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Theory of Relaxation Phenomena in Ising Antiferromagnets*

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A method combining statistical equilibrium theory and the thermodynamics of irreversible processes is used to study the magnetic relaxation behavior of *AB*-type Ising antiferromagnets; more specifically, using a Bethe-Takagi statistical expression for the magnetic Gibbs energy, the magnetic Gibbs energy produced in the irreversible process is calculated and the time derivatives of the sublattice long-range-order parameters and the short-range-order parameter are treated as fluxes conjugate to their appropriate generalized forces in the sense of Onsager's theory of irreversible thermodynamics. Three relaxation times are calculated and examined for temperatures near the Néel transition temperature T_N with the result that the "staggered" magnetization is shown to experience a critical "slowing down." Also, an expression is calculated for the dynamic initial parallel susceptibility which is appropriate within the above theoretical framework for all temperatures and frequencies and contains effectively only one phenomenological coefficient. Assuming the most simple temperature dependence for this phenomenological coefficient, the dynamic susceptibility expression is analyzed near T_N for selected frequencies.

I. INTRODUCTION

In recent years there have been large theoretical and experimental efforts devoted to the better understanding of critical phenomena in magnetic systems[1] and much progress has resulted. Largely due to the great complexity of an actual physical cooperative system, considerable theoretical interest has been directed towards finding and studying more simple mathematical models which nevertheless retain the decisive features of the physical system near a critical point. Even then, however, these simplified many-body models still present very formidable mathematical difficulties when an attempt is made to obtain their exact solutions and, as a result, many approximation methods have been studied and developed. In general, the case of nonequilibrium critical phenomena is not presently as well understood either theoretically or experimentally as the equilibrium case, and further efforts on these challenging time-dependent problems should promise to be rewarding in the future. Although admittedly there remains an important need for more fundamental understanding of dynamical aspects of critical phenomena, some noticeable developments which have used phenomenological arguments to some degree or another have emerged in the very last few years. In this regard, particular mention should be made for theories of dynamical scaling[2,3] and mode-mode coupling,[4–6] and for results found using the stochastic kinetic Ising model.[7]

The present paper considers an *AB*-type Ising model of an antiferromagnet. Then, utilizing the fact that the statistical Bethe-Takagi theory conveniently offers an approximate expression for the magnetic Gibbs energy surface, one next uses a simple method[8] which combines the statistical equilibrium theory with the Onsager theory of irreversible thermodynamics in order to study relaxation behavior in the spin system. Three relaxation times are calculated and examined for temperatures closely surrounding the Néel transition temperature T_N, and the "staggered" (or sublattice) magnetization is found to exhibit a critical "slowing down." Also an expression is calculated for the *dynamic* initial parallel susceptibility which is appropriate within this theoretical framework for all temperatures and frequencies and contains effectively only one phenomenological coefficient which may be found from experimental fit. Assuming the most simple temperature dependence for this phenomenological coefficient, the dynamic susceptibility expression is analyzed near T_N for selected frequencies.

II. BETHE-TAKAGI EQUILIBRIUM RESULTS FOR *AB*-TYPE ISING ANTIFERROMAGNETISM

The Ising model under study is an assembly of N spins localized on lattice points with the properties that each spin can only be up (positive) or down (negative) along the z axis and that the spin-spin interactions are short (nearest-neighbor) in range. Consider only lattices which may be decomposed into *two* interpenetrating sublattices A and B with the property that any lattice point belonging to a particular sublattice has *all* its nearest-neighbor lattice points on the other sublattice (such lattices may be termed "loosely packed"). For the case of Ising antiferromagnetism, call the A (B) sublattice the one which has spins up (down) at the absolute zero of temperature.

Introducing the localized Ising spin variable μ_i whose values are given by

$$\mu_i = \begin{cases} +1, & \text{spin on } i\text{th site up} \\ -1, & \text{spin on } i\text{th site down,} \end{cases}$$

one may define the following order parameters:

$$R_A \equiv \frac{2}{N} \sum_{k\in A} \mu_k, \quad R_B \equiv -\frac{2}{N} \sum_{l\in B} \mu_l,$$

$$\sigma \equiv -\frac{1}{Q} \sum_{\langle i,j\rangle} \mu_i \mu_j, \tag{1}$$

where N is the total number of Ising spins, $Q=\frac{1}{2}Nz$ is the total number of nearest-neighbor spin pairs (z, the lattice coordination number, being the number of nearest-neighbor spins surrounding any spin), and $\sum_{\langle i,j\rangle}\cdots$ signifies a summation over all nearest-neighbor spin pairs. R_A and R_B are called the *sublattice long-range-order* parameters while σ is called the *short-range-order* parameter.

Assuming the Ising spin system to be placed in an external magnetic field H applied along the z axis and to be in thermal contact with a temperature bath having absolute temperature T, one appropriately wishes to construct a statistical expression for the magnetic Gibbs energy $G_m \equiv U - TS - HM_{\text{tot}}$, where U, S, and M_{tot} are the spin-system configurational interaction energy, entropy, and total magnetic moment, respectively. The configurational interaction energy U and the total magnetic moment M_{tot} of the Ising antiferromagnetic spin system can easily be written, using (1), explicitly in terms of R_A, R_B, and σ as follows:

$$U = \frac{J}{2} \sum_{\langle i,j\rangle} \mu_i\mu_j = -\tfrac{1}{4} NzJ\sigma \quad (J>0), \tag{2}$$

$$M_{\text{tot}} = \mu_0 \sum_n \mu_n = \mu_0 \Big(\sum_{k\in A} \mu_k + \sum_{l\in B} \mu_l \Big) = \tfrac{1}{2} N\mu_0 (R_A - R_B), \tag{3}$$

where $\frac{1}{2}J$ is the strength of the nearest-neighbor Ising spin-spin interaction ($J>0$ for antiferromagnetism) and μ_0 is the magnetic moment of an Ising spin. In order to form the magnetic Gibbs energy G_m, however, one needs still to construct the statistical expression for the entropy given by Boltzmann's formula

$$S = k \ln W(R_A, R_B, \sigma), \tag{4}$$

where k is the Boltzmann constant and where $W(R_A, R_B, \sigma)$ is the number of ways of arranging the Ising spins consistent with given order-parameter values R_A, R_B, σ. As is well known, this purely combinatorial problem is an extremely difficult one (e.g., there exists at present no exact solution for any three-dimensional lattice) and therefore, at this stage, one must in general introduce some mathematical approximation for $W(R_A, R_B, \sigma)$. The present paper only considers the Takagi[9] approximation for $W(R_A, R_B, \sigma)$, which approximation is known to be the statistical-mechanical interpretation of the earlier Bethe[10] approximation. The results of the Bethe-Takagi approximation are well known and since they will be used in the present paper are entered below in a condensed form for convenience.

The magnetic Gibbs energy G_m in this approximation is given by

$$\begin{aligned} G_m &= U - TS - H\,M_{\text{tot}} \\ &= -\tfrac{1}{4}N\,z\,J\sigma - \tfrac{1}{2}N\,\mu_0\,H(R_A - R_B) - NkT\ln 2 \\ &\quad - \tfrac{1}{4}N\,kT(z-1)\,[(1+R_A)\ln(1+R_A) + (1-R_A)\ln(1-R_A) + (1+R_B)\ln(1+R_B) + (1-R_B)\ln(1-R_B)] \\ &\quad + \tfrac{1}{8}N\,kT\,z[(1+R_A-R_B-\sigma)\ln(1+R_A-R_B-\sigma) + (1-R_A+R_B-\sigma)\ln(1-R_A+R_B-\sigma) \\ &\qquad + (1+R_A+R_B+\sigma)\ln(1+R_A+R_B+\sigma) + (1-R_A-R_B+\sigma)\ln(1-R_A-R_B+\sigma)]. \end{aligned} \tag{5}$$

The equilibrium conditions $\delta G_m = 0$ result in the following set of coupled transcendental equations:

$$\frac{\partial G_m}{\partial R_A} = 0: \quad 2(z-1)\ln\frac{1+R_A}{1-R_B} = z\ln\frac{(1+R_A-R_B-\sigma)(1+R_A+R_B+\sigma)}{(1-R_A+R_B-\sigma)(1-R_A-R_B+\sigma)} - \frac{4\mu_0 H}{kT}, \tag{6a}$$

$$\frac{\partial G_m}{\partial R_B} = 0: \quad 2(z-1)\ln\frac{1+R_B}{1-R_B} = z\ln\frac{(1-R_A+R_B-\sigma)(1+R_A+R_B+\sigma)}{(1+R_A-R_B-\sigma)(1-R_A-R_B+\sigma)} + 4\,\frac{\mu_0 H}{kT}, \tag{6b}$$

$$\frac{\partial G_m}{\partial \sigma} = 0: \quad \ln\frac{(1+R_A-R_B-\sigma)(1-R_A+R_B-\sigma)}{(1+R_A+R_B+\sigma)(1-R_A-R_B+\sigma)} = -\frac{2J}{kT}. \tag{6c}$$

Setting $H=0$, the equilibrium conditions (6) offer the solutions

$$R_A=R_B=R_0, \quad \sigma=\sigma_0,$$

where R_0, σ_0 are the familiarly known solutions of the Bethe equations

$$2(z-1)\ln\frac{1+R_0}{1-R_0}=z\ln\frac{1+2R_0+\sigma_0}{1-2R_0+\sigma_0}\ , \tag{7a}$$

$$\ln\frac{(1-\sigma_0)^2}{(1+2R_0+\sigma_0)(1-2R_0+\sigma_0)}=-\frac{2J}{kT}\ . \tag{7b}$$

For temperatures *below* the Néel transition temperature T_N, these equilibrium order-parameter solutions R_0, σ_0 can both be written conveniently in terms of the so-called Bethe long-range-order parameter δ through the relations

$$R_0=\tanh z\delta, \tag{8a}$$

$$\sigma_0=1-2\,\frac{\sinh(z-2)\delta}{\sinh(2z-2)\delta\cosh z\delta}\ , \tag{8b}$$

where the temperature dependence of δ is given by

$$e^{-J/kT}=\frac{\sinh(z-2)\delta}{\sinh z\delta}\ . \tag{8c}$$

Using (8a) and (8c) together, the Néel transition temperature T_N marking the disappearance of equilibrium sublattice long-range order R_0 is found within this approximation as

$$J/kT_N=\ln[z(z-2)^{-1}]\ . \tag{8d}$$

For temperatures *above* the Néel transition temperature T_N, the equilibrium order-parameter solutions R_0, σ_0 are given, respectively, by

$$R_0=0, \tag{9a}$$

$$\sigma_0=\tanh(J/2kT). \tag{9b}$$

Considering the antiferromagnetic Ising spin system to be placed in a weak, uniform, static external magnetic field H applied along the z axis, one may write, in the state of thermodynamic equilibrium,

$$R_A=R_0+R_1\ , \tag{10a}$$

$$R_B=R_0-R_1\ , \tag{10b}$$

$$\sigma=\sigma_0+\sigma_1\ , \tag{10c}$$

where the induced quantities R_1, σ_1 are both small and vanish in the absence of H [one can show that $R_1\sim O(H)$ and $\sigma_1\sim O(H^2)$]. Substituting (10) into the equilibrium conditions (6a) and (6b) and using the Bethe equation (7a), one obtains

$$z\left(\frac{1}{1-\sigma_0}-\frac{(1-z^{-1})}{1-R_0^2}\right)R_1=\frac{\mu_0 H}{kT}+O(H^3). \tag{11}$$

Using (3), (10a), (10b), and (11), the magnetization M is found to be

$$M\equiv\frac{M_{\text{tot}}}{V}=\frac{N\mu_0}{2V}\ (R_A-R_B)=\frac{N\mu_0}{V}\ R_1$$

$$=\frac{N}{V}\ \frac{\mu_0^2}{zkT}\left(\frac{1}{1-\sigma_0}-\frac{(1-z^{-1})}{1-R_0^2}\right)^{-1}\ H+O(H^3), \tag{12}$$

which gives the static initial parallel susceptibility χ_{static} in the Bethe-Takagi approximation as

$$\chi_{\text{static}}=\frac{N}{V}\ \frac{\mu_0^2}{zkT}\left(\frac{1}{1-\sigma_0}-\frac{(1-z^{-1})}{1-R_0^2}\right)^{-1}\ . \tag{13}$$

The expression (13) is the same result as that furnished by the constant-coupling approximation[11] of Kasteleyn and Van Kranendonk applied to Ising antiferromagnetism.

Upon analyses, it is easy to show that the Bethe-Takagi (BT) expression for χ_{static} [(13)] has a finite-jump discontinuity in slope at the Néel transition temperature T_N, in comparison with the infinite-slope singularity found by Fisher and Sykes[12] (FS) using exact series expansions. More specifically, the FS results give, for temperatures in the neighborhood of $T_N^{(\text{FS})}$, the result that

$$\chi_{\text{static}}^{(\text{FS})}=\frac{N\mu_0^2}{kT}\left(\alpha_N+\epsilon_{\pm}[1-T_N^{(\text{FS})}/T]\ln\left|1-\frac{T_N^{(\text{FS})}}{T}\right|\right),$$

where α_N, $\epsilon_{\pm}$ are constants for given lattice coordination number z. For the hexagonal ($z=3$) lattice, however, the BT approximation gives qualitatively good agreement within the transition region when compared with the FS results in the sense that the BT results for the hexagonal lattice give a steep linear drop in the susceptibility as temperature is lowered slightly from T_N and, for temperatures slightly above T_N, a linear rise turning through a *maximum* at a temperature $T_{\max}$ *above* T_N (for $z=3$, $T_{\max}/T_N=1.208$, and $zJ\,\chi_{\text{static}}^{\max}/N\mu_0^2=0.4236$, while the FS results are $T_{\max}^{(\text{FS})}/T_N^{(\text{FS})}=1.688$ and $zJ\,\chi_{\text{static}}^{\max(\text{FS})}/N\mu_0^2=0.4163$). For larger lattice coordination numbers $z=4$, 6, and 8, the comparisons with FS are not as favorable since for these cases the susceptibility maxima in the BT approximation occur *at* T_N and the susceptibility curves become less steep at temperatures slightly below T_N for increasing lattice coordination number z. As one would expect, for all cases, both the higher location of the Néel transition temperatures T_N and the nature of the nonanalytic behavior at T_N exhibit the shortcomings of the BT approximation within the transition region. However, besides establishing a demarcation for the qualitative validity of the BT method applied to Ising antiferromagnetism, one might also remark, because of the fact the method only barely fails to give a maximum susceptibility above T_N for the plane square ($z=4$) lattice, that the results offer motivation for a slightly improved approximation or a better method, i.e., one which retains some three-spin correlations could be worthwhile.

III. MAGNETIC GIBBS-ENERGY PRODUCTION, KINETIC EQUATIONS, AND RELAXATION TIMES

In order to investigate relaxation phenomena in the Ising antiferromagnet, one assumes a small, uniform, external magnetic field H is applied along the z axis which removes the spin system slightly from equilibrium, and one studies how rapidly the spin system returns or relaxes back to thermal equilibrium. In order to calculate the dynamic initial parallel susceptibility, a method is used which combines the previous statistical equilibrium theory with the Onsager theory of irreversible thermodynamics. Since the external field H will always be taken sufficiently small such that the spin system may be considered close to equilibrium, this method only studies the final stage of approach to equilibrium. Before proceeding to discuss such longitudinal relaxation, some further preliminary remarks should be given concerning the method used. The Ising Hamiltonian

$$\mathcal{H}=2J\sum_{\langle j,k\rangle}S_{jz}S_{kz}, \tag{14}$$

where S_{lz} is the longitudinal component of the localized Ising spin ($S=\frac{1}{2}$) angular momentum operator S_l, does *not* admit longitudinal relaxation since the equation of motion for $\langle S_{lz}\rangle$ vanishes identically:

$$\frac{d\langle S_{lz}\rangle}{dt}=\frac{2J}{i\hbar}\sum_{\langle j,k\rangle}\langle[S_{jz}S_{kz},S_{lz}]\rangle=0, \tag{15}$$

where $\langle\cdots\rangle$ signifies the appropriate ensemble average, $2\pi\hbar$ is Planck's constant, and $[S_{jz}S_{kz},S_{lz}]$ signifies the vanishing commutator operator $S_{jz}S_{kz}S_{lz}-S_{lz}S_{jz}S_{kz}$. One therefore needs to add to the Ising Hamiltonian (14) other operator quantities which do not commute with S_{lz}, thereby inducing transitions within the spin system permitting, as a result, longitudinal relaxation. These added operator quantities should preferably contain some kind of spin-lattice coupling since it is known that longitudinal relaxation is intimately connected with the spin-lattice relaxation time, e.g., the frequently designated T_1 appearing in the longitudinal Bloch equation description of spin-lattice relaxation phenomena in solids. Since, as mentioned above, the method of the present paper contains the Onsager theory of irreversible processes (a phenomenological theory), the explicit form of the needed additional operators will not be displayed, but instead their effect will be "hidden" within Onsager phenomenological rate coefficients which themselves must be found either in principle by a more powerful theory or in practice by fit with experimental data.

To proceed, then, the magnetic Gibbs energy (5) is written in the neighborhood of equilibrium as

$$G_m(T,R_A,R_B,\sigma,H)=G_m^{(0)}(T,R_0,\sigma_0)+\Delta G_m, \tag{16}$$

where $G_m^{(0)}$ is the equilibrium magnetic Gibbs energy in the absence of H [found from (5) by setting $R_A=R_B=R_0$, $\sigma=\sigma_0$, $H=0$] and ΔG_m, the production of magnetic Gibbs energy due to the presence of small H, is given by the following quadratic form found by retaining to second order the terms in a Taylor series expansion of G_m with respect to the spontaneous equilibrium point $R_A=R_B=R_0$, $\sigma=\sigma_0$, $H=0$:

$$\begin{aligned}\Delta G_m=\tfrac{1}{2}[&A(R_A-R_0)^2+A(R_B-R_0)^2+2B(R_A-R_0)(R_B-R_0)\\&+2C(R_A-R_0)(\sigma-\sigma_0)+2C(R_B-R_0)(\sigma-\sigma_0)\\&+D(\sigma-\sigma_0)^2-2E(R_A-R_0)H+2E(R_B-R_0)H],\end{aligned} \tag{17}$$

where the expressions given below for A, B, C, D, and E are explicit functions of the known equilibrium quantities R_0, σ_0 and are, therefore, calculable as functions of the temperature:

$$\begin{aligned}A&=\left(\frac{\partial^2G_m}{\partial R_A^2}\right)_{\rm eq}=\left(\frac{\partial^2G_m}{\partial R_B^2}\right)_{\rm eq}\\&=\tfrac{1}{4}zNkT\left[\frac{1+\sigma_0}{(1+\sigma_0)^2-4R_0^2}+\frac{1}{1-\sigma_0}-\frac{2(1-z^{-1})}{1-R_0^2}\right],\end{aligned} \tag{18a}$$

$$B=\left(\frac{\partial^2G_m}{\partial R_A\partial R_B}\right)_{\rm eq}=\tfrac{1}{2}zNkT\,\frac{2R_0^2-\sigma_0(1+\sigma_0)}{(1-\sigma_0)[(1+\sigma_0)^2-4R_0^2]}, \tag{18b}$$

$$C=\left(\frac{\partial^2G_m}{\partial R_A\partial\sigma}\right)_{\rm eq}=\left(\frac{\partial^2G_m}{\partial R_B\partial\sigma}\right)_{\rm eq}=-\tfrac{1}{2}zNkT\,\frac{R_0}{(1+\sigma_0)^2-4R_0^2}, \tag{18c}$$

$$D=\left(\frac{\partial^2G_m}{\partial\sigma^2}\right)_{\rm eq}=\tfrac{1}{2}zNkT\,\frac{1+\sigma_0-2R_0^2}{(1-\sigma_0)[(1+\sigma_0)^2-4R_0^2]}, \tag{18d}$$

$$E=-\left(\frac{\partial^2G_m}{\partial R_A\partial H}\right)_{\rm eq}=\left(\frac{\partial^2G_m}{\partial R_B\partial H}\right)_{\rm eq}=\tfrac{1}{2}N\mu_0\,. \tag{18e}$$

According to the Onsager theory of irreversible thermodynamics, one obtains the generalized forces X_{R_A}, X_{R_B}, X_σ conjugate to the currents $\dot{R}_A$, $\dot{R}_B$, $\dot{\sigma}$, respectively, by differentiating ΔG_m [Eq. (17)] with respect to R_A-R_0, R_B-R_0, $\sigma-\sigma_0$, respectively:

$$\begin{aligned}X_{R_A}&=\frac{\partial(\Delta G_m)}{\partial(R_A-R_0)}\\&=A(R_A-R_0)+B(R_B-R_0)+C(\sigma-\sigma_0)-EH,\end{aligned} \tag{19a}$$

$$\begin{aligned}X_{R_B}&=\frac{\partial(\Delta G_m)}{\partial(R_B-R_0)}\\&=B(R_A-R_0)+A(R_B-R_0)+C(\sigma-\sigma_0)+EH,\end{aligned} \tag{19b}$$

$$X_\sigma=\frac{\partial(\Delta G_m)}{\partial(\sigma-\sigma_0)}=C(R_A-R_0)+C(R_B-R_0)+D(\sigma-\sigma_0). \tag{19c}$$

Again according to the Onsager theory of irreversible thermodynamics, the linear relations between the currents and forces may be written as follows in terms of a matrix of phenomenological rate coefficients:

$$\begin{pmatrix} \mathring{R}_A \\ \mathring{R}_B \\ \mathring{\sigma} \end{pmatrix} = \begin{pmatrix} \gamma_R & \nu & \xi \\ \nu & \gamma_R & \xi \\ -\xi & -\xi & \gamma_\sigma \end{pmatrix} \begin{pmatrix} X_{R_A} \\ X_{R_B} \\ X_\sigma \end{pmatrix} , \qquad (20)$$

where account has been taken of the geometrical equivalence of the two sublattices and the facts that the long-range-order parameters are odd variables while σ is an even variable under time inversion.[13] The matrix equation (20) written in component form yields upon using (19) a set of three, coupled, linear, inhomogeneous, first-order-in-time, rate equations:

$$\mathring{R}_A = (\gamma_R A + \nu B + \xi C)(R_A - R_0) + (\gamma_R B + \nu A + \xi C)(R_B - R_0) + [(\gamma_R + \nu)C + \xi D](\sigma - \sigma_0) - (\gamma_R - \nu)\, EH, \qquad (21a)$$

$$\mathring{R}_B = (\nu A + \gamma_R B + \xi C)(R_A - R_0) + (\nu B + \gamma_R A + \xi C)(R_B - R_0) + [(\gamma_R + \nu)C + \xi D](\sigma - \sigma_0) + (\gamma_R - \nu)\, EH, \qquad (21b)$$

$$\mathring{\sigma} = [\gamma_\sigma C - \xi(A+B)](R_A - R_0) + [\gamma_\sigma C - \xi(A+B)](R_B - R_0) + (\gamma_\sigma D - 2\xi C)(\sigma - \sigma_0). \qquad (21c)$$

In order to find the relaxation times, one considers the corresponding homogeneous equations resulting when there is no external stimulation, i.e., $H = 0$. Equations (21) then become

$$\mathring{R}_A = (\gamma_R A + \nu B + \xi C)(R_A - R_0) + (\gamma_R B + \nu A + \xi C)(R_B - R_0) + [(\gamma_R + \nu)C + \xi D](\sigma - \sigma_0), \qquad (22a)$$

$$\mathring{R}_B = (\nu A + \gamma_R B + \xi C)(R_A - R_0) + (\nu B + \gamma_R A + \xi C)(R_B - R_0) + [(\gamma_R + \nu)C + \xi D](\sigma - \sigma_0), \qquad (22b)$$

$$\mathring{\sigma} = [\gamma_\sigma C - \xi(A+B)](R_A - R_0) + [\gamma_\sigma C - \xi(A+B)](R_B - R_0) + (\gamma_\sigma D - 2\xi C)(\sigma - \sigma_0). \qquad (22c)$$

Assuming a solution of the form $e^{-t/\tau}$ for (22), one obtains the secular equation

$$\begin{vmatrix} \tau^{-1} + \gamma_R A + \nu B + \xi C & \gamma_R B + \nu A + \xi C & (\gamma_R + \nu)C + \xi D \\ \gamma_R B + \nu A + \xi C & \tau^{-1} + \gamma_R A + \nu B + \xi C & (\gamma_R + \nu)C + \xi D \\ \gamma_\sigma C - \xi(A+B) & \gamma_\sigma C - \xi(A+B) & \tau^{-1} + \gamma_\sigma D - 2\xi C \end{vmatrix} = 0, \qquad (23)$$

which yields the following three inverse relaxation times:

$$\tau_1^{-1} = -(\gamma_R - \nu)(A - B), \qquad (24a)$$

$$\tau_2^{-1} = -\tfrac{1}{2}[(\gamma_R + \nu)(A+B) + \gamma_\sigma D] \left\{ 1 - \left(1 - 4\,\frac{[\gamma_\sigma(\gamma_R + \nu) + 2\xi^2][D(A+B) - 2C^2]}{[(\gamma_R + \nu)(A+B) + \gamma_\sigma D]^2} \right)^{1/2} \right\}, \qquad (24b)$$

$$\tau_3^{-1} = -\tfrac{1}{2}[(\gamma_R + \nu)(A+B) + \gamma_\sigma D] \left\{ 1 + \left(1 - 4\,\frac{[\gamma_\sigma(\gamma_R + \nu) + 2\xi^2][D(A+B) - 2C^2]}{[(\gamma_R + \nu)(A+B) + \gamma_\sigma D]^2} \right)^{1/2} \right\}. \qquad (24c)$$

In order to analytically examine the spectrum (24) for temperatures *slightly below* T_N, one may use the following series expansions for (8a) and (8b):

$$R_0 = \tanh z\delta = z\delta - \tfrac{1}{3} z^3 \delta^3 + \cdots , \qquad (25a)$$

$$\sigma_0 = (z-1)^{-1}[1 + \tfrac{1}{3} z(z-2)(3z-2)\delta^2 + \cdots] , \qquad (25b)$$

where the relation between δ and the temperature slightly below T_N may be approximately written, using (8c), as

$$\tfrac{2}{3}(z-1)\,\delta^2 \simeq J\theta_- / k\, T_N , \quad \theta_- \equiv (T_N - T)/T_N . \qquad (25c)$$

Using (18) and the series expansions (25) for the order parameters, (24) may be written for temperatures T *slightly below* T_N as

$$\tau_1^{-1} = -\frac{NJ(z-1)(\gamma_R - \nu)}{(z-2)\ln[z(z-2)^{-1}]} \times \left[1 + \left(\frac{z^2 \ln[z(z-2)^{-1}]}{z-1} - 1 \right) \theta_- + \cdots \right], \qquad (26a)$$

$$\tau_2^{-1} = -\tfrac{1}{2} NJ(z-2)\gamma_\sigma^{-1}\, [\gamma_\sigma(\gamma_R + \nu) + 2\xi^2]\,\theta_- + \cdots , \qquad (26b)$$

$$\tau_3^{-1} = -\frac{NJ(z-1)^2 \gamma_\sigma}{2(z-2)\ln[z(z-2)^{-1}]} \left\{ 1 + \left[\left(\frac{z-2}{z-1} \right)^2 \times \left(\frac{(z-1)(3z^2 - 6z + 4)}{(z-2)^2} + \frac{3(z-1)\gamma_\sigma(\gamma_R + \nu) - 2\xi^2}{\gamma_\sigma^2} \right) \times \ln[z(z-2)^{-1}] - 1 \right] \theta_- + \cdots \right\} . \qquad (26c)$$

For temperatures *slightly above* T_N, one may use the following series expansion for (9):

$$R_0 = 0, \tag{27a}$$

$$\sigma_0 = \frac{1}{z-1}\left\{1 - \frac{z(z-2)\ln[z(z-2)^{-1}]}{2(z-1)}\,\theta_+ + \frac{z(z-2)\{2(z-1) - \ln[z(z-2)^{-1}]\}\ln[z(z-2)^{-1}]}{4(z-2)}\,\theta_+^2 + \cdots\right\}, \tag{27b}$$

where $\theta_+ \equiv (T - T_N)/T_N$. Using (18) and the series expansions (27), (24) may be written for temperatures *slightly above* T_N as

$$\tau_1^{-1} = -\frac{NJ(z-1)(\gamma_R - \nu)}{(z-2)\ln[z(z-2)^{-1}]} \times \left[1 - \left(\frac{z^2\ln[z(z-2)^{-1}]}{4(z-1)} - 1\right)\theta_+ + \cdots\right], \tag{28a}$$

$$\tau_2^{-1} = -\tfrac{1}{4}NJ(z-2)\gamma_\sigma^{-1}\,[\gamma_\sigma(\gamma_R+\nu) + 2\xi^2]\,\theta_+ + \cdots, \tag{28b}$$

$$\tau_3^{-1} = -\frac{NJ(z-1)^2\gamma_\sigma}{2(z-2)\ln[z(z-2)^{-1}]} \times \left\{1 - \left[\left(\frac{(z-1)\gamma_\sigma^2 + (z-2)^2\xi^2}{(z-1)^2\gamma_\sigma^2}\right) \times \ln[z(z-2)^{-1}] - 1\right]\theta_+ + \cdots\right\}. \tag{28c}$$

From (26) and (28), one concludes that, as $T \to T_N$ from either below or above,

$$\tau_1^{-1} \to -\frac{NJ(z-1)(\gamma_R - \nu)}{(z-2)\ln[z(z-2)^{-1}]}, \tag{29a}$$

$$\tau_2^{-1} \to 0, \tag{29b}$$

$$\tau_3^{-1} \to -\frac{NJ(z-1)^2\gamma_\sigma}{2(z-2)\ln[z(z-2)^{-1}]}, \tag{29c}$$

where, in (29), the Onsager coefficients are understood as evaluated at T_N. One therefore has the result that, as $T \to T_N$, the relaxation times τ_1 and τ_3 both remain finite while the remaining relaxation time τ_2 approaches infinity. Also, from physical considerations, (29) shows that

$$\gamma_R - \nu < 0, \quad \gamma_\sigma < 0. \tag{30}$$

Having found the system relaxation times, it is instructive to calculate next the normal coordinates associated with the negative reciprocals of these relaxation times. Diagonalizing the system of equations (22), one finds

$$\begin{pmatrix}\dot{Y}_1\\ \dot{Y}_2\\ \dot{Y}_3\end{pmatrix} = -\begin{pmatrix}\tau_1^{-1} & 0 & 0\\ 0 & \tau_2^{-1} & 0\\ 0 & 0 & \tau_3^{-1}\end{pmatrix}\begin{pmatrix}Y_1\\ Y_2\\ Y_3\end{pmatrix}, \tag{31a}$$

where the normal coordinates are calculated to be

$$Y_1 = (R_A - R_0) - (R_B - R_0), \tag{31b}$$

$$Y_2 = -\frac{[2\xi C - \gamma_\sigma D - \tau_2^{-1}][(R_A - R_0) + (R_B - R_0)] + 2[(\gamma_R + \nu)C + \xi D](\sigma - \sigma_0)}{\tau_2^{-1} + \tau_3^{-1} + 4\xi C}, \tag{31c}$$

$$Y_3 = \frac{[2\xi C - \gamma_\sigma D - \tau_3^{-1}][(R_A - R_0) + (R_B - R_0)] + 2[(\gamma_R + \nu)C + \xi D](\sigma - \sigma_0)}{\tau_2^{-1} + \tau_3^{-1} + 4\xi C}. \tag{31d}$$

Using (3) and (31b), the magnetization $M = M_{\text{tot}}/V$ may be written

$$M = \frac{N\mu_0}{2V}(R_A - R_B) = \frac{N\mu_0}{2V}Y_1, \tag{32a}$$

showing that the relaxation of the spin-system magnetization is characterized by the single finite relaxation time τ_1. Also, using (31c) and (31d), the system long-range-order parameter defined as $\frac{1}{2}(R_A + R_B)$, and the short-range-order parameter σ, may be written, respectively as

$$\tfrac{1}{2}(R_A + R_B) = R_0 + \tfrac{1}{2}\left(1 + \frac{\xi C}{\tau_2^{-1} + \tau_3^{-1}}\right)(Y_2 + Y_3) \tag{32b}$$

and

$$\sigma = \sigma_0 + \frac{1}{2}\,\frac{\tau_2^{-1} + \tau_3^{-1} + 4\xi C}{(\tau_2^{-1} - \tau_3^{-1})[(\gamma_R + \nu)C + \xi D]} \times \{[\tau_3^{-1} - (2\xi C - \gamma_\sigma D)]\,Y_2 + [\tau_2^{-1} - (2\xi C - \gamma_\sigma D)]Y_3\}. \tag{32c}$$

Equation (32b) shows that the relaxation of the long-range order $\frac{1}{2}(R_A + R_B)$ (proportional to the "staggered" or sublattice magnetization) is characterized by both relaxation times τ_2 and τ_3. However, for temperatures near T_N, Y_3 decays much more rapidly in time than Y_2 since τ_2 approaches infinity while τ_3 remains finite for such temperatures. One concludes therefore that the *staggered magnetization* experiences a *critical slowing down*. Equation (32c) shows that the return to equilibrium of the system short-range order is also characterized by both relaxation times τ_2 and τ_3. However, in this latter case, for temperatures near T_N, not only as mentioned above does Y_2 decay much more slowly in time than Y_3 but also for such temperatures, (32c) shows the amplitude of the normal coordinate Y_2 to be much smaller than the amplitude of the normal coordinate Y_3.

IV. STEADY SOLUTION OF KINETIC EQUATIONS LEADING TO COMPLEX MAGNETIC SUSCEPTIBILITY

The case is now studied where the spin system is stimulated by a small uniform external magnetic field H oscillating at an angular frequency ω. In the steady state, all quantities will oscillate at this same angular frequency ω; therefore,

$$H = H_1 e^{i\omega t}, \quad R_A - R_0 = R_{A1} e^{i\omega t},$$
$$R_B - R_0 = R_{B1} e^{i\omega t}, \quad \sigma - \sigma_0 = \sigma_1 e^{i\omega t}. \tag{33}$$

Substituting (33) into the kinetic equations (21) gives

$$(-i\omega + \gamma_R A + \nu B + \xi C)R_{A1} + (\gamma_R B + \nu A + \xi C)R_{B1} + [(\gamma_R + \nu)C + \xi D]\sigma_1 = (\gamma_R - \nu)\, EH_1, \tag{34a}$$

$$(\nu A + \gamma_R B + \xi C)R_{A1} + (-i\omega + \nu B + \gamma_R A + \xi C)R_{B1} + [(\gamma_R + \nu)C + \xi D]\sigma_1 = -(\gamma_R - \nu)\, EH_1, \tag{34b}$$

$$[\gamma_\sigma C - \xi(A+B)]R_{A1} + [\gamma_\sigma C - \xi(A+B)]R_{B1} + (-i\omega + \gamma_\sigma D - 2\xi C)\sigma_1 = 0. \tag{34c}$$

Solving the set of inhomogeneous equations (34) simultaneously for R_{A1}/H_1 and R_{B1}/H_1 gives

$$\frac{R_{A1}}{H_1} = -\frac{R_{B1}}{H_1} = \mathfrak{D}^{-1} \begin{vmatrix} (\gamma_R - \nu)E & \gamma_R B + \nu A + \xi C & (\gamma_R + \nu)C + \xi D \\ -(\gamma_R - \nu)E & -i\omega + \nu B + \gamma_R A + \xi C & (\gamma_R + \nu)C + \xi D \\ 0 & \gamma_\sigma C - \xi(A+B) & -i\omega + \gamma_\sigma D - 2\xi C \end{vmatrix}$$

$$= -\mathfrak{D}^{-1}(\gamma_R - \nu)E\{\omega^2 + i[(\gamma_R + \nu)(A+B) + \gamma_\sigma D]\omega - [\gamma_\sigma(\gamma_R + \nu) + 2\xi^2][D(A+B) - 2C^2]\}, \tag{35}$$

where the determinatal denominator $\mathfrak{D}$ is the same as the secular determinant used in (23) for calculating the reciprocal relaxation times except for the replacement of τ^{-1} by $-i\omega$, i.e.,

$$\mathfrak{D} \equiv \begin{vmatrix} -i\omega + \gamma_R A + \nu B + \xi C & \gamma_R B + \nu A + \xi C & (\gamma_R + \nu)C + \xi D \\ \nu A + \gamma_R B + \xi C & -i\omega + \nu B + \gamma_R A + \xi C & (\gamma_R + \nu)C + \xi D \\ \gamma_\sigma C - \xi(A+B) & \gamma_\sigma C - \xi(A+B) & -i\omega + \gamma_\sigma D - 2\xi C \end{vmatrix}$$

$$= (-i\omega - \tau_1^{-1})(-i\omega - \tau_2^{-1})(-i\omega - \tau_3^{-1}). \tag{36}$$

Equations (35) and (36) are needed in order to calculate the complex initial magnetic susceptibility $\chi^{(c)}(\omega)$. This may be seen as follows. The antiferromagnetic Ising-model induced magnetization (total induced magnetic moment per unit volume) is given by

$$M - M_\infty = \frac{N\mu_0}{2V}\mathrm{Re}[R_A - R_B]$$
$$= \frac{N\mu_0}{2V}\mathrm{Re}[(R_{A1} - R_{B1})e^{i\omega t}], \tag{37}$$

where M_∞ is the magnetization induced by a magnetic field oscillating at ∞ – frequency and where use was made of (33). Also, by definition, the expression for $\chi^{(c)}(\omega)$ may be written

$$M - M_\infty \equiv \mathrm{Re}[\chi^{(c)}(\omega) H_1 e^{i\omega t}], \tag{38}$$

where $\chi^{(c)}(\omega) = \chi'(\omega) - i\chi''(\omega)$ with $\chi'(\omega)$, $\chi''(\omega)$ being the magnetic dispersion and absorption factor, respectively. Comparing (37) and (38), one obtains

$$\chi^{(c)}(\omega) = (N\mu_0/2V)(R_{A1} - R_{B1})/H_1\,. \tag{39}$$

Therefore, in order to construct the dynamic susceptibility (39), one uses (35) and (36) in order to write

$$(R_{A1} - R_{B1})/H_1 = 2(\gamma_R - \nu)E\tau_1\tau_2\tau_3\, \frac{\omega^2 + i[(\gamma_R + \nu)(A+B) + \gamma_\sigma D]\omega - [\gamma_\sigma(\gamma_R + \nu) + 2\xi^2][D(A+B) - 2C^2]}{(1 + i\omega\tau_1)(1 + i\omega\tau_2)(1 + i\omega\tau_3)}\,. \tag{40}$$

Since (24) give both

$$[(\gamma_R + \nu)(A+B) + \gamma_\sigma D] = -(\tau_2^{-1} + \tau_3^{-1}) \tag{41a}$$

and

$$[\gamma_\sigma(\gamma_R + \nu) + 2\xi^2][D(A+B) - 2C^2] = \tau_2^{-1}\tau_3^{-1}\,, \tag{41b}$$

one is able to write (40) explicitly in terms of the relaxation times as

$$(R_{A1}-R_{B1})/H_1=N\mu_0(\gamma_R-\nu)\tau_1\tau_2\tau_3$$
$$\times\frac{\omega^2-i(\tau_2^{-1}+\tau_3^{-1})-\tau_2^{-1}\tau_3^{-1}}{(1+i\omega\tau_1)(1+i\omega\tau_2)(1+i\omega\tau_3)}$$
$$=-N\mu_0(\gamma_R-\nu)\,\frac{\tau_1}{1+i\omega\tau_1}\,. \tag{42}$$

Using (42), the complex initial susceptibility (39) becomes

$$\chi^{(c)}(\omega)=\chi'(\omega)-i\chi''(\omega)$$
$$=-\frac{(N\mu_0)^2}{2V}\,(\gamma_R-\nu)\,\frac{\tau_1}{1+i\omega\tau_1}\,, \tag{43}$$

a standard Debye form containing only the finite relaxation time τ_1 and containing effectively only one phenomenological coefficient $(\gamma_R-\nu)$. Within the present theoretical framework, the Debye form (43) is applicable for all values of temperature and frequency and, in particular, setting $\omega=0$ and using (24a), (18a), and (18b), one recovers the previous static susceptibility result [Eq. (13)] thus affording a consistency check upon the present dynamic calculation. Although not treated in the present paper, it should be mentioned that the staggered dynamic susceptibility can easily be displayed as containing a linear superposition (with temperature-dependent coefficients) of two Debye forms characterized by the remaining relaxation times τ_2 and τ_3, respectively.

Separating (43) into real and imaginary parts gives the magnetic dispersion and absorption factor as, respectively,

$$\chi'(\omega)-\chi_\infty=-\frac{(N\mu_0)^2}{2V}\,(\gamma_R-\nu)\,\frac{\tau_1}{1+\omega^2\tau_1^2}\,, \tag{44a}$$

$$\chi''(\omega)=-\frac{(N\mu_0)^2}{2V}\,(\gamma_R-\nu)\,\frac{\omega\tau_1^2}{1+\omega^2\tau_1^2}\,. \tag{44b}$$

Substituting the series expansions (26a) and (28a) into (44), the magnetic dispersion and absorption factor may be examined analytically for both low and high frequencies at temperatures closely surrounding the Néel transition temperatures T_N. Specifically, defining

$$\tau_{1N}\equiv-\frac{(z-2)\ln[z(z-2)^{-1}]}{NJ(z-1)(\gamma_R-\nu)}\,, \tag{45}$$

one obtains the following results for the *low-frequency region* characterized by $\omega\tau_{1N}\ll1$:

$$\chi'(\omega)-\chi_\infty=\begin{cases}K\left[1-\left(\dfrac{z^2\ln[z(z-2)^{-1}]}{z-1}-1\right)\theta_-+\cdots\right], & T<T_N \quad (46a)\\[2ex] K\left[1+\left(\dfrac{z^2\ln[z(z-2)^{-1}]}{4(z-1)}-1\right)\theta_++\cdots\right], & T>T_N \quad (46b)\end{cases}$$

$$\chi''(\omega)=\begin{cases}K\omega\tau_{1N}\left[1-2\left(\dfrac{z^2\ln[z(z-2)^{-1}]}{z-1}-1\right)\theta_-+\cdots\right], & T<T_N \quad (46c)\\[2ex] K\omega\tau_{1N}\left[1+2\left(\dfrac{z^2\ln[z(z-2)^{-1}]}{4(z-1)}-1\right)\theta_++\cdots\right], & T>T_N\,; \quad (46d)\end{cases}$$

while for the *high-frequency region* characterized by $\omega\tau_{1N}\gg1$, one obtains

$$\chi'(\omega)-\chi_\infty=\begin{cases}K(\omega\tau_{1N})^{-2}\left[1+\left(\dfrac{z^2\ln[z(z-2)^{-1}]}{z-1}-1\right)\theta_-+\cdots\right], & T<T_N \quad (47a)\\[2ex] K(\omega\tau_{1N})^{-2}\left[1-\left(\dfrac{z^2\ln[z(z-2)^{-1}]}{4(z-1)}-1\right)\theta_++\cdots\right], & T>T_N \quad (47b)\end{cases}$$

$$\chi''(\omega)=\begin{cases}K(\omega\tau_{1N})^{-1}\left[1-2(\omega\tau_{1N})^{-2}\left(\dfrac{z^2\ln[z(z-2)^{-1}]}{z-1}-1\right)\theta_-+\cdots\right], & T<T_N \quad (47c)\\[2ex] K(\omega\tau_{1N})^{-1}\left[1+2(\omega\tau_{1N})^{-2}\left(\dfrac{z^2\ln[z(z-2)^{-1}]}{4(z-1)}-1\right)\theta_++\cdots\right], & T>T_N \quad (47d)\end{cases}$$

where

$$K\equiv\frac{1}{2}\left(\frac{N}{V}\right)\frac{\mu_0^2}{J}\left(\frac{z-2}{z-1}\right)\ln[z(z-2)^{-1}],\ \theta_\pm=\pm\,\frac{T-T_N}{T_N}\,. \tag{48}$$

First, as expected, the $\chi'-\chi_\infty$ low frequency expressions (46a) and (46b) may also be easily obtained from the BT static susceptibility χ_{static} [(13)] by using the series expansions (25) and (27), respectively. Since the remaining dynamic susceptibility expressions (46c), (46d), and (47) contain (through τ_{1N}) the single Onsager phenomenological coefficient quantity $(\gamma_R - \nu)$, one should be able to fit such susceptibility expressions with experimental data in the transition region.

From an analytic point of view, one is able to extract some qualitative features from (46) and (47) if one *assumes* that the temperature variation of $(\gamma_R - \nu)$ in the transition region is negligible. Then (46a) and (47a) show that, for temperatures slightly beneath T_N and for $z = 3$, 4, 6, and 8, there exists a change in sign of the slope of the magnetic dispersion curve $\chi' - \chi_\infty$ vs temperature as one passes between the low- and the high-frequency regions (positive slope for the low-frequency region, negative slope for the high-frequency region). Also, for both the low- and high-frequency regions, the curve χ'' vs temperature has a positive slope at temperatures slightly beneath T_N, but for temperatures slightly above T_N, the χ'' curve has a positive slope for $z = 3$ and a negative slope for $z = 4$, 6, and 8. In addition one finds that, for the high-frequency region and for temperatures slightly above T_N, the curve $\chi' - \chi_\infty$ vs temperature has a negative slope for $z = 3$ but a positive slope for $z = 4$, 6, and 8. Therefore, under the initially mentioned assumption, it occurs once again that the qualitative features for $z = 3$ differ from those for $z = 4$, 6, and 8 crystal lattices. Concerning such differences, in this dynamical calculation the production of magnetic Gibbs energy ΔG_m was derived by power series from G_m and was crucial in order to apply the Onsager theory of irreversible thermodynamics. It is therefore desirable to choose the shape of the G_m surface as accurately as possible, i.e., to construct the best possible starting point for the application of the Onsager theory. Recalling, then, the earlier equilibrium Sec. II, and realizing that the general *qualitative* features of the equilibrium susceptibility derived by Fisher and Sykes from exact series expansions are rather similar for both two and three dimensions, it can be concluded by comparison with the FS exact series results that the Bethe-Tagaki approximation of the G_m surface is most accurate for the case of the hexagonal ($z = 3$) lattice. Therefore, in the present dynamical analyses, one should probably prefer the hexagonal lattice results whenever any ambiguity arises. For example, under the previous assumption that the temperature variation of $(\gamma_R - \nu)$ in the transition region is negligible, the stated dynamic results found above are taken to imply the existence of a maximum in the magnetic absorption factor curve χ'' vs temperature at temperatures above T_N for both the low- and high-frequency regions. Naturally, such assumed negligible temperature dependence for $(\gamma_R - \nu)$ should be tested either by experiment or by a more powerful theory. In this regard, Kikuchi[14] has shown by using a statistical-mechanical method called path probability that, for order-disorder configurational relaxation treated within a superposition approximation in a bcc *AB*-type lattice, the diagonal Onsager coefficients tend to finite values while the off-diagonal coefficient tends to zero as temperature tends to the critical temperature T_c (in fact, Kikuchi shows within the superposition approximation that the off-diagonal coefficient equals zero for $T > T_c$). From an experimental point of view, perhaps the substance $CoCs_3Cl_5$ can be used as an example of application since experimental measurements[15] suggest that this salt is, in a good approximation, a three-dimensional almost simple-cubic Ising antiferromagnet.

V. CONCLUSIONS

Using the method of equilibrium statistical mechanics, the static initial parallel susceptibility has been calculated in the BT approximation for loose-packed Ising antiferromagnets. These results have been compared with the results of FS based upon exact series expansions. For the hexagonal ($z = 3$) lattice, the BT results within the transition region gave qualitatively good agreement when compared against the FS results in the sense that one obtained a *maximum* in the susceptibility-vs-temperature curve at a temperature slightly *above* the Néel transition temperature T_N and one obtained a steep drop in the susceptibility curve as one lowered the temperature slightly from T_N. However, for the other lattice coordination numbers $z = 4$, 6, and 8, the qualitative agreement of BT compared with FS was not as good within the transition region since the BT results showed the susceptibility maxima to occur *at* T_N and showed the susceptibility curves to drop less steeply for increasing lattice coordination number z as one lowered temperature slightly from T_N.

Next a method combining the statistical equilibrium theory with the Onsager theory of irreversible thermodynamics was used to study relaxation behavior in the spin system and to calculate the dynamic initial parallel susceptibility. Although the results are expected to be rather inadequate for temperatures extremely close to a critical temperature due to the nature of the approximate theory used, the study illustrates a simple method whereby some qualitative features of the relaxation phenomena can be obtained in forms amenable to experimental fitting. Having used a three-order-parameter description, one obtained as a result three relaxation times where, as $T \rightarrow T_N$, one relaxation time tends to infinity while both remaining relaxation times stay finite. Besides finding a critical "slowing down" of the "staggered" magnetization, the dynamic susceptibility was found to be a standard Debye form containing a single (finite) relaxation time and containing effectively a single phenomenological coefficient which could be fitted with experiment. Assuming the most simple temperature dependence for this phenomenological coefficient, some qualitative conclusions were able to be drawn, which included a change in sign of the slope of the magnetic-dispersion–vs–temperature curve for temperatures slightly below T_N as one passed between so-called low- and high-frequency regions and the existence of a maximum in the magnetic-absorption-factor–vs–temperature above T_N for both the low- and high-frequency regions.

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Acoustical Faraday Effect in Antiferromagnetic Cr_2O_3[†]

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The rotation of the plane of polarization of an ultrasonic wave in both the antiferromagnetic and flopped phases of the antiferromagnetic crystal Cr_2O_3 has been calculated and measured at 4.2 °K using 9-GHz sound waves. The Faraday effect is found to arise from a resonant interaction between phonons and low-frequency magnon modes, by single-ion magnetostriction. The values of magneto-elastic coupling constants G_{44} and G_{46} of Cr^{3+} in Cr_2O_3 are determined: $|G_{44}| \propto 4.5\ \mathrm{cm}^{-1}$, $|G_{46}| \simeq 2.5\ \mathrm{cm}^{-1}$.

I. INTRODUCTION

Rotation of the plane of polarization of an acoustical transverse wave in a magnetically polarized crystal has been recognized, for a long time, to be a tool for the study of magneto-elastic coupling in magnetic materials. Kittel[1] was the first to draw attention to the possibility of a Faraday effect in ferromagnetic crystals. The first experimental results have been reported on yttrium iron garnet crystals and the values of the transverse magnetostriction were deduced at different temperatures.[2,3]

Similar experiments were done on paramagnetic crystals[4] and in metals.[5] In this last case, differ-