétique ($\beta = 0,3$) est en accord avec celle attendue dans le cas d'un modèle d'Ising à 3 dimensions. La dispersion des magnons a été étudiée dans les directions (100) et (001) à 5 K. On rend compte de la dispersion observée en utilisant un échange isotrope à l'intérieur du doublet fondamental issu du terme 4T_1 . La renormalisation des ondes de spins met en évidence un fort couplage magnon-phonon dans la direction (001).

Abstract. — Inelastic neutron scattering has been used to investigate both the acoustic phonons and the magnetic excitations in CoBr_2 . The acoustic phonon dispersion curves were measured in three principal symmetry directions at room temperature. From the initial slopes the sound velocities have been derived and the elastic constants calculated. The two dimensional nature of the crystallographic structure is apparent in the difference between the sound velocities parallel and perpendicular to the closed packed layers although the difference is less than in other halides. Furthermore, the order parameter of the magnetic lattice was found to be $\beta = 0.3$ a value not inconsistent with that expected for a 3 D Ising model. The magnon dispersion was investigated in the (100) and (001) directions at 5 K. The observed dispersion could be explained using a model containing an isotropic exchange within the ground state doublet of the 4T_1 term. Renormalization of the spin waves revealed a strong magnon-phonon coupling in the (001) direction.

1. Introduction. — A vast number of experiments have recently been performed on transition metal dihalides. The principal reason for this extensive interest is that the compounds have a particularly simple « layer » structure which influences the dimensionality of both the lattice dynamics and the magnetic properties. For instance, the compound FeCl_2 is an excellent example of a two dimensional ferromagnet [1] with the static component of the crystal field producing a strong single ion anisotropy. In addition the crystal field strongly perturbs the orbital motion of the electron outside the half closed shell in the Fe^{2+} ion. This modulation is transmitted to the spin system by spin orbit coupling [2]. The components of the crystal field linear in the displacements of the ferrous and ligand ions from their

equilibrium positions creates a coupling between the magnons and phonons. Magnon-phonon hybridization between the acoustic magnons and the transverse acoustic phonons in the Σ direction has been observed using high resolution neutron spectroscopy [3]. In the present paper we are primarily interested in establishing the dimensionality of the lattice dynamics and magnetic properties and the possible existence of magnon-phonon coupling in the related compound CoBr_2 .

2. Crystal structure and related properties. — Anhydrous cobalt dibromide has the hexagonal CdI_2 structure with space group $P3m1(D_{3d}^3)$. The structure is composed of hexagonal sheets of ferromagnetically aligned cobaltous ions with a trigonal crystal field

distortion producing a strong anisotropy fixing the moment direction in the plane. A much weaker anisotropy, arising from a slight monoclinic distortion of the crystal field, aligns the spin direction within the plane along a (210) axis [4]. A very weak antiferromagnetic coupling exists between the hexagonal planes, which are sandwiched between two layers of bromide ions, so the overall magnetic structure is antiferromagnetic with a Néel temperature of 19 K [5]. Thus the magnetic structure is similar to that observed in CoCl_2 which has the CdCl_2 crystallographic structure, space group $R\bar{3}m$, but in which the Co^{2+} ions have the same site symmetry as in CoBr_2 .

3. The specimen. — The single crystal of CoBr_2 which was grown from the melt by S. Legrand of Saclay was of a high quality with a mosaic width of $< 0.3^\circ$. A sample of dimensions suitable for inelastic neutron scattering investigations, $30 \times 5 \times 2 \text{ mm}^3$, was cut from the boule together with several smaller pieces for diffraction studies. Preparation of the specimen was carried out under a dry atmosphere of helium gas and each crystal was wrapped in thin aluminium foil in order to avoid hydration.

4. Static properties. — **4.1 SUBLATTICE MAGNETIZATION.** — The order parameter in an antiferromagnet is simply the sublattice magnetization $M(T)$ and in the asymptotic limit the sublattice magnetization should exhibit a power law dependence on the reduced temperature.

$$\frac{M(T)}{M(0)} = D \left(1 - \frac{T}{T_N} \right)^\beta. \quad (1)$$

4.2 EXPERIMENTAL. — The sublattice magnetization was measured as a function of temperature by integrating the magnetic Bragg peaks using the thermal neutron diffractometer D2 at the ILL Grenoble. The crystal was aligned with a (110) axis vertical inside a variable temperature cryostat mounted on the diffractometer. The diffractometer was used in normal beam geometry with a wavelength of $1.22 \text{ \AA}$ provided by a germanium (311) crystal, to minimize $\lambda/2$ contamination. Temperature stability of better than $\pm 0.01 \text{ K}$ was monitored by a Si diode and a carbon resistor thermometer placed in thermal contact with the sample. Integrated intensities of several magnetic reflections (003) (101) (009) etc. in the zero layer were measured by performing ω scans. Care was taken following each change in temperature to ensure that the sample had in fact reached thermal equilibrium by monitoring the intensity until it became constant.

4.3 RESULTS. — The temperature variation of the magnetic Bragg peaks (003) (101) and (009) is shown in figure 1. It may be seen that within the experimental error the intensities obtained from each Bragg peak

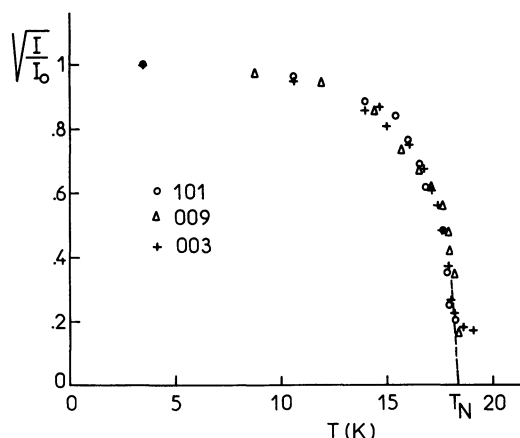


Fig. 1. — The thermal variation in the Bragg intensities of the magnetic reflections (101), (003) and (009).

as a function of temperature fall on the same curve. As the Néel temperature was approached, $T_N = 18.9 \text{ K}$, the monitor count was increased to minimize the statistical error consistent with other errors and a longer time was required before the sample reached thermal equilibrium. The Néel temperature had previously been established by observing the thermal variation of the magnetic critical scattering. Above 16 K, dI/dT was large and although temperature stability was monitored to better than $\pm 0.1 \text{ K}$ the absolute temperature at the sample position could not be estimated better than $\pm 0.05 \text{ K}$.

The relative magnetization shown in figure 2, was estimated assuming that the magnetization at 5 K was the saturation value. Taking $M(5 \text{ K}) = M(0)$, then at each temperature

$$\frac{M(T)}{M(0)} = \frac{I(T)^{1/2}}{I(5 \text{ K})^{1/2}}$$

and it is this quantity which is plotted in figure 1. Corrections for critical scattering were performed by carefully analysing the profile of the observed magnetic peaks and assuming the critical component to have a Lorentzian shape. The contribution of critical scattering to the observed magnetic intensities was found to be negligible except for the three temperatures closest

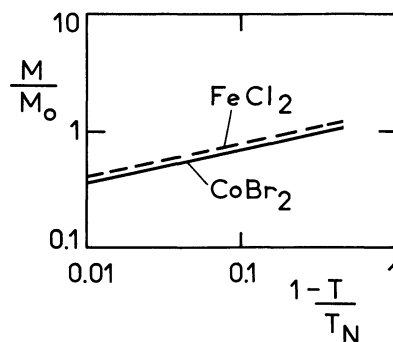


Fig. 2. — The temperature dependence of the sublattice magnetization.

to T_N at which measurements were made. A least square refinement of the magnetization was carried out using equation (1) with the intensity weighted according to their statistical significance. The best fit of the reduced sublattice magnetization to the power law, equation (1), is shown in figure 2. Only points above 15 K were included in the fit since the power law only applies close to T_N . The parameters obtained with the best fit were

$$\begin{aligned} D &= 1.36 \pm 0.08 \\ \beta &= 0.3 \pm 0.01 \\ T_N &= 18.9 \pm 0.05 \text{ K} . \end{aligned}$$

The values obtained for β and D are in reasonable agreement with the values calculated for the three dimensional Ising model [6] namely $\beta = 0.312$ and $D = 1.43$, although the value obtained for D depends on the assumption that $M(5 \text{ K}) = M(0)$. The magnetization below 5 K is effectively temperature independent and any difference is less than the experimental error in D . As in FeCl₂ [7] the critical behaviour in CoBr₂ is three dimensional but in FeCl₂ the magnetic excitations and lattice dynamics exhibit two dimensional properties. The dimensionality of the magnetic and lattice dynamics in CoBr₂ will now be discussed.

5. Dynamic properties. — **5.1 EXPERIMENTAL.** — Inelastic neutron scattering experiments were carried out using the three axis spectrometer IN8 located on a thermal source at the ILL, Grenoble. For energy transfers less than 1.5 THz pyrolytic graphite was used as both monochromator, analyser and filter. The spectrometer was operated with a fixed final wave vector of $2.662 \text{ \AA}^{-1}$ and the filter was aligned in the scattered beam. Horizontal collimation of 50', 40', 40' and 40' with respect to the outgoing beam was employed producing an energy resolution of 0.12 THz. For energy transfers larger than 1.5 THz the (111) reflection from a copper crystal was used as monochromator. The graphite filter was removed and final wave vectors of between 3 and $4 \text{ \AA}^{-1}$ were employed. For some scans requiring improved energy resolution the copper monochromator and a final wave vector of $2.662 \text{ \AA}^{-1}$ were used.

Measurement of the acoustic phonons in the Δ , Σ and Λ directions were carried out at room temperature. The magnetic excitations were studied at 5.5 K in the Λ and Σ symmetry directions only. For the magnetic measurements the large single crystal of CoBr₂ was mounted with a (110) axis vertical inside the same cryostat as used in the investigation of the static properties. In addition to studying the magnon dispersion, selected neutron groups were studied as a function of temperature until they disappeared close to T_N .

5.2 ACOUSTIC PHONONS. — Skeletal neutron scattering data concerning the dispersion of acoustic phonons in the compounds FeCl₂ [8] and CoCl₂ [9]

and more extensive results on FeBr₂ [10] have been reported. These results together with zone centre phonon frequencies obtained from Raman and Infra red measurements have enabled comparison with detailed model calculations. Pasternak [11] extended the static displaced shell model to include static intra ionic polarization and short range central forces between neighbouring shells and obtained reasonable agreement with the experimental curves. However, the physical significance of some the parameters derived in the model calculation has been questioned by Benedek [12]. In particular the ionicities (ionic charge of the cations) were found to be severely reduced, for example 0.06, 0.41 and 0.324 for FeCl₂, CoCl₂ and FeBr₂ respectively. The absence of van der Waals forces in Pasternak's model could account for the low ionicities. More recently, an extended shell model for ionic layer compounds has been proposed by Frey [13]. This model which includes both static and dynamically induced electronic dipoles has been applied to several dibromides and dichlorides with success. For the short range interactions they included central forces up to fourth nearest neighbour, that is, four intra layers and one inter layer interaction. The polarization part is composed of two shell charges and two isotropic shell-core springs. An excellent fit to the observed phonon dispersion was obtained for those materials for which neutron results existed. The parameters obtained from the model were physically reasonable and varied from compound to compound in a systematic manner. In particular the ionicities obtained for FeCl₂ CoCl₂ and FeBr₂ were 0.86, 0.8 and 0.83 respectively. Particularly, good agreement between theory and experiment was obtained for MnCl₂ for which detailed phonon dispersion curves including both acoustic and optic branches have been published. The cation Mn^{2+} in MnCl₂ [14] is in an S state and is therefore unlikely to produce significant coupling to the ligand lattice and therefore magnon-phonon coupling. A certain degree of covalency has been observed in MnF₂ [15] by polarized neutron diffraction but such effects in compounds containing Mn^{2+} ions are of less importance than in compounds containing Fe^{2+} ions. The degree of ionicity obtained for the model calculations of MnCl₂ was 0.91. The effects of covalency in CoBr₂ [16] are important and the possibility of a strong interaction between the metal ions and the ligand lattice may give rise to significant magnon phonon coupling. We have therefore undertaken an investigation of the acoustic phonons to facilitate identification of possible hybridized modes.

The dispersion of acoustic phonons propagating in the three principal symmetry directions are presented in figure 3. The abscissa has been scaled in reciprocal Ångströms to facilitate comparison. Six independent elastic constants, consistent with the $3m$ crystal symmetry may be estimated from the derived sound velocity. In table I are recorded the sound velocities and the elastic constants determined from the appropriate

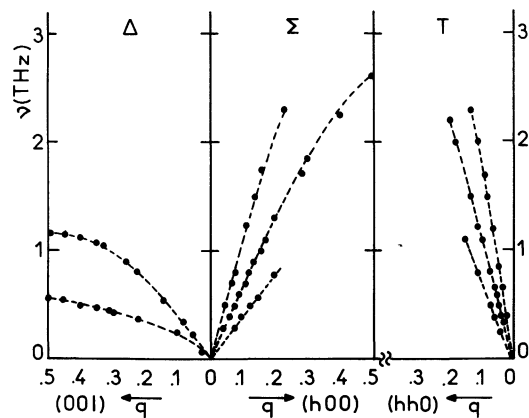


Fig. 3. — Acoustic phonon dispersion in CoBr₂ at 300 K.

initial slopes of the phonon branches. The elastic constants shown in table I are in excellent agreement

with those calculated by Benedek using the extended shell model. The polarization of each branch was also established. Comparison of the longitudinal sound velocity, « in » and perpendicular to the plane yielded a ratio of 1.2. In CoCl₂ this ratio was close to 2 [9] indicating the two dimensional character of phonon dispersion in the bromide is less pronounced than in the chloride.

As observed in other halides e.g. FeCl₂ and CoCl₂ the zone boundary frequencies of the phonons propagating in the (00ζ) direction are much lower than zone boundary frequencies in either the (ζζ0) or (ζ00) directions. Such behaviour is associated with the material being quite soft and weakly bound along the *c* axis. A consequence of this, is that these materials cleave quite easily perpendicular to *c* and are therefore difficult to cut to the optimum dimension for neutron experiments.

Table I. — Sound velocities and derived elastic constants of CoBr₂ at 300 K ($\rho = 4.923 \text{ g/cm}^3$).

Wave propagation direction	modes	sound velocity m.sec ⁻¹	elastic constants	wave polarization direction
[001]	Δ ₁ LA	2720 ± 100	C ₃₃ = 32 ± 1 C ₄₄ = 7.1 ± 1	[001]
	Δ ₃ TA	1070 ± 50		[001]
[100]	-T ₁ LA	3390 ± 100	C ₁₁ = 57 ± 5	[100]
	-T ₂ TA	2120 ± 100		near [120]
	-T ₃ TA	1360 ± 50		near [001]
[120]	-Σ ₁ LA	3220 ± 100	C ₆₆ = 26 ± 2 C ₁₂ = 14 ± 8 C ₁₄ = -7 ± 10	near [120]
	-Σ ₁ TA	1260 ± 100		near [120]
	-Σ ₂ TA	2100 ± 100		[100]

5.3 MAGNETIC EXCITATIONS. — The dispersion of magnons propagating in the Σ and Δ symmetry-direction at 5.5 K has previously been reported [17, 18]. Yoshizawa *et al.* [19] have also reported measurements made in the Δ direction. The results of the present investigation are shown in figure 4. In the (001) direction the degeneracy of the modes has been lifted with the upper mode exhibiting very little dispersion. However, in the Σ direction the lower branch disperses quite strongly to the zone boundary. The upper branch in the (ζ00) direction is not shown in figure 4 since the associated neutron groups proved to be less well defined for wave vectors smaller than ζ = 0.2. However, the broad groups observed suggested that the mode dispersed gradually out from the zone centre and at ω = 0.67 THz and eventually at approximately ζ = 0.2 followed closely the lower branch out to the

zone boundary. The modes appeared to be degenerate at the zone boundary. The upper branch involves spin fluctuations along the C* axis. Since only components of magnetization perpendicular to the scattering vector give rise to magnetic neutron scattering it is possible to identify the fluctuations by arranging the scattering geometry accordingly. The proposed spin direction (210) [4] and the associated structure factors hinders complete fulfillment of the geometrical requirements.

Thus with the imposed scattering plane it was necessary to perform scans involving less favourable resolution conditions in order to observe the upper mode in the (h00) direction. Therefore the observed groups tended to be much broader than those obtained under conditions of focus. Furthermore, since the spin direction lies within the plane, three domains are formed when cooling the crystal through the Néel

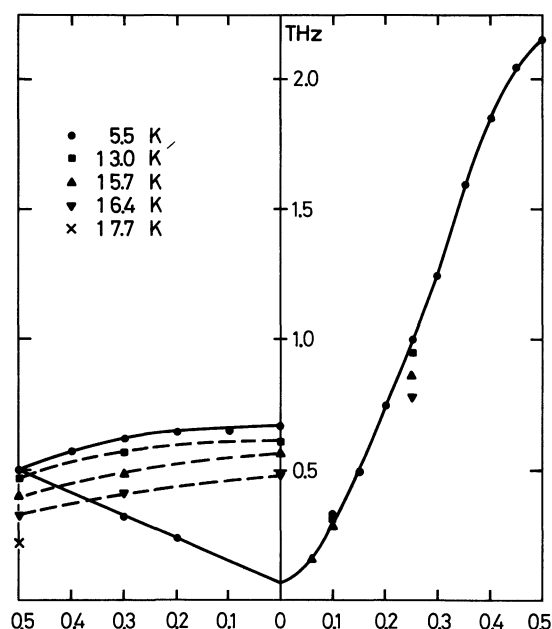


Fig. 4. — The observed magnon dispersion in the ($\zeta 00$) and (00ζ) directions of CoBr₂ at 5 K.

temperature, making identification of the modes difficult. A predominantly single domain crystal was formed by cooling the crystal in a magnetic field of 2T.

From figure 3 it may be seen that at the zone centre the energy gap associated with the upper branch corresponds to 0.67 ± 0.04 THz. This value is in excellent agreement with the antiferromagnetic resonance value of 0.675 ± 0.008 [4, 20]. The intrinsic energy resolution of the instrument (IN8) did not permit the energy gap of the lower branch to be established. A value for the zone centre frequency, of 0.064 ± 0.006 THz was established by A.F.M.R. [4].

Analysis of the inelastic neutron measurements of the spin dynamics in CoBr₂ at 5.5 K was carried out using an effective spin Hamiltonian. The original analysis [18] was performed using tensor algebra but to facilitate a discussion of the derived parameter the more familiar vector notation, as used by Yoshizawa [19], will be employed here.

5.4 MODEL CALCULATION. — The Hamiltonian appropriate to the Co²⁺ ions in CoBr₂ has been discussed by [21, 22]. The free-ion 3d⁷(⁴F) multiplet is split by the predominantly cubic field of the surrounding ligand ions, i.e. bromide, into ⁴T_{1g}, ⁴T_{2g} and ⁴A_{2g} states. Although the ground state ⁴T_{1g} is slightly perturbed by T_{1g}(⁴P) state the lowest triplet may be considered an effective ⁴P state with orbital momentum $l = 1$. Within this ⁴P state the Hamiltonian, in the absence of an applied field, may be written

$$\mathcal{H} = \sum_i \left\{ -\frac{3}{2} \lambda k l_i \cdot S_i - \delta \left([l_i^2] - \frac{2}{3} \right) \right\} - \sum_{i < j} 2 S_i \tilde{J}_{ij} S_j$$

where z is taken to be parallel to the crystallographic C axis. The first summation corresponds to single ion terms which include spin orbit coupling and trigonal crystal field distortions and the second summation comprises the bilinear exchange term, between ion pairs. The exchange term is small in comparison with the spin orbit and crystal field terms and it may therefore be treated as a perturbation. Under the influence of the spin orbit coupling and the residual components of the crystal field the ground state splits into six Kramers doublets. In the Hamiltonian the free ion value of the coupling constant is represented by λ and k is a reduction factor ($k < 1$) introduced by Tinkham to account for the covalent bonding and the mixing of the ⁴P level. From a measurement of the Raman spectrum at 21 K, i.e. above the Néel temperature, Lockwood *et al.* [23] obtained $k\lambda = -151 \text{ cm}^{-1}$ (-4.53 THz) $k = 0.85$ and $\delta = -272 \text{ cm}^{-1}$ (-8.16 THz). The resulting energy level diagram is shown in figure 5. As in CoCl₂ [4] the parameter δ is negative indicating that the c axis is the hard direction of magnetization whereas for FeBr₂ and FeCl₂ δ is positive and the spins are fixed along the c direction. The exchange interactions giving rise to magnetic order below 18.9 K are much smaller than the separation between the lowest doublets namely $\sim 8 \text{ THz}$ (382 K) and therefore admixtures of higher levels into the ground state may be neglected. This assumption is further justified by the knowledge that the zone boundary magnon frequencies are less than 2.5 THz. Thus the magnetic properties of CoBr₂ can be described using a model in which an isotropic exchange between ionic spins $S = 3/2$ is projected onto the ground state doublet of the ⁴T_{1g} term to produce an anisotropic exchange interaction for an effective spin $S' = 1/2$. The contribution of the exchange interaction to the Hamiltonian is then written as

$$\mathcal{H}_{\text{ex}} = -2 \sum_{i > j} (J_{ij} S'_i \cdot S'_j + D_{ij} S'_{ix} S'_{jx})$$

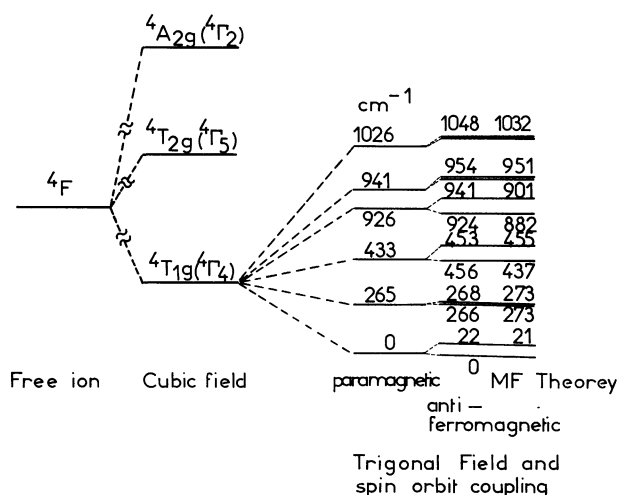


Fig. 5. — The proposed energy level diagram of Co²⁺ in CoBr₂ at 5.5 K.

where the anisotropy in the interaction is negative and given by

$$\frac{D}{J} = (\alpha_{\parallel}^2 - \alpha_{\perp}^2)/\alpha_{\perp}^2$$

$\alpha_{\parallel}$ and $\alpha_{\perp}$ are the principal components of the tensor coupling the real and effective spins. The subscripts $\parallel$ and $\perp$ refer to the hexagonal C axis which is taken to be the x direction in the coordinate system. Since the spins are confined in the plane the ratio $\alpha_{\parallel}^2/\alpha_{\perp}^2$ which is a function of the trigonal crystal field components, is less than 1.

After diagonalization the dispersion relationships for the two magnon branches become

$$E_1 = \bar{S} \{ (A - B)(A - C + E - F) \}^{1/2}$$

$$E_2 = \bar{S} \{ (A - C)(A - B + E + F) \}^{1/2}$$

where the intra atomic exchange interaction between neighbours i is given by

$$A = \sum_{i=1}^n Z_i J_i (1 - \gamma_{iq}); \quad E = \sum_{i=1}^n Z_i D_i \gamma_{iq}$$

and the inter atomic exchange between spins on adjacent planes by

$$B = Z' J' (1 - \gamma_q) \quad \text{and} \quad C = Z' J' (1 + \gamma_q)$$

$$F = Z' D' \gamma_q$$

Z_i is the number of i th neighbours at positions δ_i . The average spin is given by $\bar{S}$ and the prime superscript refers to the inter layer parameters. The Fourier transform γ is given by $\gamma_{iq} = (Z_i)^{-1} \sum \exp(iq \delta_i)$ yielding for the $(h0l)$ plane

$$\gamma_1 = \frac{2}{3} \cos 2\pi h$$

$$\gamma_2 = \frac{1}{3} (\cos 4\pi h + 2 \cos 2\pi h)$$

$$\gamma_3 = \frac{2}{3} (\cos 4\pi h) \text{ etc.}$$

and

$$\gamma' = \cos 2\pi l.$$

The interaction parameters J_i and D_i were determined by a least square fit of the dispersion relation to the observed frequencies. The observed frequencies had previously been corrected for the effects of instrumental resolution using the method of Cooper and Nathans [24]. In table II are displayed the parameters associated with the best fit as indicated by a χ^2 test. Significant improvement to the fit was obtained by extending the exchange interaction within the plane out to third nearest neighbours.

Comparison of the parameters shown in table II with those obtained by Yoshizawa *et al.* [19] indicates close agreement. The ratio of the inter to intra planar exchange interactions — J'/J is 0.47. A similar value of

Table II. — *The results of the best fit of the dispersion relation to the observed spin wave spectrum at 5 K.*

J_1	=	0.503 THz
J_2	=	0.005 THz
J_3	=	-0.084 THz
J'	=	-0.237 THz
D_1	=	0.280 THz
D_2	=	0.002 THz
D_3	=	0.002 THz
D'	=	-0.005 THz

0.482 was obtained by Ziebeck *et al.* [25] for the related compound CoCl_2 . Such large ratio are indicative of poor two dimensionality. The ideal X - Y system would have $D_i/J_i = 1$ and for the Heisenberg model this ratio would be zero. In CoBr_2 $\frac{D_i}{J_i}$ was found to have an intermediate value of 0.56. Originally lines [15] predicted $D_i/J_i = 0.425$. Although Ziebeck *et al.* obtained $D_i/J_i = 0.42$ from observation of the magnetic excitations propagating along C^* in CoCl_2 , Hutchings [26] found it necessary to have $D/J \neq D'/J'$. The principal difference between the Hamiltonian used by Ziebeck and that of Hutchings was the inclusion of an intra planar anisotropy term of the form $v(l_x^2 - l_y^2)$ arising from a monoclinic distortion of the crystal field.

5.5 TEMPERATURE DEPENDENCE. — A series of scans in both the (00ζ) and $(\zeta 00)$ directions in which the wave vector was held constant were carried out as a function of temperature. The neutron groups observed at four different temperatures for a wave vector of $q = 0.1$ in the $(\zeta 00)$ and $q = 0.5$ in the (00ζ) are shown in figure 6. The renormalization of the upper mode at the zone centre was found to decrease, within experimental limits, in a manner proportional to the reduced magnetization with reduced temperature. Thus above T_N the system was unable to support collective excitations consistent with a randomly aligned spin assembly [27].

With reference to figure 6 it may be seen that as the frequency of the magnetic excitation in the (00ζ) direction decreases with temperature its width becomes larger. Eventually at the highest temperature shown in figure 6 two peaks may just be resolved. The position of the upper peak was found not to change with temperature and its frequency was consistent with that of the degenerate transverse acoustic phonon branch. This was confirmed by other scans at different wave vectors. However, all measurements of the magnetic excitations were carried out in the magnetic Brillouin zone i.e. around magnetic Bragg peaks. Since the magnetic structure has a c axis twice that of the nuclear cell the zone centre of the magnetic Brillouin zone, along C^* , is the zone boundary of the nuclear zone. Therefore the only way that a phonon could be observed at its appropriate frequency but with its wave vector defined with respect to the magnetic lattice is

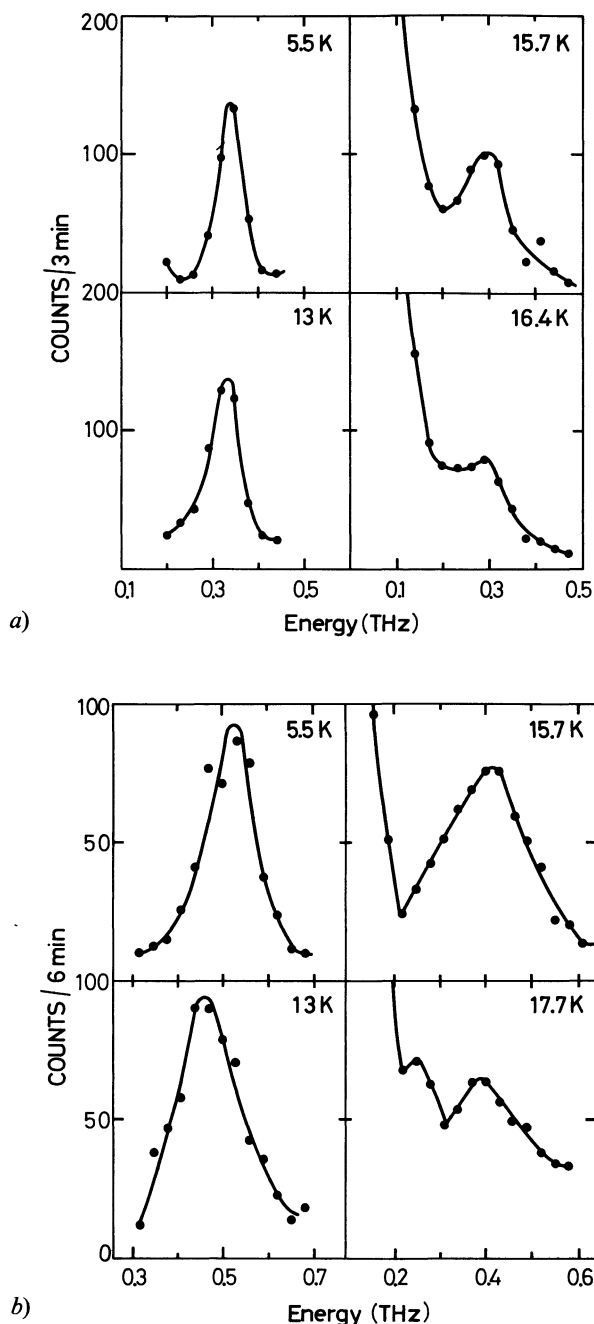


Fig. 6. — The thermal evolution of two neutron groups located at $q = 0.1$ in the $(\zeta 00)$ direction (a) and at $q = 0.5$ in the (00ζ) direction (b).

by hybridization. Cracknell [28] has shown by arguments of symmetry, that only the transverse acoustic phonon in the (00ζ) direction can couple to the magnon branch. Unlike FeCl_2 the degeneracy of the magnon modes, in the absence of an external field, in CoBr_2 is lifted by the anisotropic exchange. Lovesey [2] calculates an energy gap at the anticrossing point in the $(\zeta 00)$ direction in FeCl_2 of some 0.1 THz. Whilst the present results are not inconsistent with this estimate further measurements using long wavelength neutrons and possibly polarization analysis are required before a detailed understanding of the coupling can be obtained.

6. Summary. — The measurements reported in this paper indicate that although the crystallographic structure enables CoBr_2 to be described as a layer compounds the dynamic properties of the nuclear and magnetic lattice clearly do not have X-Y character.

The absence of two dimensionality is not unusual for magnetic compounds having the CdCl_2 or CdI_2 structures, as indicated previously. The poor two dimensionality in CoBr_2 is exemplified by the value of 0.3 obtained for the critical exponent of the sublattice magnetization as a function of the temperature. Within experimental error this value is not significantly different from the value $\beta = 0.29$ obtained for FeCl_2 [7] or from the theoretical rigid lattice Ising value $\beta = 0.312$. Since both Fe^{2+} and Co^{2+} ions are strongly coupled with the ligand lattice, the effects of renormalization and magnon phonon hybridization cannot be ignored in the interpretation of the experimentally determined critical exponents. This may account for the slight differences obtained for β and departure from the rigid lattice Ising value. As the temperature is raised the magnetic excitations renormalise close to T_N and collective excitations cease to propagate above. The decreasing frequency of the upper magnon branch in the (00ζ) direction produces a coupling to the transverse acoustic phonon branch. The origin of the hybridization is probably the Van Vleck mechanism of spin lattice relaxation but further measurements are required to quantify the interaction.

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