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MINIWORKSHOP ON STRONG CORRELATIONS
AND QUANTUM CRITICAL PHENOMENA
(4 - 22 July 1994)

EXPERIMENTAL STUDIES OF THE HALDANE CONJECTURE IN THE QUASI-ONE-DIMENSIONAL ANTIFERROMAGNETS

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D. PETITGRAND, LABORATOIRE LEON BRILLOUIN, CEN-SACLAY
} NEUTRONS

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* WE STUDY THE COMPOUNDS WITH THE LOCALIZED SPINS, WHICH ARE BELIEVED TO INTERACT AS

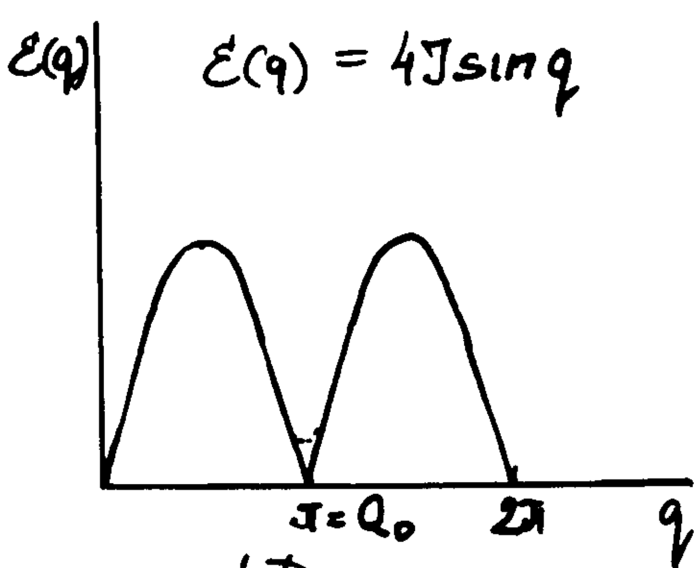
$$(1) \quad \hat{\mathcal{H}} = \sum_{i,j} J_{ij} \hat{\vec{S}}_i \hat{\vec{S}}_j + D \sum_i (\hat{S}_i^z)^2 - \gamma \vec{H} \sum_i \hat{\vec{S}}_i$$

MOST ELABORATED THEORETICAL DESCRIPTION - SPIN-WAVE THEORY BASED ON SEMICLASSICAL $1/S$ EXPANSION. IT GIVES:

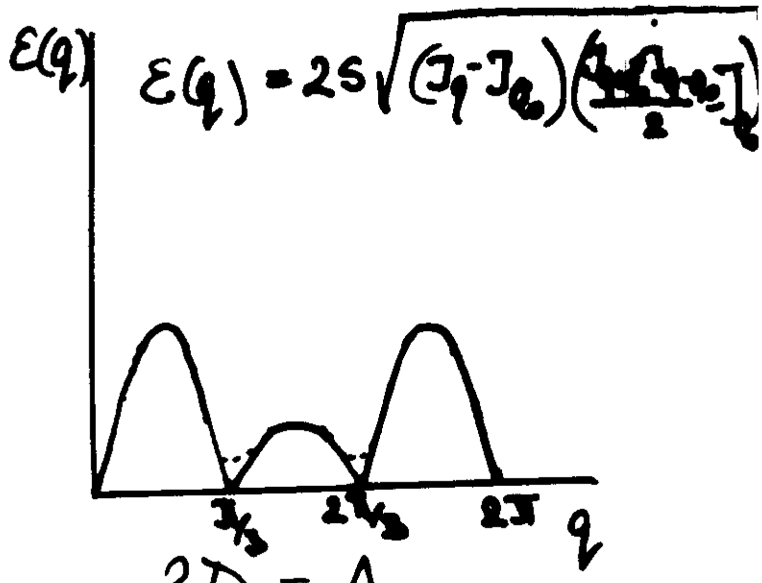
* THE GROUND STATE, NEEL ORDERED FOR AFM
  $|G.S.\rangle = |\uparrow\downarrow\uparrow\downarrow\uparrow\downarrow\uparrow \dots\rangle$ → TWICE DEGENERATE + CONTINUOUS DEGENERACY FOR $D \neq 0$

* IN MORE THAN 1 DIMENSION THE G.S. IS USUALLY DESCRIBED BY A HELIX WITH SOME PROPAGATION VECTOR Q_0. FOR 2D TRIANGULAR AFM $Q_0 = (\frac{1}{3}; \frac{1}{3})$, CORRESPONDS TO 120° ORDERING.

* THE ELEMENTARY EXCITATIONS - MAGNONS, $\varepsilon(q)$, "SOFTENING" AT $q = 0, Q_0$ FOR $D = 0$ IN ACCORDANCE WITH THE GOLDSTONE THEOREM.



1D



2D - Δ

* ORDER PARAMETER = SUBLATTICE AVERAGE SPIN

(2) $\langle \hat{S}_i^{z_i} \rangle_{G.S.} = S - \langle \hat{a}_i^+ \hat{a}_i \rangle_{G.S.} = \frac{1}{2}\left\{ \int \frac{d^n q}{\mathcal{E}(q)} A_q - 1 \right\}$

EVIDENTLY DIVERGING IN 1D (n=1) DUE TO THE GOLDSTONE MODES.

↓

IN 1D THE S.W.T. IS NOT SELF-CONSISTENT.

* IN 1966 MERMIN AND WAGNER, FOLLOWING HOHENBERG, PROVED THE ABSENCE OF THE NEEL ORDER IN 1 and 2 dimensions AT ANY FINITE TEMPERATURE.

* FOLLOWING SIMILAR ARGUMENTS, IN 1991 L. PITAEVSKII AND STRINGARI ESTABLISHED THE EXACT INEQUALITY

(3) $I(q) = \int d\omega\, S(q,\omega) \geq A \frac{\langle S_i^{z_i} \rangle_{G.S.}}{q}$

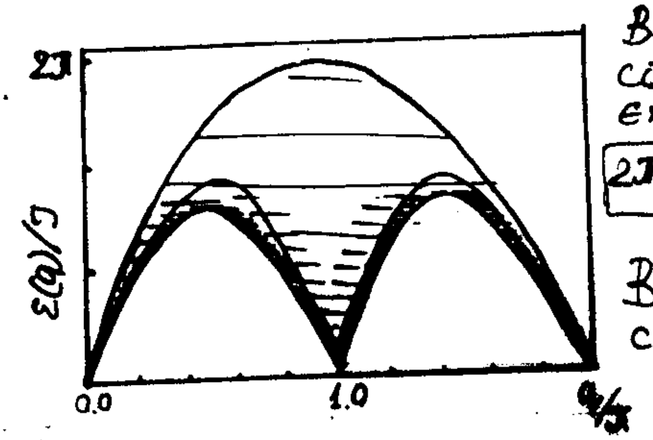
THUS PROVING THE ABSENSE OF THE A.F.M. ORDER ALSO IN THE GROUND STATE FOR 1D (AT T=0).

* ISING ANISOTROPY OR 3D INTERACTIONS CAN LIFT THE DIVERGENCY IN (2), (3) MAKING S.W.T. SELF-CONSISTENT AND 3D ORDER PERMITTED.

↑

CRUCIALLY DEPENDS ON THE STRUCTURE OF THE 1D G.S.: BEHAVIOR OF THE SPIN-SPIN CORRELATIONS

$\langle S_i^\alpha S_j^\alpha(t) \rangle_{G.S.} \rightarrow ? \leftarrow S_\alpha(q,\omega) = \int dt \sum_{\vec{r}_j} e^{i q \vec{r}_j - i\omega t} \langle S_i S_j(t) \rangle$



BETHE-ANZATZ CONTINUUM OF THE EXCITATIONS:

$2J\sin\frac{q}{2} < \mathcal{E}(q) < J\sin q$

BLUE LINE - SEMI-CLASSICAL SPIN WAVE

* ANZATZ SOLUTION (BETHE) FOR $S=\frac{1}{2}$ GIVES:
  * DISORDERED G.S. FOR THE A.F.M, WITH THE POWER-LIKE DECAY OF THE SPIN-SPIN CORRELATIONS:

$$(4)\quad \langle S_i S_j \rangle_{G.S.} \sim \frac{(-1)^{r_{ij}}}{|r_{ij}|}$$

  * GAPLESS EXCITATIONS FORMING A BAND AROUND $\mathcal{E}(q) \simeq \pi J \sin q$ (DES CLOISEAUX-PEARSON)
  * ACTUALLY NOT TOO FAR FROM SPIN-WAVE THEORY!?

* HALDANE (1983) CONSIDERING MAPPING OF A SPIN CHAIN ON THE NONLINEAR σ-MODEL THAT IS TRUE FOR $S \gg 1$ CLAIMED THE ABOVE BEHAVIOR TO BE THE CASE FOR $S=\frac{1}{2}\cdot$INTEGER DUE TO APPEARANCE OF ADDITIONAL TOPOLOGICAL TERM. FOR $S=$ INTEGER THE G.S. WAS CLAIMED TO CORRESPOND TO THAT OF σ-MODEL, I.E.:
  * G.S. IS NONDEGENERATE SINGLET, WITH THE EXPONENTIAL DECAY OF CORRELATIONS:

$$(5)\quad \langle S_i S_j \rangle_{G.S.} \sim \frac{(-1)^{r_{ij}}}{\sqrt{|r_{ij}|}} e^{-\varkappa |r_{ij}|} \;;\quad |G.S.\rangle = |S_{tot}=0\rangle$$

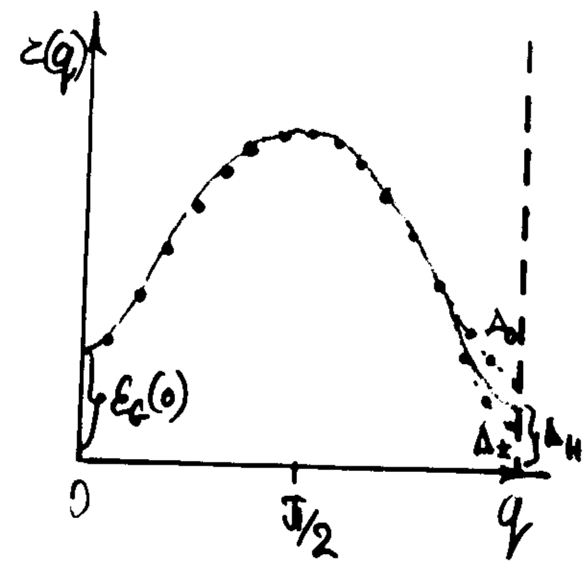
  * HENCE, THE ELEMENTARY EXCITATIONS - THE GOLDSTONE BOSONS IN THE SPIN-WAVE PICTURE, ACQUIRE A DYNAMICALLY GENERATED GAP,

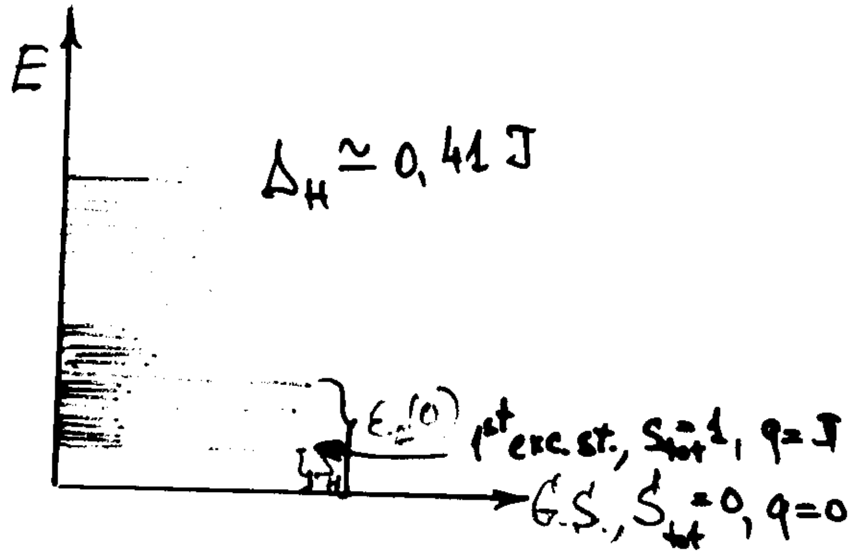
$$(6)\quad \min \mathcal{E}(q) = \mathcal{E}(\pi) = \Delta_H \sim e^{-\pi S}$$

HALDANE SPIN CHAIN FEATURES

* $|\text{GROUND STATE}\rangle = |\cdots 0\uparrow\downarrow\uparrow 0\downarrow\uparrow 000\downarrow\uparrow\downarrow\uparrow\cdots\rangle = |S_{tot}=0, q=0\rangle$

* EXCITATIONS SPECTRUM AND ENERGY LEVELS PICTURE





* $|1^{st}\text{ exc. STATE}\rangle = |S_{tot}=1,\ S^z_{tot}=0,\pm 1,\ q=\pi\rangle = |0,\pm 1; q=\pi\rangle$

MANY SPIN EXCITED STATES WITH OTHER q IN THE BRILLOUIN ZONE BUT STILL WITH $S_{tot}=1$, $S^z_{tot}=0,\pm 1$.

* ANISOTROPY IN THE HAMILTONIAN

$$\hat{H} = J\sum_i \vec{S}_i \vec{S}_j + D\sum \hat{S}_i^{z\,2} + E\sum (\hat{S}_i^{x\,2} - \hat{S}_i^{y\,2}) = \hat{H}_0 + \hat{V}_A$$

SPLIT THE TRIPLET AT EACH q. AT $D, E \ll J$

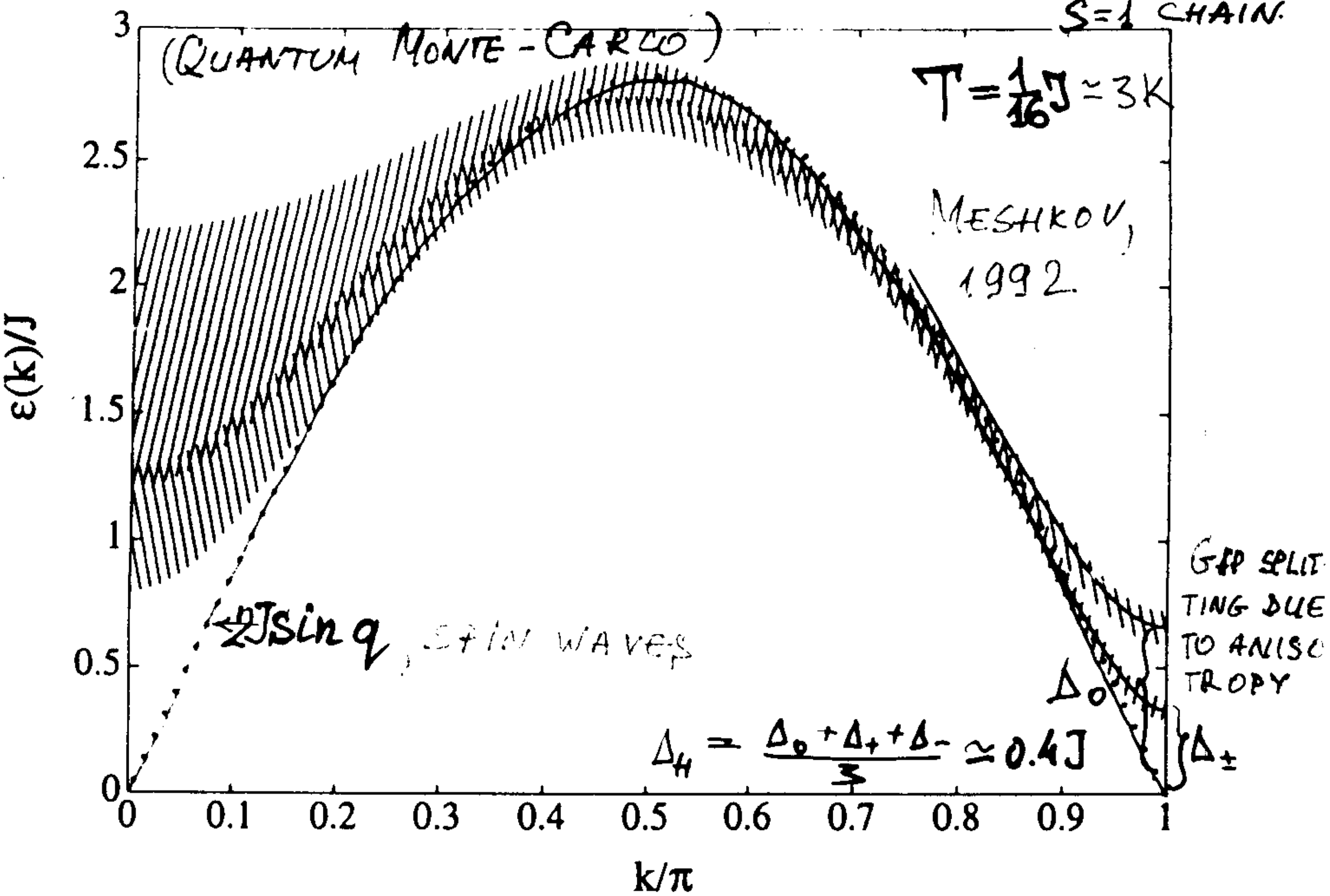
ONE CAN TREAT IT PERTURBATIVELY (FIRST $E=0$):

$$\Delta_H \to \begin{cases} \Delta_0 = \Delta_H + \langle 0,q|\hat{V}_A|0,q\rangle - N\frac{D}{3}S(S+1) \\ \Delta_\pm = \Delta_H + \langle \pm 1,q|\hat{V}_A|\pm 1,q\rangle - N\frac{D}{3}S(S+1) \end{cases}$$

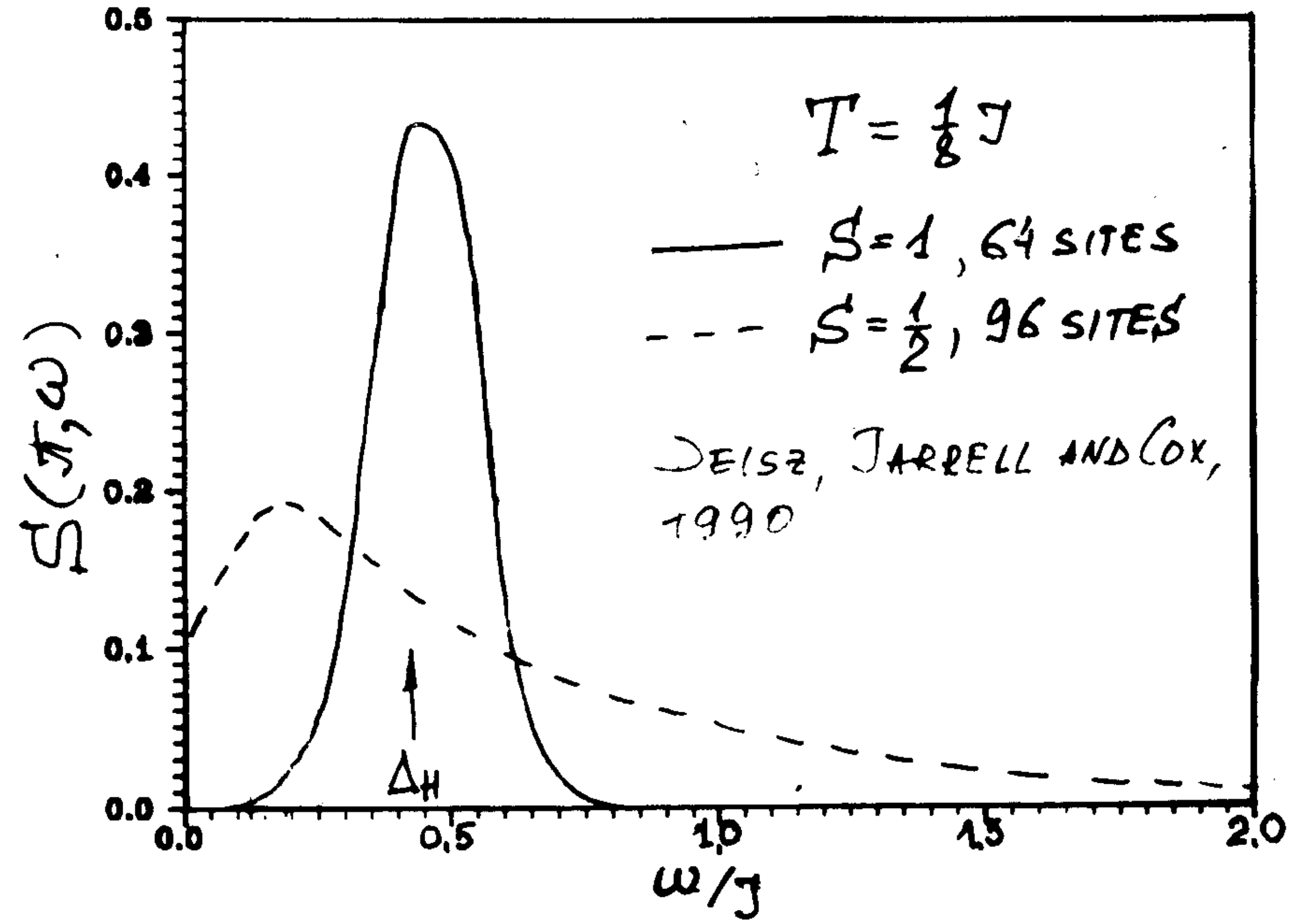
* ACCORDING TO THE NUMERICAL STUDIES OF FINITE CHAINS

$$\begin{cases} \Delta_0 = \Delta_H + 1{,}36D \\ \Delta_\pm = \Delta_H - 0{,}67D \end{cases}$$

THE AVERAGE GAP IS EQUAL TO Δ_H, DOES NOT CHANGE.



DIFFERENCE IS CLEARLY SEEN IN THE CALCULATED SHAPES OF THE DYNAMICAL STRUCTURE FACTOR

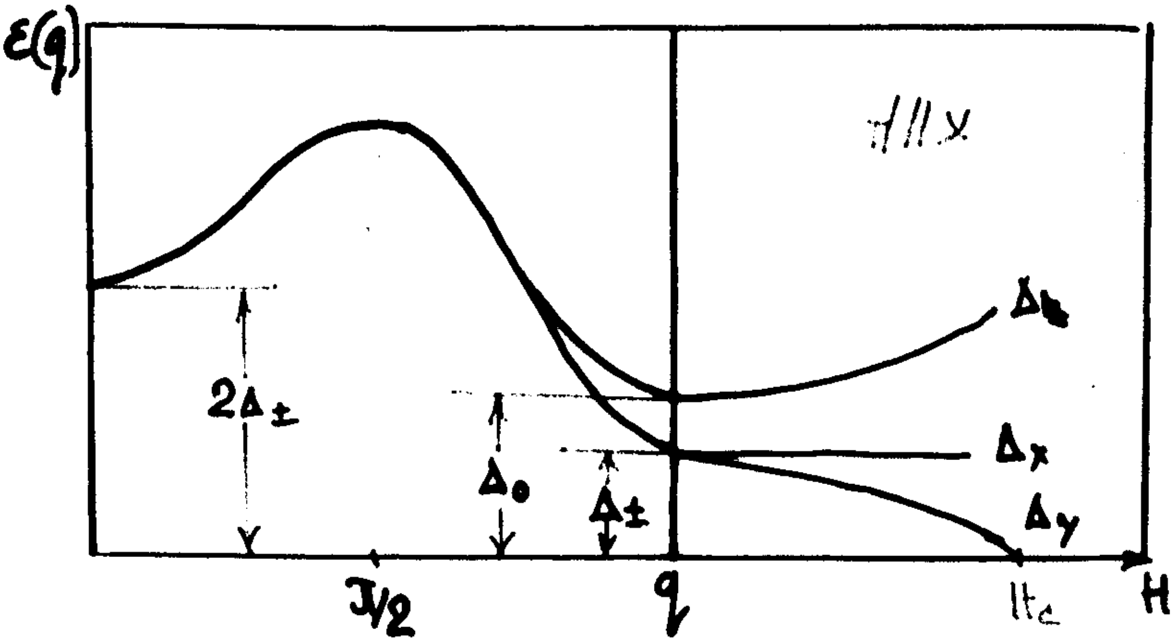


PLETELY. SO DOES THE MAGNETIC FIELD.

$$\hat{\mathcal{H}}_z = \gamma \vec{H} \sum_i \vec{S}_i = \gamma \vec{H} \vec{S}^z_{tot}$$

$\Delta_0 \to \Delta_z$

$\Delta_\pm \to \Delta_x, \Delta_y$



* THUS IN THE PERTURBATION THEORY THE EXCITATIONS ENERGIES AT EACH POINT IN THE BRILLOUIN ZONE SPLIT LIKE SINGLE-ION LEVELS:

H∥z

$$\begin{cases} \mathcal{E}_{1,2}(q,H) = \dfrac{\mathcal{E}_x(q) + \mathcal{E}_y(q)}{2} \mp \sqrt{\left(\dfrac{\mathcal{E}_1(q) - \mathcal{E}_2(q)}{2}\right)^2 + (\gamma H)^2} \\ \mathcal{E}_3(q,H) = \mathcal{E}_z(q) \end{cases}$$

* THE THERMODYNAMIC PROPERTIES AT $T \lesssim \Delta_\pm$ ARE DRIVEN BY THESE EXCITATIONS FROM THE VICINITY OF $q = \pi$ WITH THE DISPERSION

$$\mathcal{E}_\alpha(q) \simeq \sqrt{\Delta_\alpha^2 + C_\alpha^2 q^2} \quad \text{AT} \quad \mathcal{E}_\alpha(q) \lesssim 2\Delta_\pm$$

* THUS ONE EASILY ESTIMATE THE FREE ENERGY

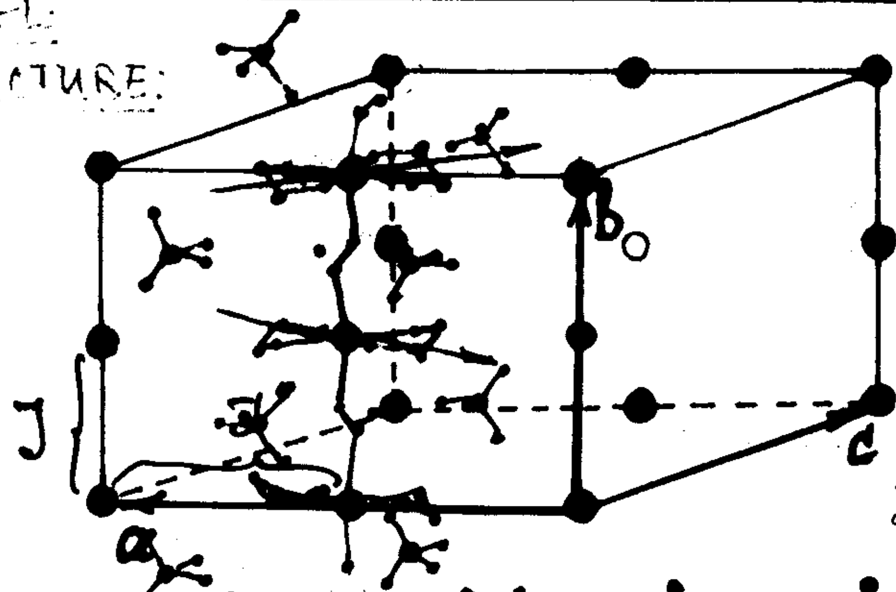
$$\mathcal{F}_m = (\mp) \sum_\alpha \sum_q \ln\left(1 (\pm) e^{-\frac{\mathcal{E}_\alpha(q)}{T}}\right)$$

? ?

AND IT'S DERIVATIVES – HEAT CAPACITY AND MAGNETIZATION

NENP = $Ni(C_8H_8N_2)_2NO_2ClO_4$

* CRYSTAL STRUCTURE:



●– Ni^{2+}; a=14,55Å, b=10,25Å, c=8,3Å
•–Cl ; •–O ; •–N ; •–H;

$J \simeq 46K \simeq$
$\simeq 1THz \simeq$
$\simeq 4{,}15 meV$

FROM THE DISPERSION IN [011] DIRECTION
$J'/J \lesssim 10^{-4}$

* SUCCEPTIBILITY SHOWS NO SIGNS OF THE MAGNETIC ORDERING DOWN TO $\sim 10 mK$

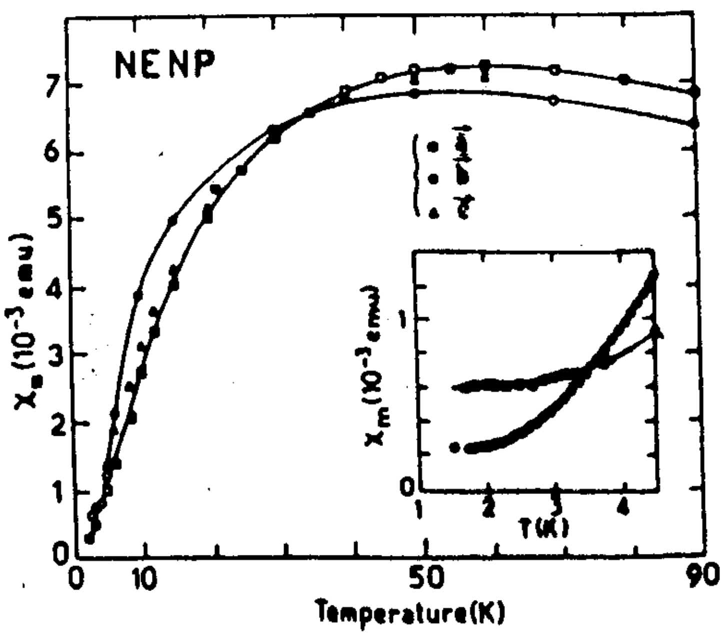
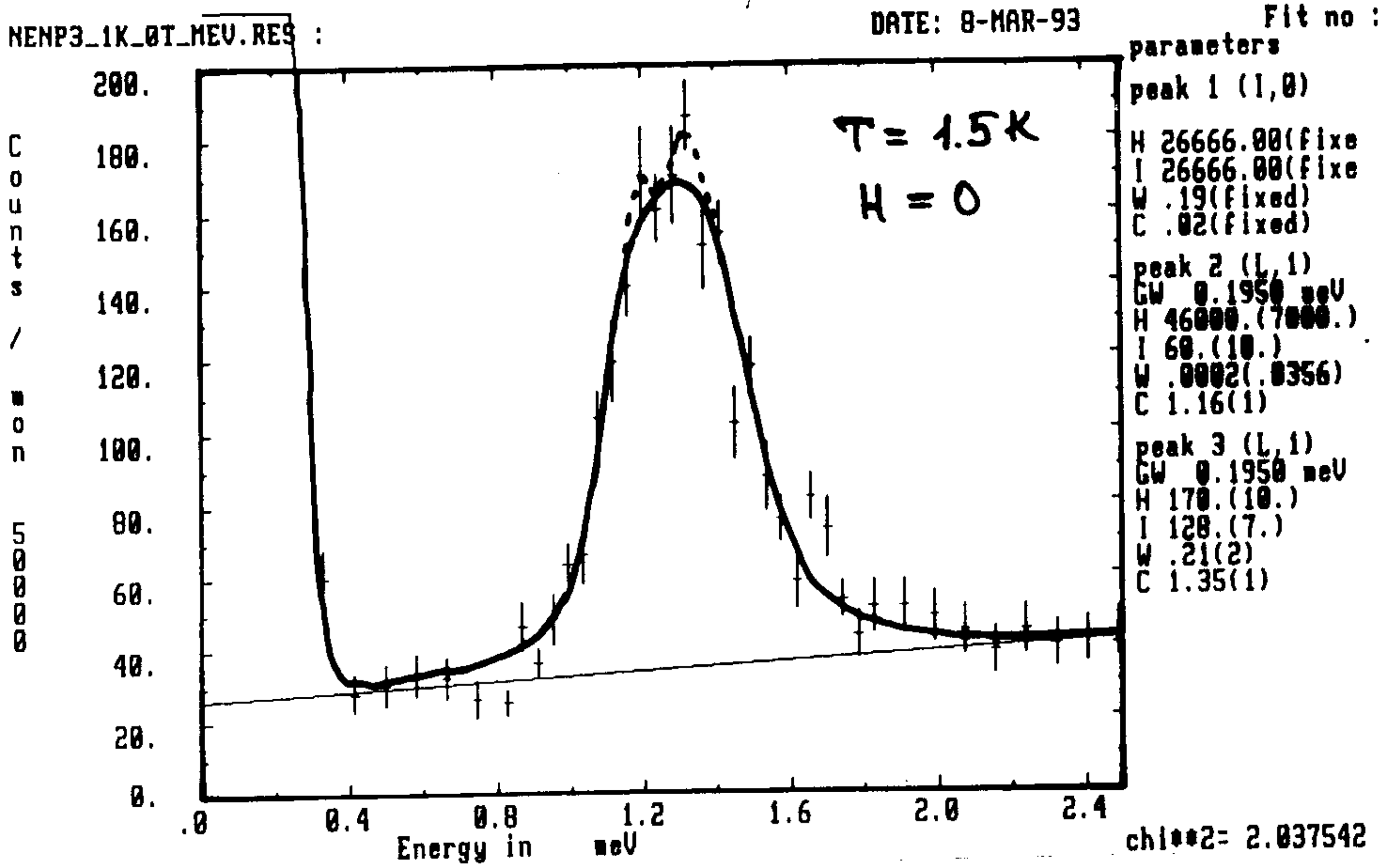
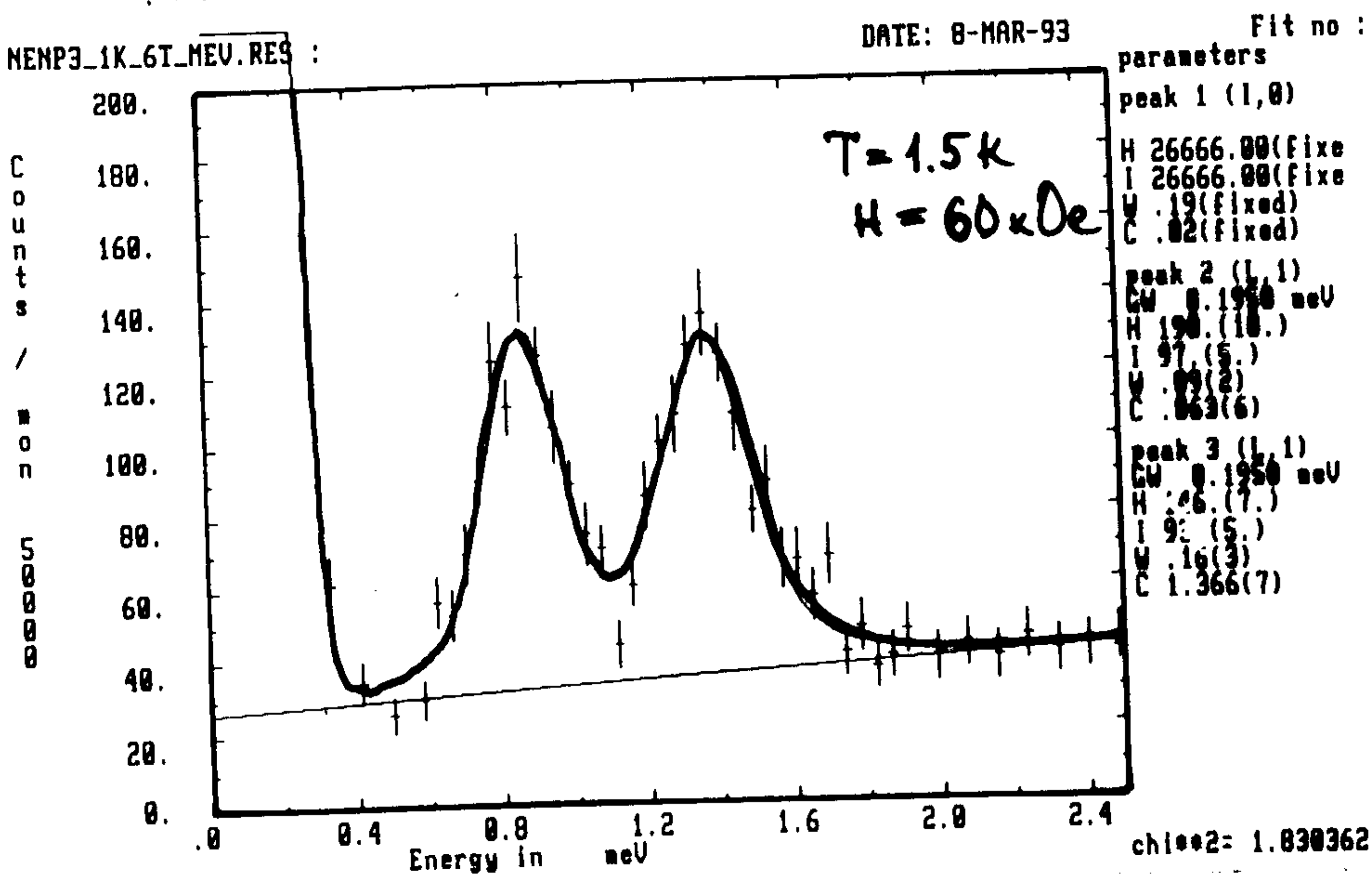


Fig. 1 Molar susceptibilities of NENP as a function of temperature .



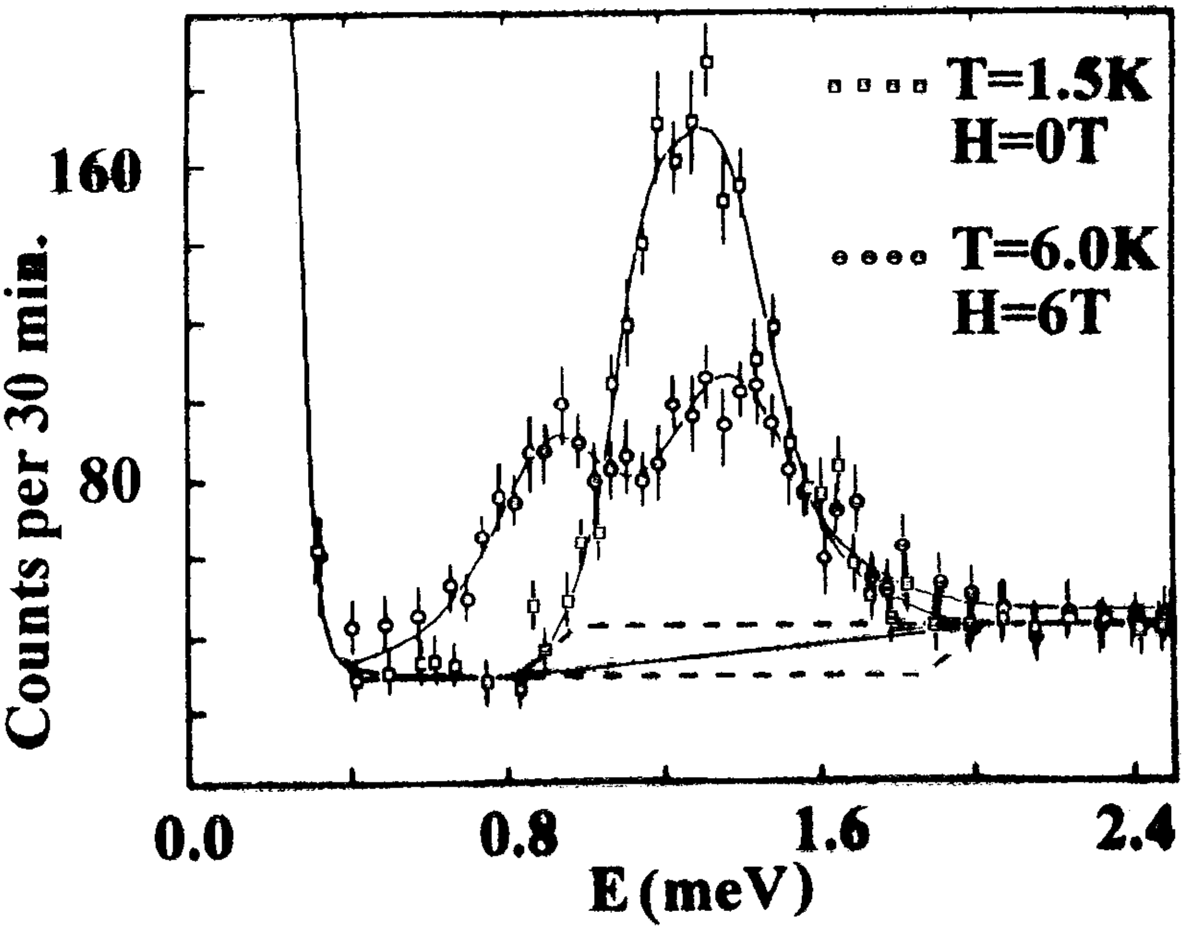
SPLITTING OF THE DOUBLET BY THE MAGNETIC FIELD



SCATTERING AMPLITUDE (VERTICAL AXES):

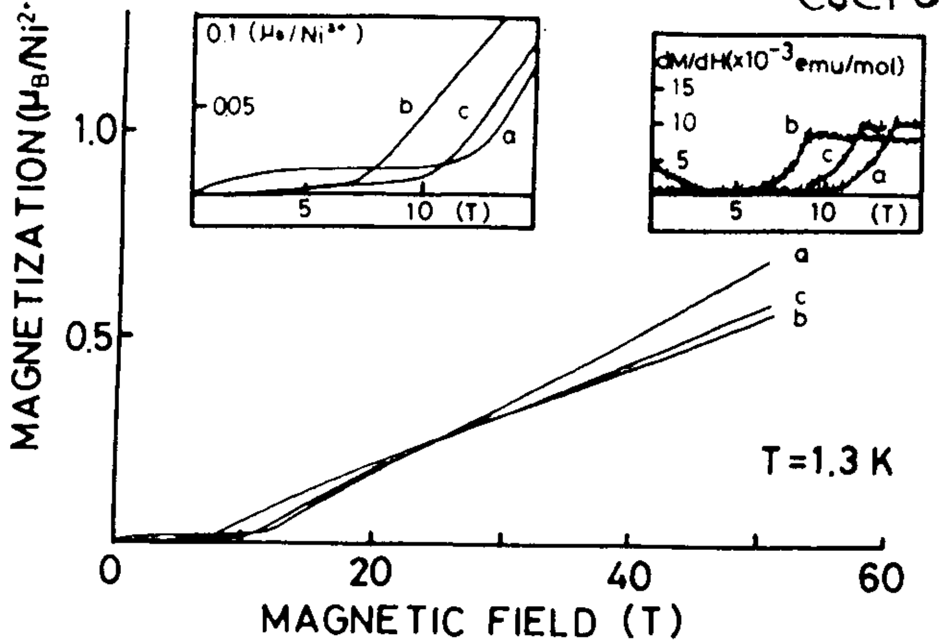
$$\frac{d^2\sigma}{dq\,d\Omega} \sim S(q,\omega) = \mathcal{F}\{\langle S_i S_j(t)\rangle\} = \left(1-e^{-\frac{\omega}{T}}\right)^{-1}\chi''($$

EVOLUTION OF THE EXCITATIONS PEAKS WITH TEMPERATURE: DESPITE THE BROADENING THEY ARE STILL WELL DEFINED UP TO T ~ Δ ~ 0.4J

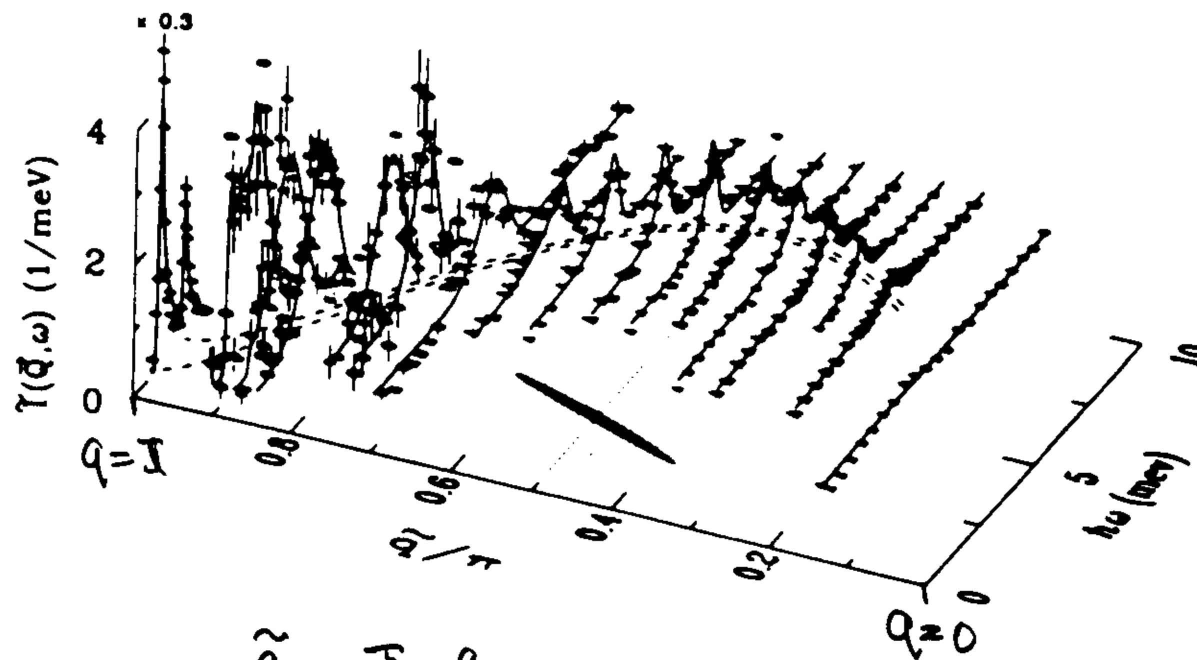


L.-P. REGNAULT, I. A. ZALIZNYAK, C. VETTIER AND J. P. RENARD, Phys. Rev. B, SEPTEMBER 1994.

Кривые намагничивания VENP для трех ориентаций поля. Виден переход в магнитное состояние.



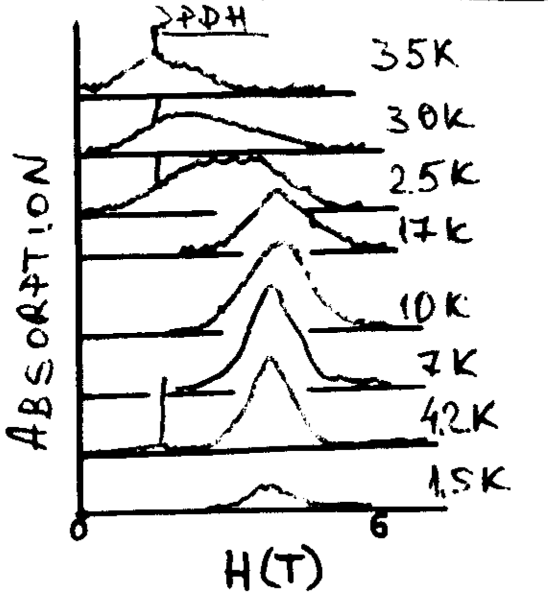
Измерение дисперсии элементарных возбуждений неупругим рассеянием нейтронов при T=1.3K. В окрестности $q=\pi$ данный эксперимент позволяет различить 2 пика. Структура нижнего сост. из 2^x не видна.

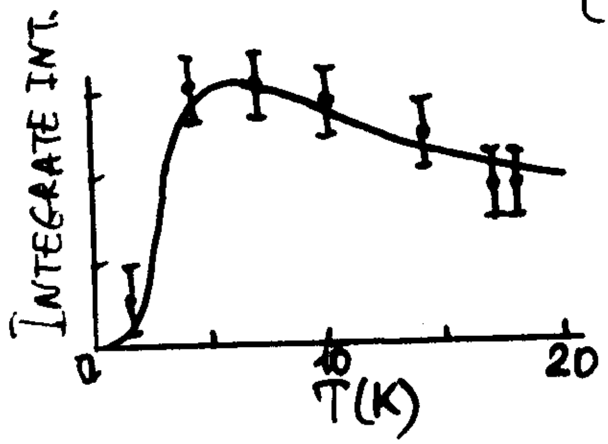


$$\tilde{q} = \pi - q$$

FIRST EXPERIMENTAL REPORT: M. DATE AND K. KINDO, Phys. Rev. Lett. 65, p. 1659 (1990).

$f = 470\,GHz$





* RATHER BROAD LINE AT ~ 4T WITH ACTIVATION-LIKE TEMPERATURE DEPENDENCE, TRANSFORMING INTO PARAMAGNETIC AT T ~ J.

* WHAT IS EXPECTED?

$$\chi''_{\alpha}(q\approx 0,\omega) \sim (1-e^{-\omega/T}) \sum_{i,j} e^{-E_i/T} |\langle i|\hat{M}^{\alpha}_{tot}|j\rangle|^2 \times \delta(\omega-(E_j-E_i)) =$$

$$= (1-e^{-\omega/T}) \sum_j |\langle G.S.|\hat{M}^{\alpha}_{tot}|j\rangle|^2 \cdot \delta(\omega-E_j) +$$

TRANSITIONS FROM THE G.S.

$$+ \sum_{\{\alpha,\beta=\pm,0\}} \left(e^{-\frac{\epsilon_\alpha(q)}{T}} - e^{-\frac{\epsilon_\beta(q)}{T}}\right) \cdot |\langle\alpha,q|\hat{M}^{\alpha}_{tot}|\beta,q\rangle|^2 \cdot \delta(\omega-(\epsilon_\beta(q)-\epsilon_\alpha(q)))$$

TRANSITIONS BETWEEN THE 1ST EXCITED TRIPLET

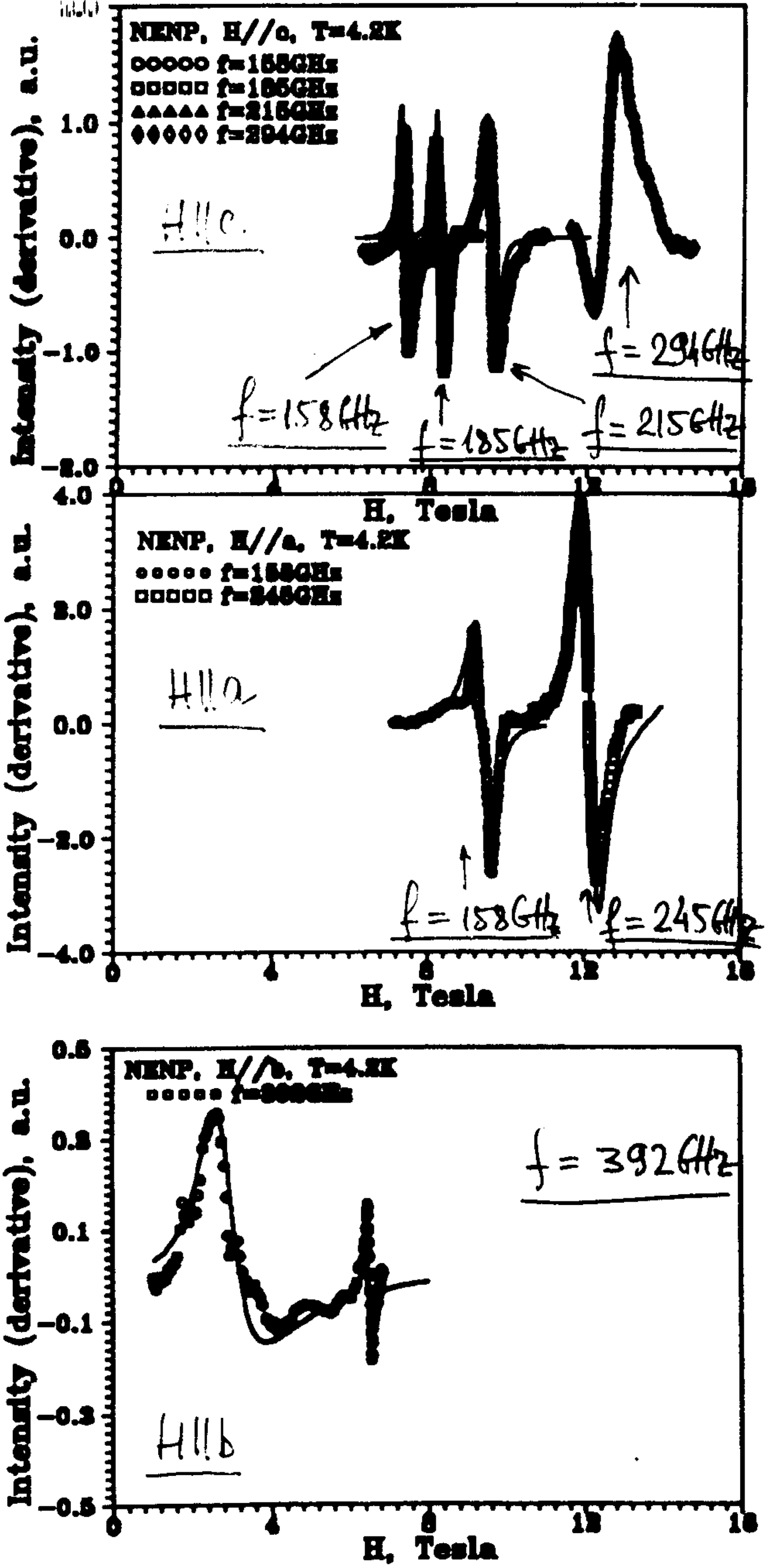
$+ \dots$

* FOR TRANSLATIONALLY INVARIANT G.S. TRANSITIONS TO THE 1ST EXCITED TRIPLET ARE PROHIBITED.
* NO TRANSLATIONALLY INVARIANT PERTURBATION $\hat{V}_A$ WILL ENABLE THEM.
* FOR PURE HEISENBERG CHAIN WITH $S_{TOT}=0$ IN THE GS ... ARE PROHIBITED BY ...

ESR ABSORPTION LINES AS OBSERVED IN NENP AT T=4.2K

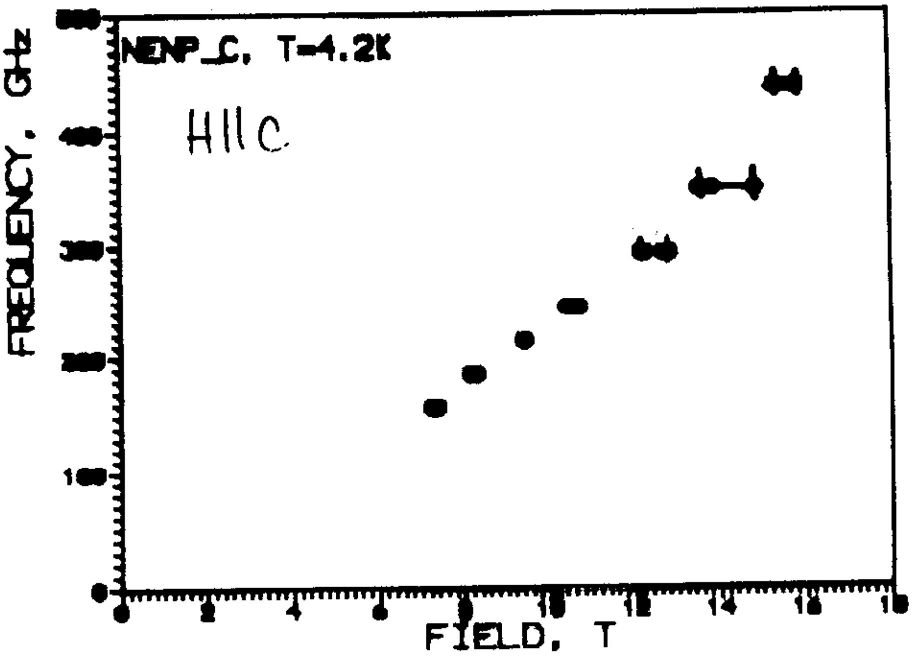
L.-C. BRUNEL, T. BRILL, I. ZALIZNYAK, J.-P. BOUCHER AND J.-P. RENARD

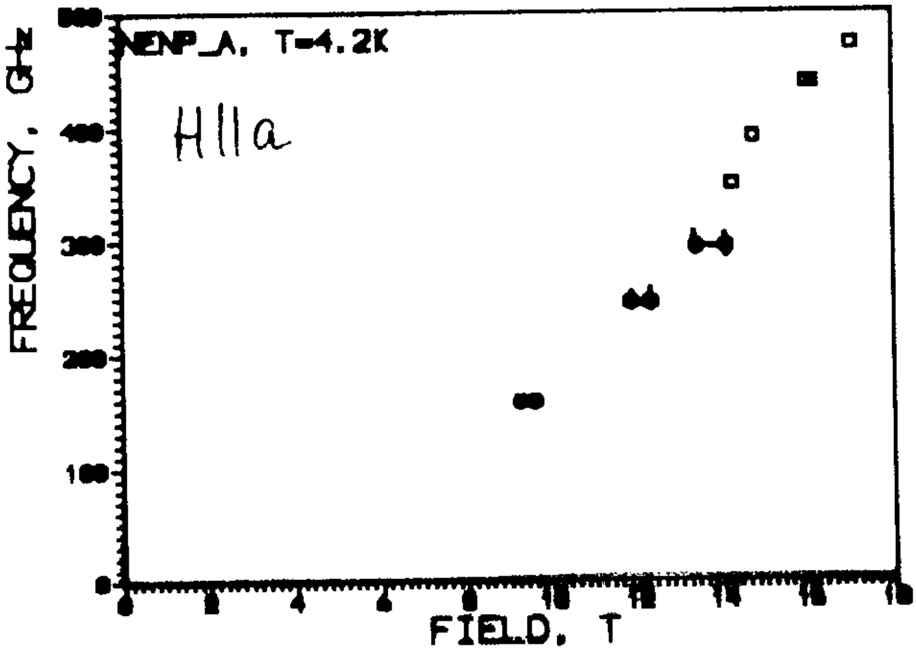
Phys. Rev. Lett. 69, p. 1699 (1992).

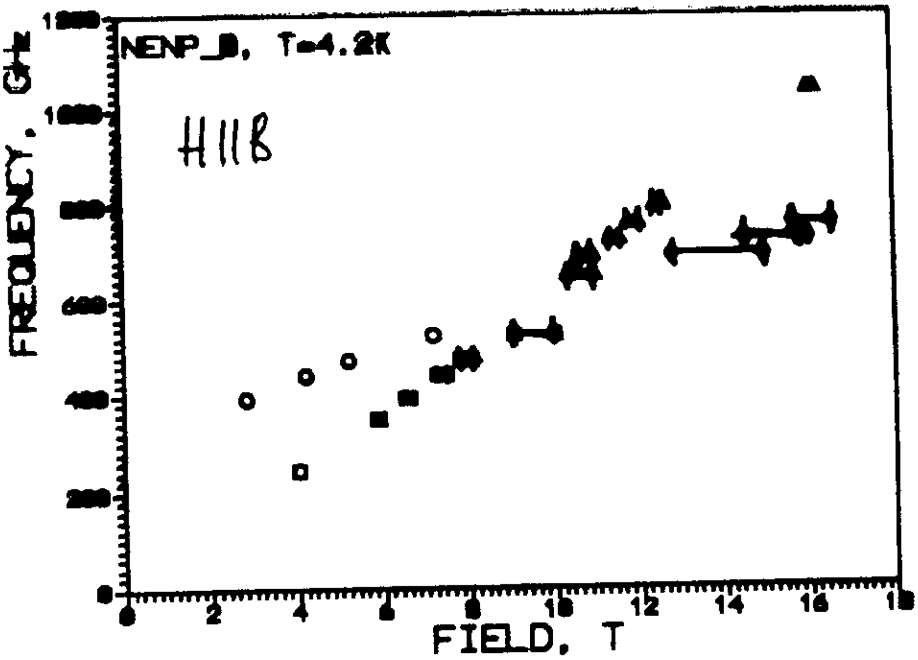


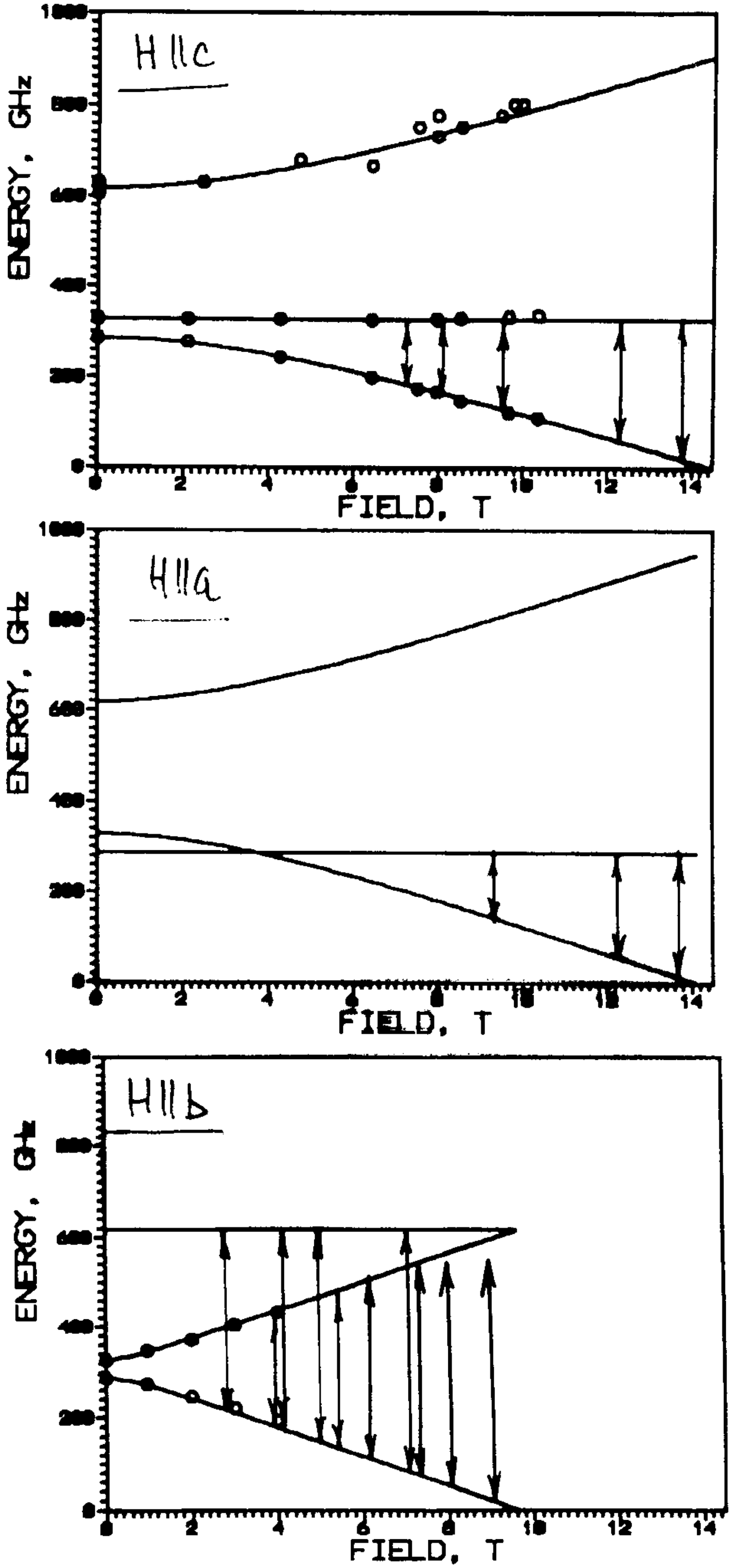
ABSORPTION.

T = 4.2 K









AT $q=\pi$

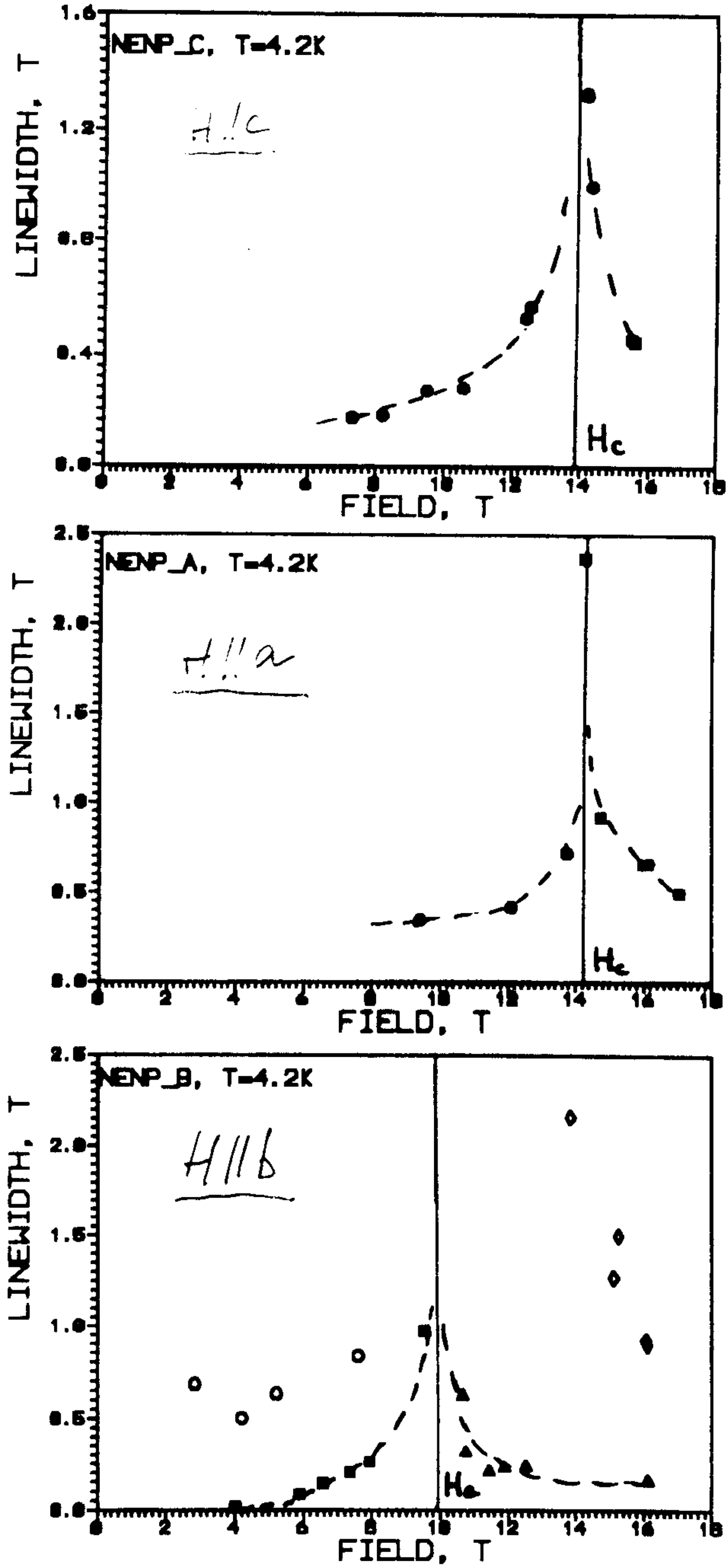
o – RESULTS OF NEUTRON SCATTERING IN NENP AT $T = 1.7K$

—— CALCULATIONS IN THE PERTURBATION THEORY (SPLITTING OF A MAGNETIC TRIPLET)

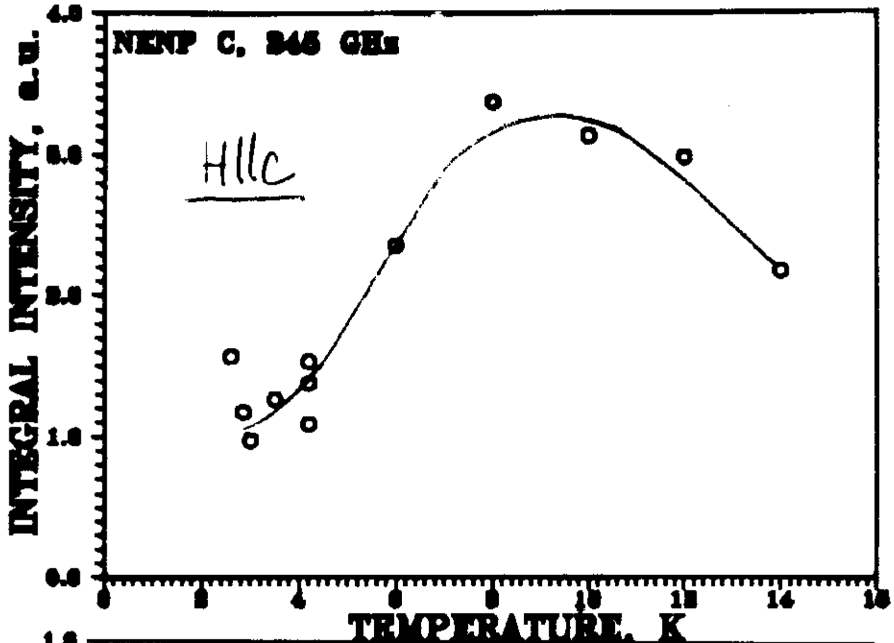
↕ TRANSITIONS PROBED BY ESR AT $T = 4.2K$

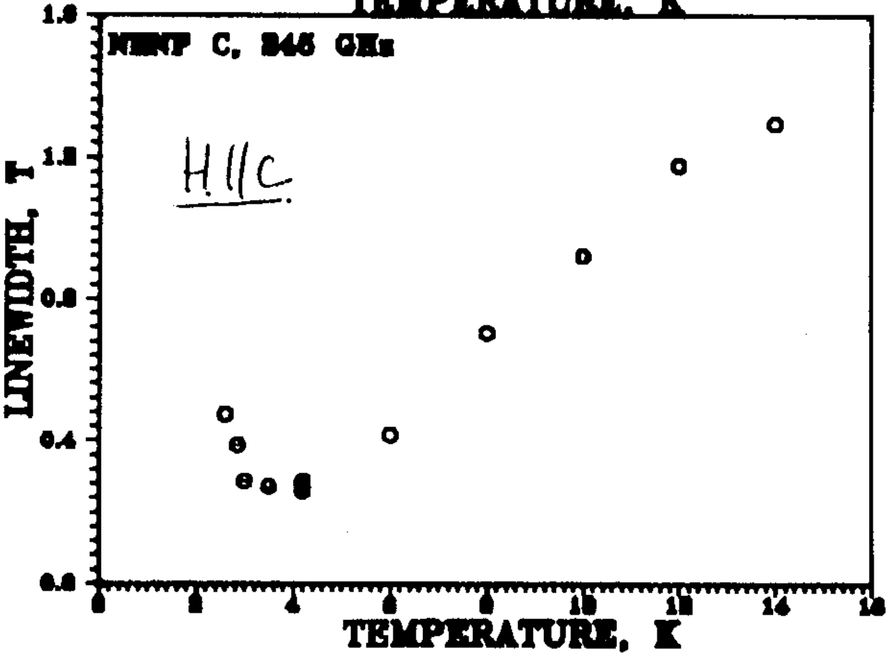
LINE IN NENP NEAR THE CRITICAL FIELDS H_c

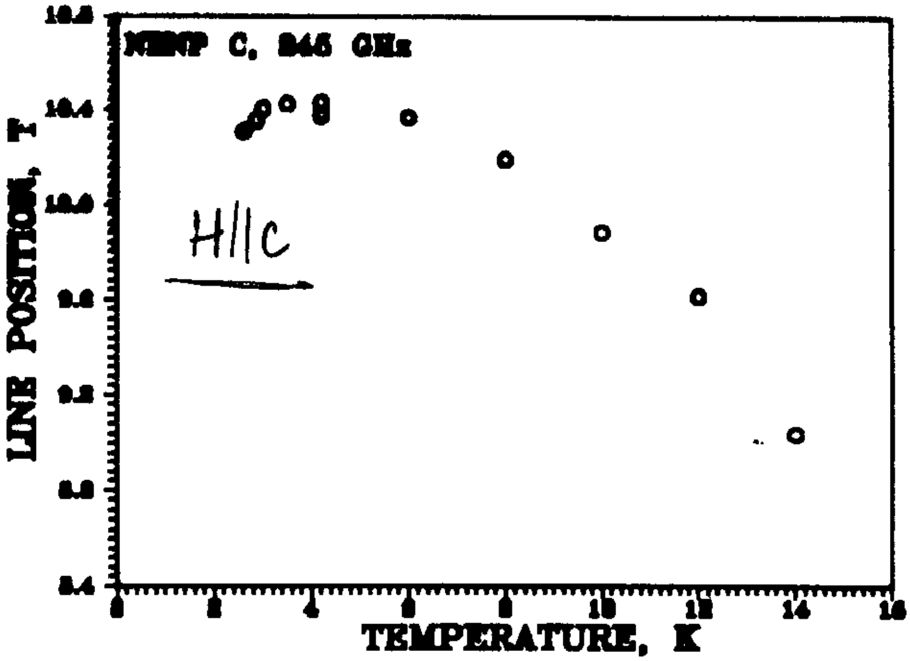
T = 4.2K

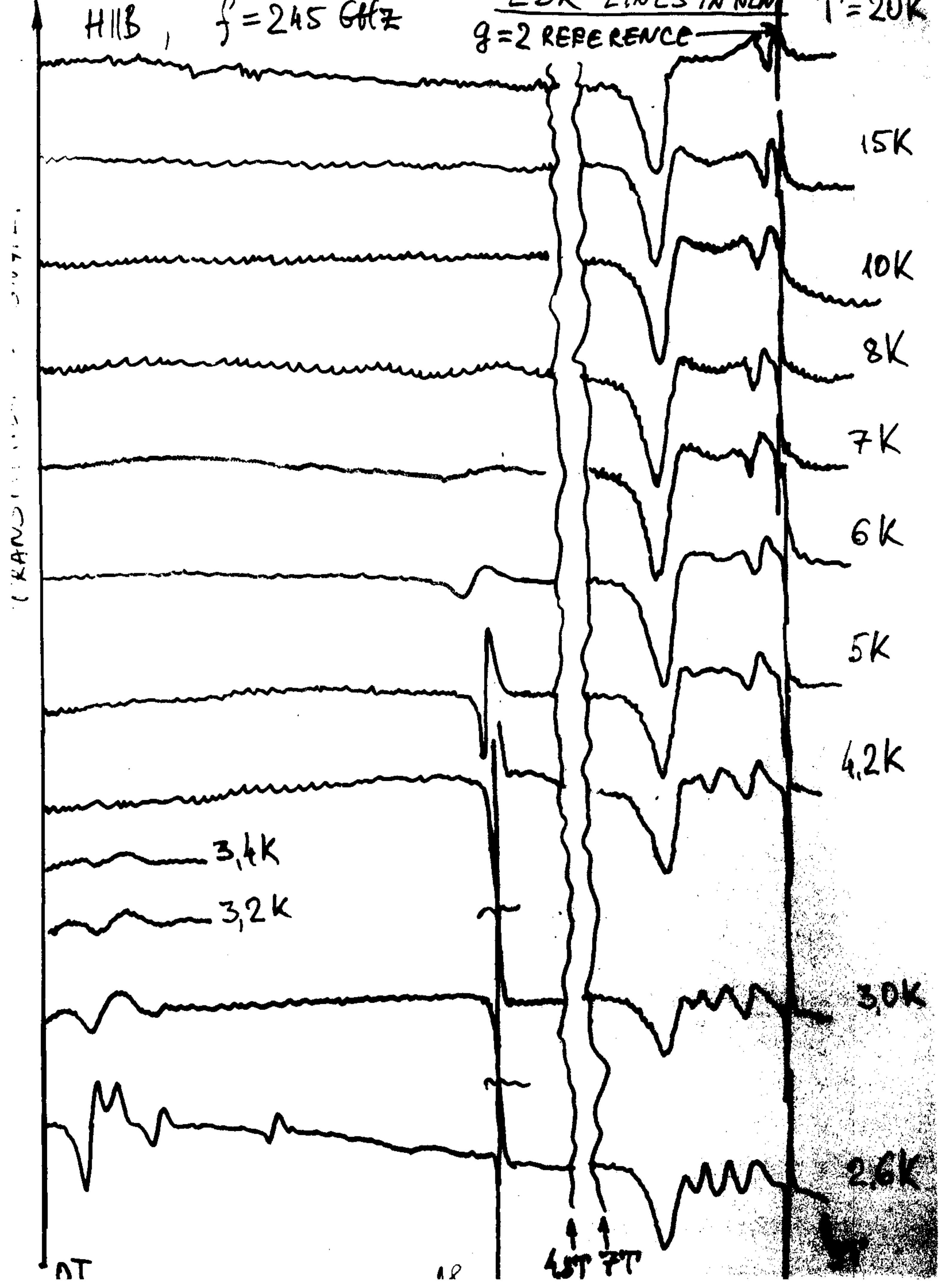


PARAMETERS.

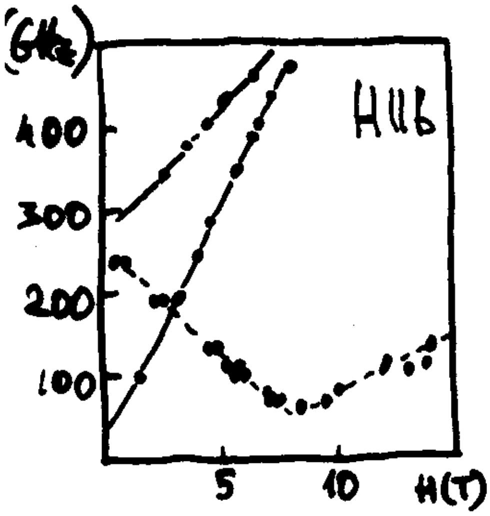

NENP C, 245 GHz
H||c
INTEGRAL INTENSITY, a.u.
TEMPERATURE, K


NENP C, 245 GHz
H||c
LINEWIDTH, T
TEMPERATURE, K


NENP C, 245 GHz
H||c
LINE POSITION, T
TEMPERATURE, K



H||B , f = 245 GHz
g = 2 REFERENCE
T = 20K
15K
10K
8K
7K
6K
5K
4,2K
3,4K
3,2K
3,0K
2,6K
7T



SAME RESULTS WERE OBTAINED BY B. LÜTHI'S GROUP

W. PALME ET. AL., INT. J. MOD. PHYS. B7, 1016 (1993)

GREEN POINTS DENOTING THE LINES APPEARING AT LOW T INCORPORATE DATA OF W. LU et. AL., Phys Rev. Lett. V.67, p. 3716 (1991)

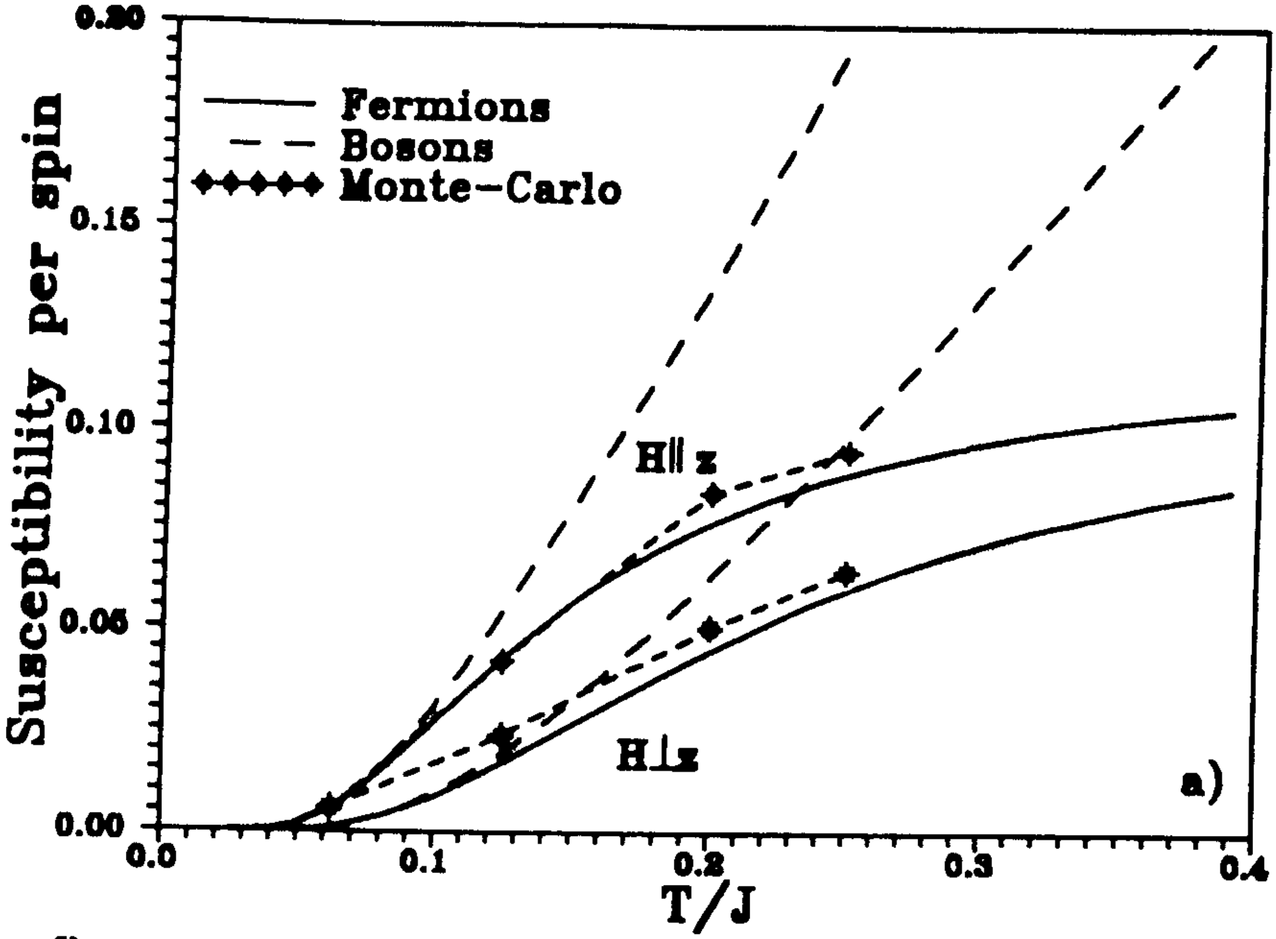
CONCLUSIONS.

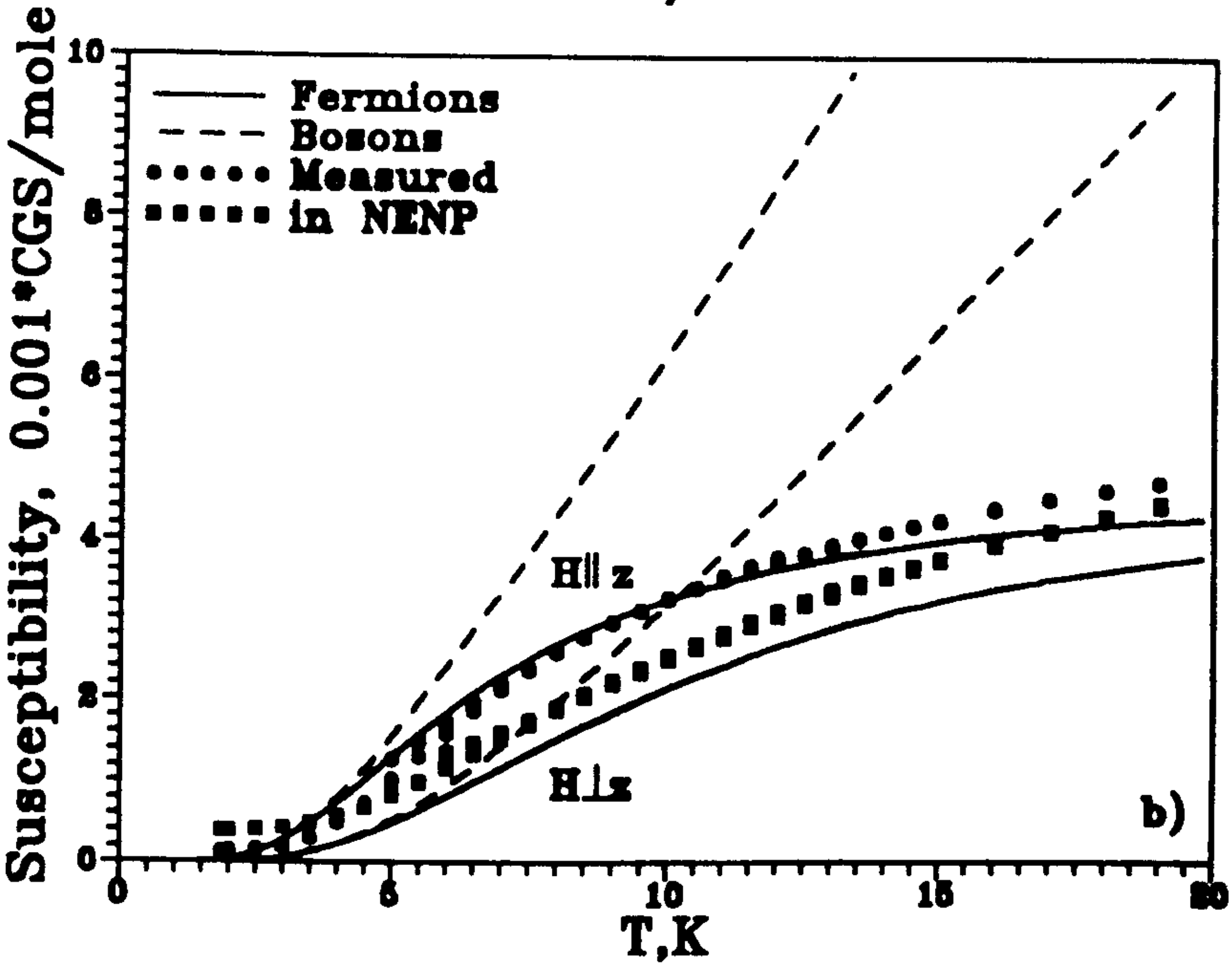
* MAGNETOSTATIC AND NEUTRON SCATTERING DATA DEMONSTRATE SINGLET GROUND STATE AND GAPPED EXCITATIONS IN QUASI-ONE-DIMENSIONAL ANTIFERROMAGNET NENP, AS PREDICTED BY HALDANE.

* ESR ABSORPTION DUE TO THE TRANSITIONS BETWEEN THE STATES OF THE EXCITED TRIPLET IS OBSERVED. IT DOMINATES χ'' OF THE SYSTEM AT FINITE TEMPERATURES, AS IS EXPECTED FOR SINGLET G.S. SYSTEMS.

* AT LOW TEMPERATURES MULTIPLE NEW ESR LINES APPEAR, PROBABLY CORRESPONDING TO THE TRANSITIONS FROM THE GROUND STATE
? COULD BE EXPLAINED BY THE STAGGERED [illegible]

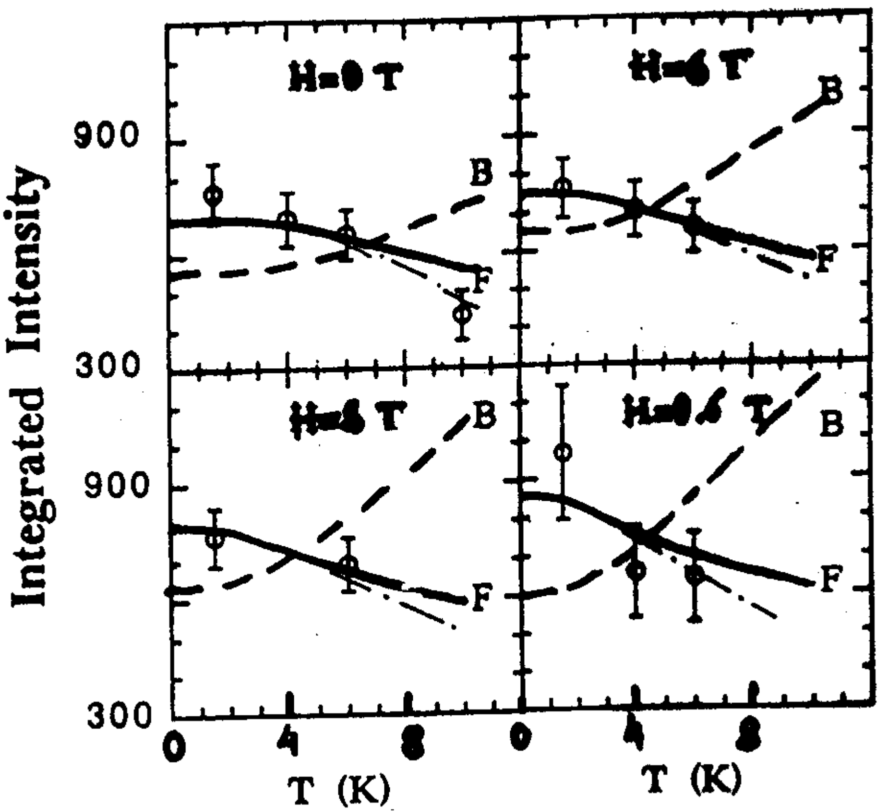
* ONE SHOULD BE VERY CAREFUL, SPEAKING [illegible] THE END'S OF CHAINS SPINS [illegible]


Fermions
Bosons
Monte-Carlo
Susceptibility per spin
H‖z
H⊥z
a)
T/J
0.00
0.05
0.10
0.15
0.20
0.0
0.1
0.2
0.3
0.4


Fermions
Bosons
Measured
in NENP
Susceptibility, 0.001*CGS/mole
H‖z
H⊥z
b)
T,K
0
2
4
6
8
10
0
5
10
15
20

TRACES OF THE TEMPERATURE DEPENDENCIES OF THE EXCITATION'S INTEGRATED INTENSITIES.

WHAT IS THE STATISTICS OF THE EXCITATIONS?



THE BEHAVIOR IS EXPECTED TO DEPEND ON THE STATISTICS OF THE EXCITATIONS, SIMPLY LIKE

$$I_q \sim \langle \hat{a}_q \hat{a}_q^+ \rangle_T \sim \begin{cases} 1 - n_F & \text{FOR FERMIONS} \\ 1 + n_b & \text{FOR BOSONS} \end{cases}$$

SO THE INCREASE IS EXPECTED FOR BOSONS AND THE DECREASE - FOR FERMIONS.

DEFINITIVE CONCLUSION IS NOT POSSIBLE DUE TO THE POSSIBLE TEMPERATURE DEPENDENT [illegible]

NEUTRON SCATTERING STUDY OF THE SPECTRAL DISTRIBUTION OF SPIN CORRELATIONS IN $CsNiCl_3$ ABOVE NEEL ORDERING.

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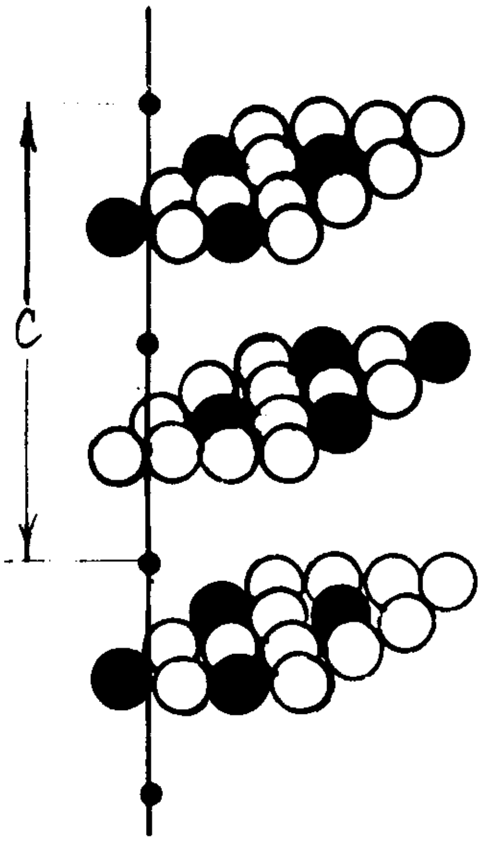
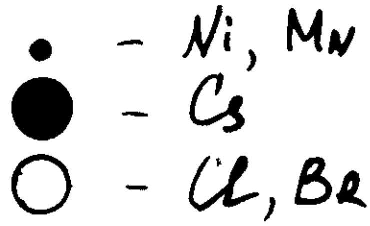
CONTENTS

QUASI-ONE-DIMENSIONAL HEXAGONAL ANTIFERROMAGNETS WITH 3D ORDERING: $CsNiCl_3$, $CsMnBr_3$





ONE-DIMENSIONALITY:

$J'/J \sim 10^{-2} \div 10^{-3}$

$CsNiCl_3$

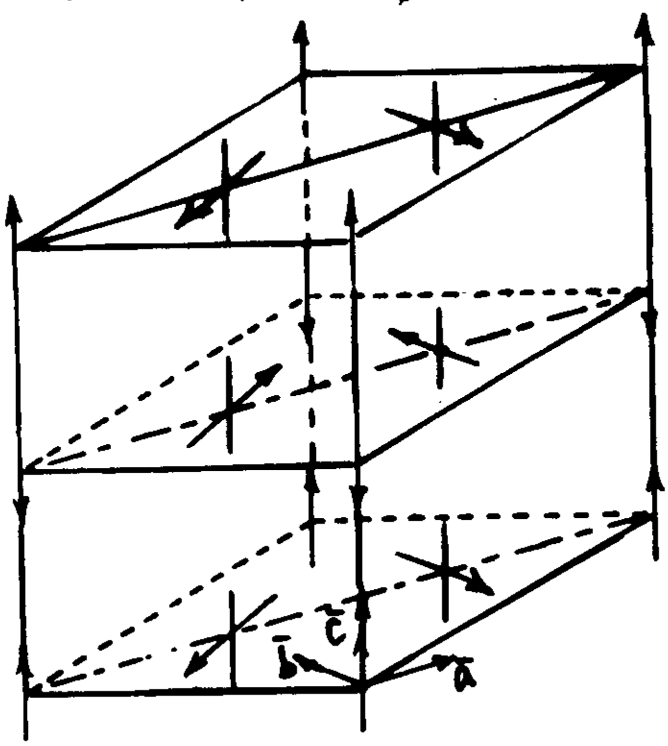
$J = 345$ GHz, $J' = 8$ GHz,

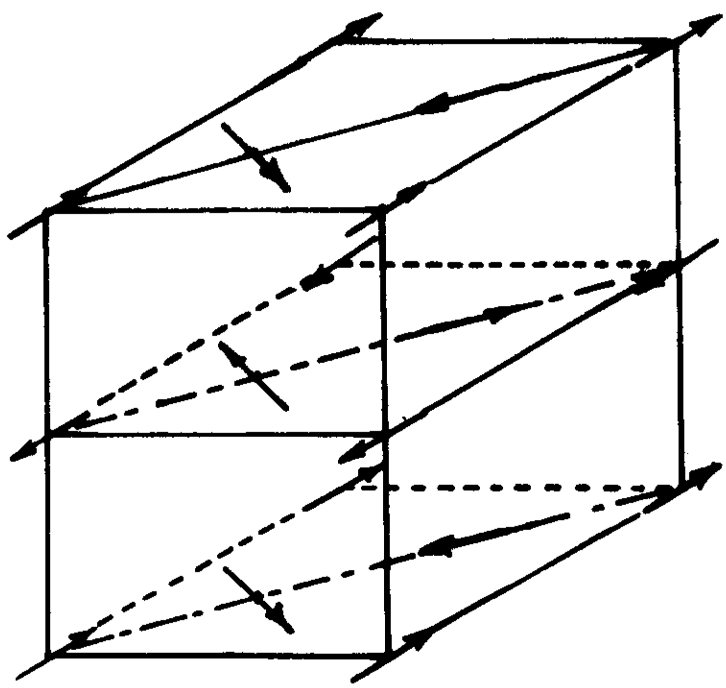
$D = -0{,}6$ GHz

$CsMnBr_3$

$J = 214$ GHz, $J' = 0{,}5$ GHz

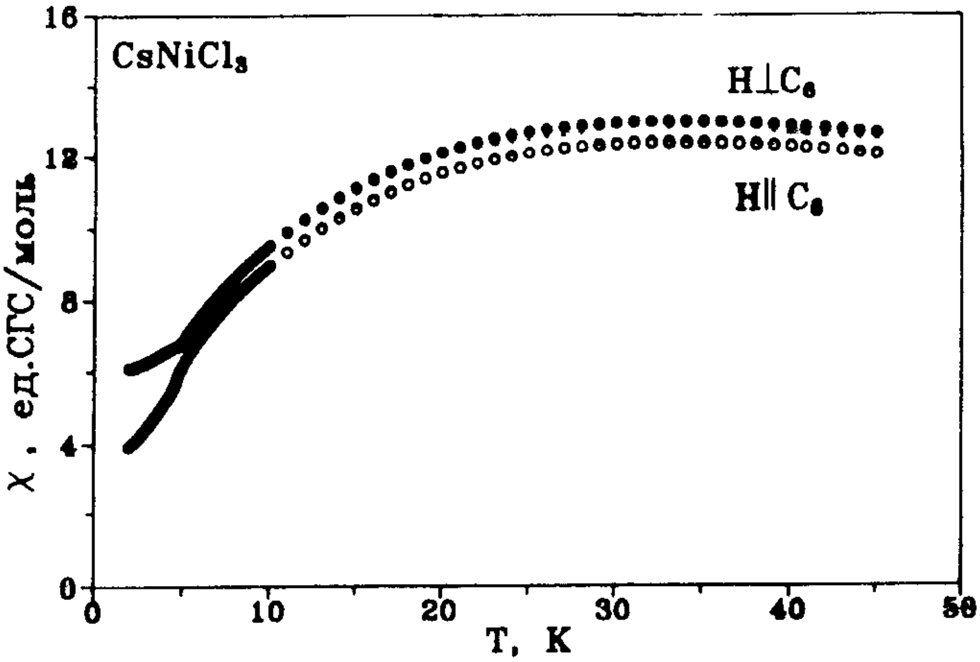
$D = +1{,}95$ GHz

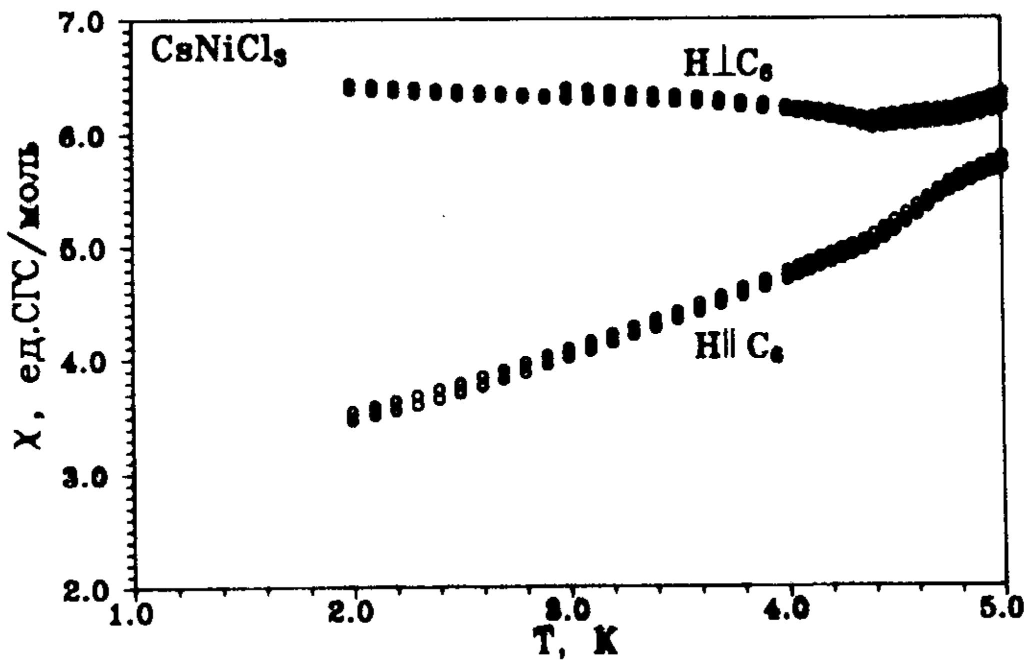




$T_{N_1} = 4.4$K, $T_{N_2} = 4.84$K

$T_N = 8{,}3$K


CsNiCl$_3$
H⊥C$_6$
H|| C$_6$
χ , ед.СГС/моль
T, K
16
12
8
4
0
0
10
20
30
40
50


CsNiCl$_3$
H⊥C$_6$
H|| C$_6$
χ , ед.СГС/моль
T, K
7.0
6.0
5.0
4.0
3.0
2.0
1.0
2.0
3.0
4.0
5.0

NEUTRON SCATTERING STUDY OF MAGNONS IN $CsNiCl_3$ WELL BELOW T_N

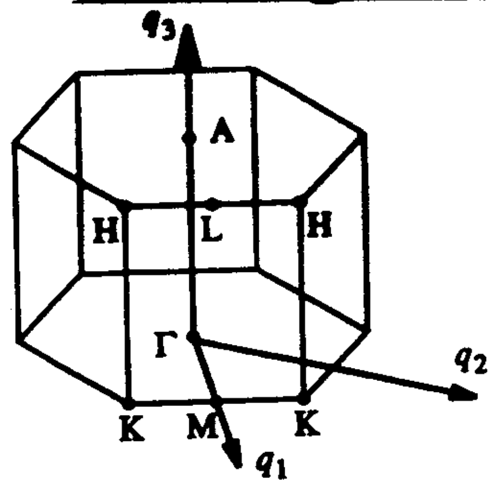


Fig. 1

* FIT OF THE LINEAR SWT TO THE MEASURED POINTS

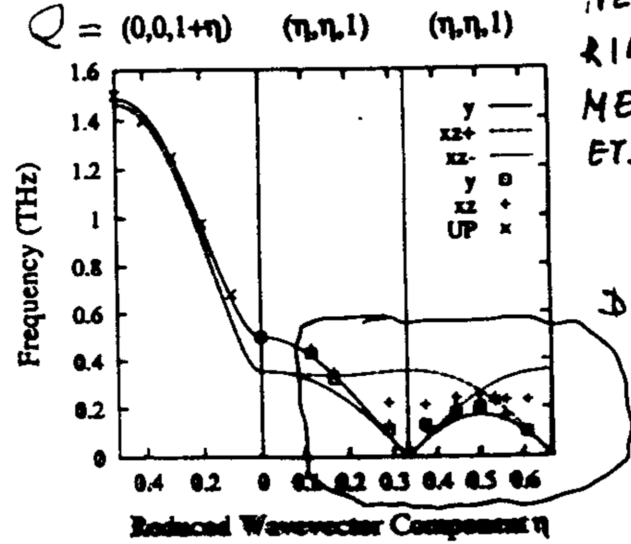


Fig. 3

NEUTRON SCATTERING MEASUREMENTS BY BUY... ET. AL., 1988

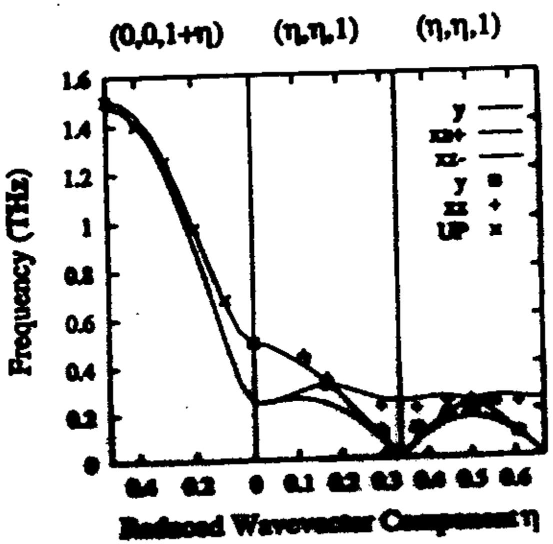


Fig. 2

* FIT OF THE CORRECTED DUE TO THE PRESENCE OF THE HIGH-ORDER TERMS SPIN-WAVE THEORY: GOOD AGREEMENT!

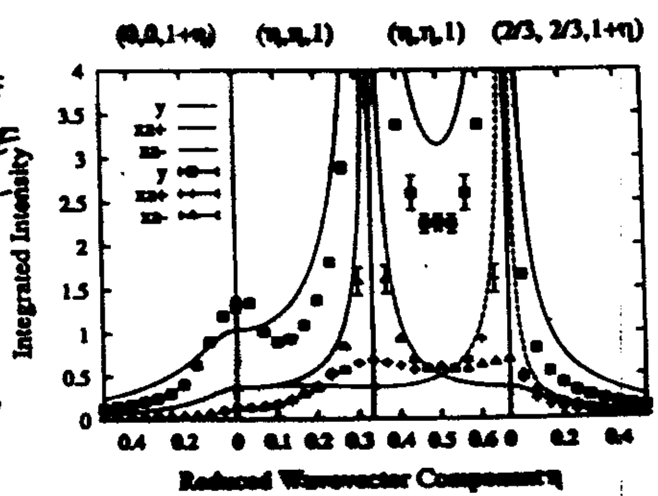
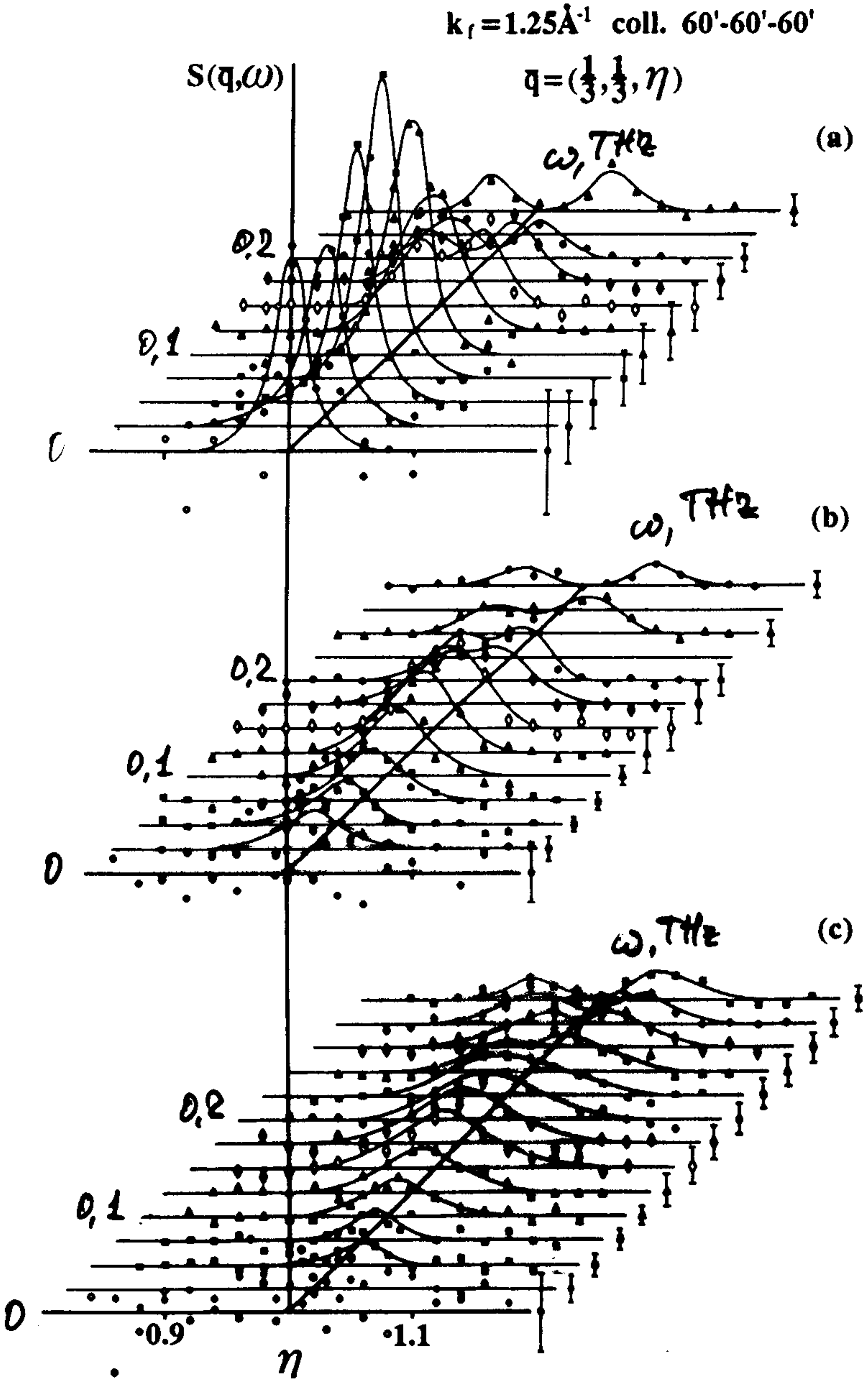
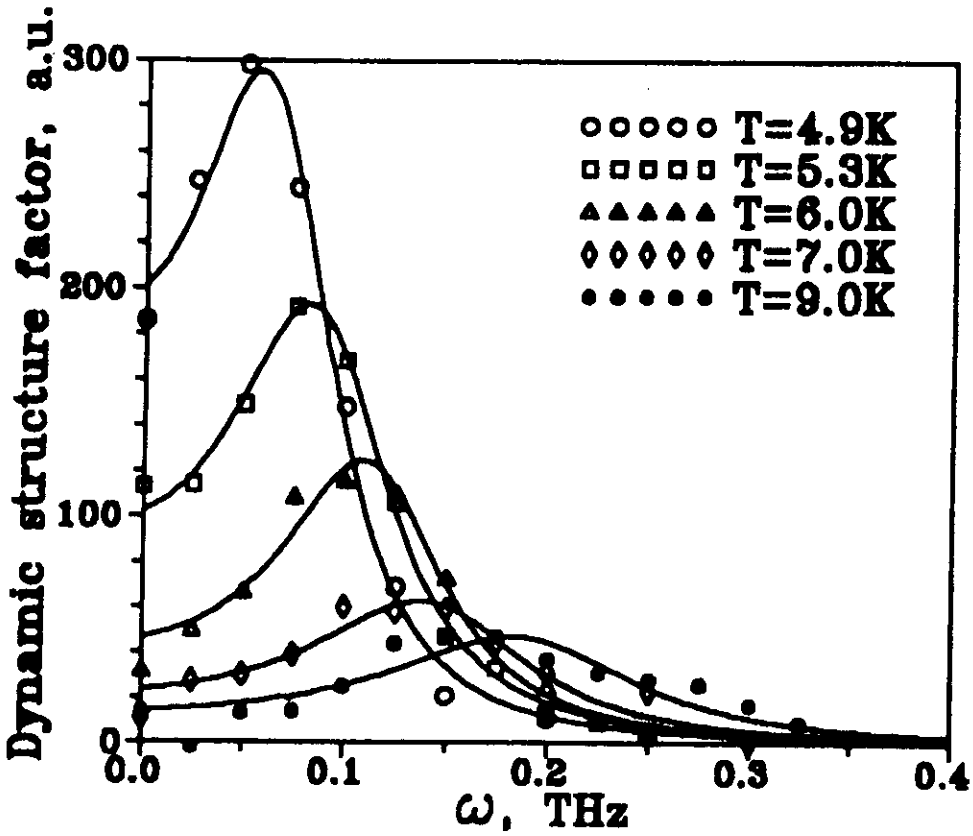


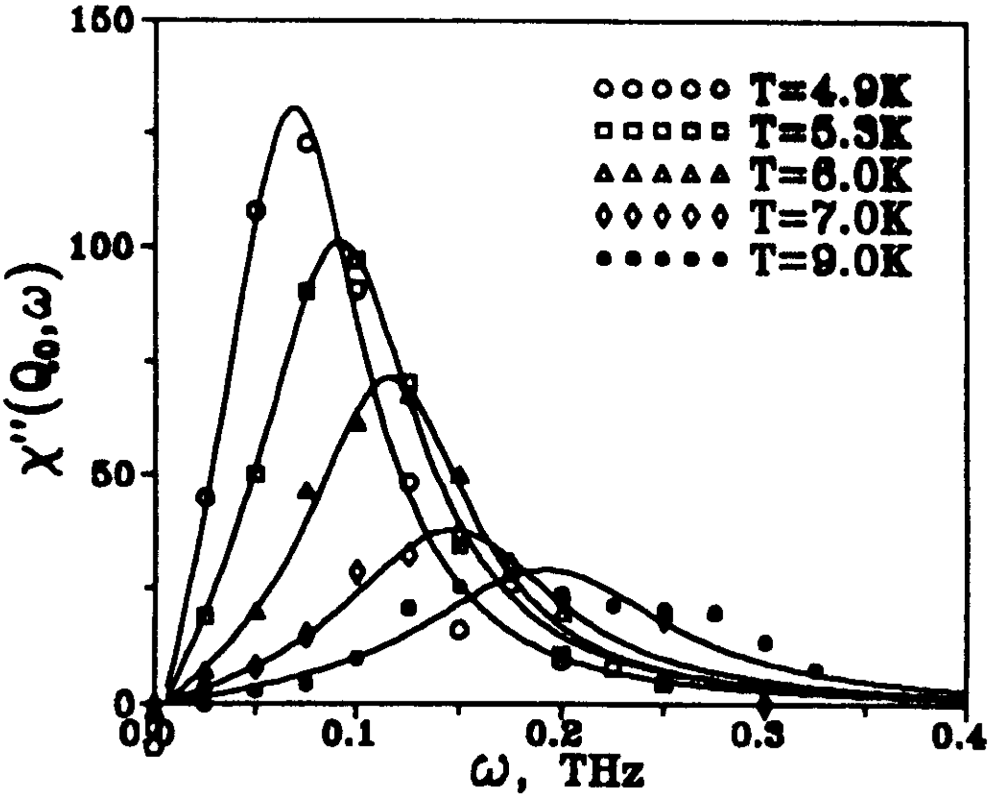
Fig. 4

T. OHYAMA, H. SHIBA, 1993, PREPRINT



k_f = 1.25Å^-1 coll. 60'-60'-60'
S(q,ω)
q=(1/3, 1/3, η)
(a)
ω, THz
0,2
0,1
0
(b)
ω, THz
0,2
0,1
0
(c)
ω, THz
0,2
0,1
0
0.9
1.1
η


Dynamic structure factor, a.u.
300
200
100
0
0.0
0.1
0.2
0.3
0.4
ω, THz
T=4.9K
T=5.3K
T=6.0K
T=7.0K
T=9.0K


χ''(Q₀,ω)
150
100
50
0
0.0
0.1
0.2
0.3
0.4
ω, THz
T=4.9K
T=5.3K
T=6.0K
T=7.0K
T=9.0K

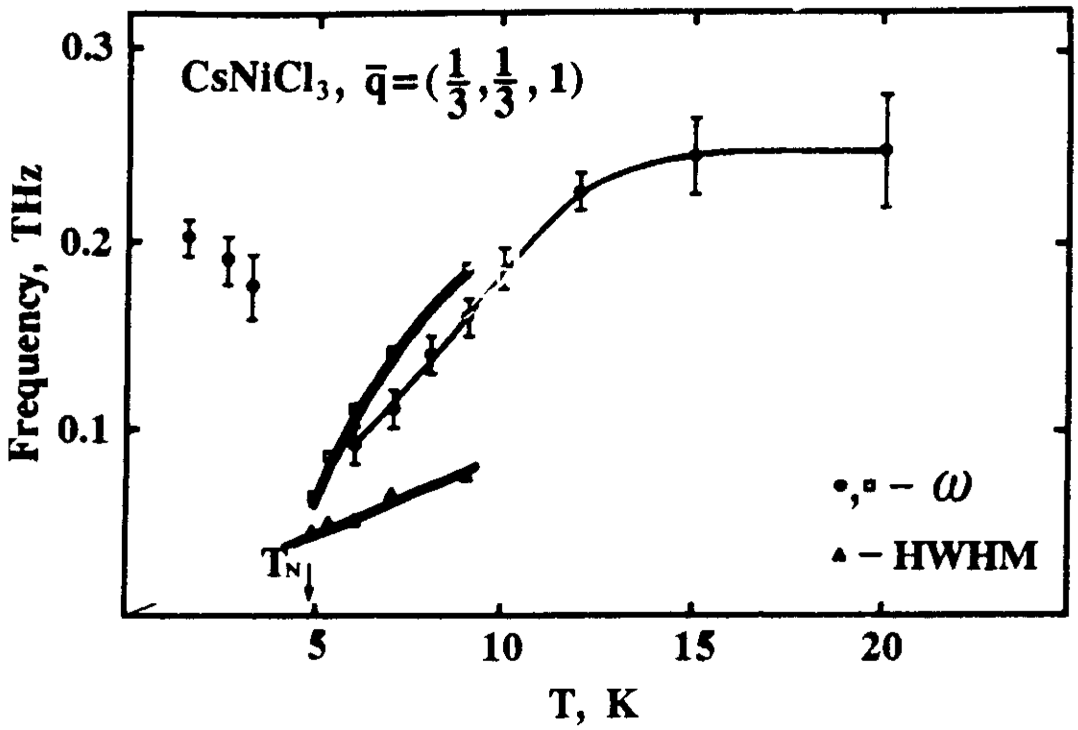


Table 1. Summary of the correlation lengths and excitations parameters measured in $CsNiCl_3$ at different temperatures above T_N. The variables used are defined in the text. Asterisk marks the peak parameters known with the poor accuracy from the E_N-scans with thermal neutrons.

Temperature, K	$\xi_\perp/a$ DN1	$\xi_\parallel/\frac{c}{2}$ DN1	$\xi_\parallel/\frac{c}{2}$ 4F2	$\xi_\parallel^{(qst)}/\frac{c}{2}$ 4F2	Peak frequency ω_q, THz	Peak width $2\Gamma_\omega$, THz
4.89 K	—	—	15.7	29.7	0.0626	0.0872
5.0 K	4.3	14.5	—	—	—	—
5.32K	—	—	12.95	24.5	0.0854	0.0974
6.0K	—	—	10.3	18.2	0.110	0.0982
7.0K	2.3	9.1	7.43	—	0.141	0.129
9.0K	1.9	7.9	4.9	—	0.186	0.149
11.0K	1.7	7.0	—	—	0.265*	0.218*

CONCLUSIONS:

* STUDY OF THE $S(q,\omega)$ SHAPES BY NEUTRON SCATTERING IN $S=1$ QUASI-ONE-DIMENSIONAL COMPOUNDS SUPPORT THE HALDANE CLAIM FOR A GAPPED SPECTRUM

* IN THE ANTIFERROMAGNETS WITH 3D ORDERING (LIKE $CsNiCl_3$) ONLY THE TEMPERATURE EVOLUTION OF $S(q,\omega)$ ABOVE THE NEEL TEMPERATURE CAN HELP IN CLARIFICATION OF HALDANE HYPOTHESIS.

* EXPERIMENTS WITH $S=\frac{1}{2}$ · INTEGER SPIN COMPOUNDS WITH 3D ORDERING ARE REQUIRED TO BE COMPARED TO THE OBTAINED DATA IN $CsNiCl_3$ FOR DEFINITIVE CONCLUSION ON THE DIFFERENCIES IN THEIR 1D SPIN DYNAMICS (ABOVE ORDERING).

* IT'S INTERESTING TO LOOK AT THE EVOLUTION OF THE HALDANE TRIPLET BELOW THE 3D ORDERING: WHERE THE LONGITUDINAL MODE GOES? IS IT OBSERVABLE?