PHYSICAL REVIEW B VOLUME 35, NUMBER 13 1 MAY 1987

Resonant modes in Ising antiferromagnets at the critical magnetic field

G. G. Cabrera
Instituto de Física "Gleb Wataghin," Universidade Estadual de Campinas, 13081 Campinas, São Paulo, Brazil

Alzira M. Stein-Barana
Instituto de Física "Gleb Wataghin," Universidade Estadual de Campinas, 13081 Campinas, São Paulo, Brazil
and Instituto de Geociências e Ciências Exatas, Universidade Estadual Paulista,
*Campus Rio Claro, 13500 Rio Claro, São Paulo, Brazil**
(Received 21 November 1986)

Magnetic-resonance properties of Ising antiferromagnets near the critical field H_c are examined. It is found that H_c separates two regimes of completely different behavior. At H_c *one* line exhibits always vanishing amplitude. It is interesting and somewhat surprising, however, that the vanishing line is the dominant one when the field is varied from H_c in either direction. The vanishing line can be used in magnetic-resonance experiments to determine the critical field H_c.

INTRODUCTION

The Ising model continues to attract the attention of theorists more than forty years after Onsager's exact solution.[1] Recent interest is related to the equivalence between classical lattice models and Hamiltonian formulation of field theories,[2] thus broadening the domains where the Ising model is used.

Nonequilibrium properties of the model were first studied by Glauber[3] through the master-equation approach, where he assumes that the system interacts with its surroundings without explicitly identifying the interactions. It is known that master equations are obtained through a coarse-graining process during which some information is lost: Macroscopic irreversibility is then described through a probabilistic formulation.

A different approach, which is the one we follow here, introduces explicitly a particular dynamics in the Ising Hamiltonian.[4] For small fields, the linear response near equilibrium is given by the fluctuation-dissipation formula,[5] which is an exact result as long as the system is not significantly disturbed.

Transverse correlations for the Ising model have long been extensively studied.[6] However, the present authors have recently obtained some general results concerning the transverse dynamic susceptibility,[4] which show that resonance frequencies can be determined exactly (for ferromagnets and antiferromagnets) for arbitrary dimensions and coordination numbers. These resonance frequencies are expressed, in reduced units, as a set of small integers which are related to different configurations of nearest-neighbor clusters.[4] Intensities of modes as functions of temperature *cannot be solved in exact analytical form* in the general case, but qualitative information for ferromagnets can be drawn from the exact solution for the linear chain, and from approximate treatments of higher-dimensional systems.[4,7]

Concerning antiferromagnets, a calculation analogous to the one described in Ref. 4 shows novel features which we want to report here. The power spectrum clearly displays the presence of a *longitudinal critical field* H_c with striking effects on the intensities of lines (this critical field is equal to the maximum local field induced by nearest-neighbor coupling[8]).

The phenomenon we want to describe in this paper is twofold and based on very general grounds.

(i) When the longitudinal magnetic field is constant and the temperature is varied, two different behaviors can be distinguished, depending on whether the magnetic field is smaller or greater than the critical value H_c. This effect can be easily understood in terms of the competition between the spin-spin coupling and the spin alignment by the applied field. It has been previously noted in a particular calculation for the linear chain.[9]

(ii) For constant temperature and varying magnetic field, the dominant line in either regime shows *vanishing intensity at the critical field.* This particular line is associated with a cluster configuration where all the spins are parallel to the applied magnetic field. At the critical field there is a delicate balance between the exchange coupling and the applied field, so that individual spins can flip without extra energy.

The above-mentioned effects are universal for Ising antiferromagnets, independent of dimension or coordination number. They will be tested for simple cases in this Brief Report, which includes exact calculations for linear chains and approximate treatments for higher-coordination systems.

CALCULATION OF EXAMPLES

The geometry is the same as the one described in Ref. 4 (longitudinal field along the z axis and incident microwave radiation polarized in the x-y plane). For the sake of simplicity we have chosen lattice structures which can be separated into two interpenetrating equivalent sublattices α and β.[7] For fields smaller than the critical value and for low temperatures, two resonant lines show saturation to the same intensity value, while all the others decay to zero for $T \rightarrow 0$. Those two dominant lines at low temperature are related to antiferromagnetic ordering for small applied fields (or the tendency to antiferromagnetic ordering in the case of the linear chain), the flipped spin in the transition being located at different sublattice sites. In the high-field regime (H greater than H_c), the applied magnetic field polarizes both sublattices along the same direction, inhibiting the antiferromagnetic ordering. Concerning the line intensities, this latter case is qualitatively similar to that of ferromagnets discussed in Refs. 4 and 7.

Each resonant line and its corresponding resonance frequency are associated with a given cluster configuration.[4,7] For antiferromagnets, two elementary clusters with the same spin configuration but centered at different sublattice sites, contribute to the same line intensity.

A few changes in the notation of Ref. 4 are in order. The Ising Hamiltonian is now written as

$$\mathcal{H}_0 = -J \sum_{\langle\alpha,\beta\rangle} \sigma(\alpha)\sigma(\beta) - H_\alpha \sum_\alpha \sigma(\alpha) - H_\beta \sum_\beta \sigma(\beta) \ , \quad (1)$$

where J is the exchange constant (negative for antiferromagnets) and H_α and H_β are the Zeeman energies for the different sublattices. The symbol $\langle\alpha,\beta\rangle$ stands for the sum over all the nearest neighbors. In its general form, as written in (1), the Ising Hamiltonian also includes the so-called ferrimagnetic case where $H_\alpha \neq H_\beta$, allowing different magnetic moments for the species at the two sublattices.

The transverse dynamical susceptibility is obtained through the knowledge of the following Green's functions:[10]

$$G_{k,j}^{(\nu,\mu)}(\omega) \equiv \langle\langle \sigma_k^+(\nu); \sigma_j^-(\mu) \rangle\rangle_\omega \ , \quad (2)$$

where $\sigma_k^\pm(\nu)$ are the transverse spin components at lattice site k for the ν sublattice ($\nu=\alpha,\beta$). Except for these minor differences, the calculation is similar to the one described in Ref. 4.

Three resonant lines are present for the antiferromagnetic linear chain. Their intensities are shown in Fig. 1 as a function of the applied field for a particular value of temperature. All the figures are displayed in terms of the reduced variables

$$\tau = k_B T/|J| \ , \quad h = H/|J| \ , \quad (3)$$

where the temperature and the Zeeman energy are given in units of $|J|$, the exchange constant. In this case the critical field is simply given by the coordination number $h_c = 2$.

If we denote by $I(+)$ the intensity of the line where all the spins in the cluster of nearest neighbors are aligned in the direction of the applied field, we can see there a kink exactly at the critical field with vanishing intensity. This singularity marks the separation between the low- and high-field regimes. *In both situations $I(+)$ is always the dominant line.* At the critical condition, the local field induced by nearest-neighbor coupling—for this particular configuration—is exactly equal to the applied field, *thus costing no energy to flip the central spin.* We remark that the calculation for the linear chain is exact and can be put in closed analytical form.

When the magnetic moments associated with the different sublattices are not the same, each of the above resonant lines splits into two. For the linear chain this is an interesting case, since we get "antiferrimagnetic" ordering for fields below the critical value. The magnetization parameter for the β sublattice changes sign at the critical field, where β denotes the sublattice with the smallest magnetic moment. The critical condition is now

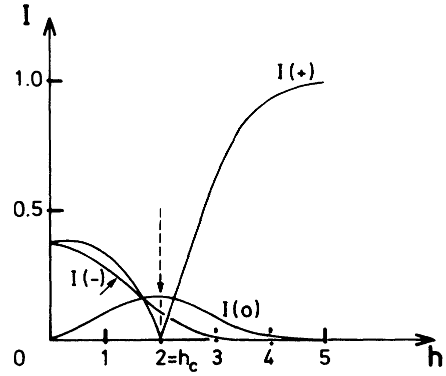

FIG. 1. Line intensities as function of the applied field for the antiferromagnetic linear chain. The graph is obtained from exact results for the one-dimensional Ising model at the reduced temperature $\tau = 1.0$. The critical field, in reduced units, is equal to the coordination number for the linear chain ($h_c = 2$). Resonance frequencies are determined following the procedure of Ref. 4, and the power absorption can be interpreted in terms of spin-flip transitions in different cluster configurations. In our notation + (−) labels the cluster where the two nearest-neighbor spins are parallel (antiparallel) to the applied field, and 0 stands for the case when one neighbor spin is parallel and the other antiparallel to the field. Note the maximum of $I(0)$ and the kink of $I(+)$ at the critical field. Intensities are given in arbitrary units.

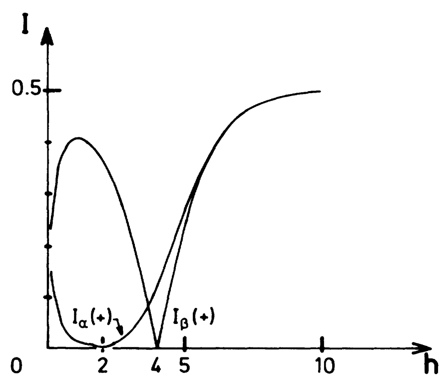

FIG. 2. The two lines split off from $I(+)$ when the magnetic moments associated with the sublattices are different. β always denotes the smallest moment ($\mu_\beta = 0.5$ in the figure). The critical field is now $h_c = 4$, as can also be seen in Fig. 3, with the inversion of the sublattice magnetization M_β. The $I_\beta(+)$ intensity is now associated with a ferromagnetic cluster centered at a β-magnetic moment with α spins as neighbors [and vice versa for the $I_\alpha(+)$ intensity]. No power absorption is present for $I_\alpha(+)$ at $h = 2$, but no kink is produced there. Temperature is $\tau = 1.0$.

given by

$$z\,|J| = \mu_\beta H_c \ , \tag{4}$$

where μ_β is a number ($\mu_\beta < 1$) related to the β magnetic moment. The two lines split from $I(+)$ are shown in Fig. 2 for $\mu_\beta = 0.5$ (and thus $h_c = 4$) for a particular value of the temperature. The corresponding order parameters are also shown in Fig. 3, along with the short-range order correlation η. More details are given in the figure captions.

For higher coordination numbers, and relying in part on approximate calculations, the following general predictions can be formulated.

(i) The appearance of more lines is expected, whose resonant frequencies are given by the scheme developed in Ref. 4.

(ii) The critical field is determined by the same physical argument given above in the text. It can be calculated using formula (4). For a given magnetic moment, h_c is an indication of both the exchange constant and the coordination number z.

(iii) The $I(+)$ intensity of the dominant line always vanishes and displays a kink at the critical field. It saturates in the limit $T \to 0$ for both the low- and high-field regimes. At $T=0$ we get

$$I(+, h > h_c) = 2I(+, h < h_c) \ .$$

(iv) In the low-field region there is another line saturating at low temperature. We will call it $I(-)$ and it is associated with the cluster where all the neighbor spins are antiparallel to the applied field. For $T=0$ this line saturates to the same value as $I(+)$. This case is illustrated in Fig. 4 for coordination number $z=6$ and $h=3$ (while the critical field in this case is $h_c=6$). In the high-field regime, $h > h_c$, $I(-)$ decays to zero at low temperatures.

(v) As in the case of the linear chain discussed here, the set of resonant lines suffers a splitting when $H_\alpha \neq H_\beta$, the splitting being

$$2\,|H_\alpha - H_\beta|/\hbar$$

in frequency units.

All the results presented here suggest that electron spin resonance, within the technical limitations imposed by the availability of frequency bands and magnetic fields, is a useful tool for characterizing antiferromagnetic exchange in Ising-like systems. The surprising behavior of the dominant line at the critical field can be used in turn to locate h_c in a typical experiment, yielding important information concerning the magnitude of the exchange and the coordination number.

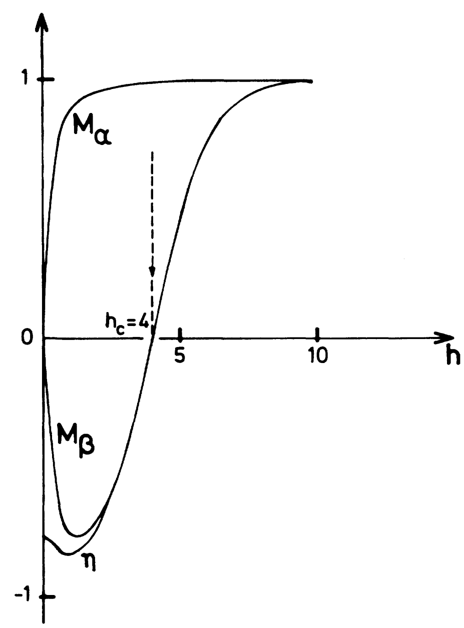

FIG. 3. The sublattice magnetizations M_α and M_β (order parameters) and the short-range order correlation η for the ferrimagnetic linear chain, as functions of the applied field. Exact calculations can be performed using the transfer matrix method described in Ref. 8. We show that "antiferrimagnetic ordering" is present below the critical field, where the M_β magnetization and the short-range order parameter are negative. Note that both $M_\alpha, M_\beta \to 0$ for $h=0$, indicating that ordering is not allowed in the absence of the field. This graph is also done for $\tau = 1.0$.

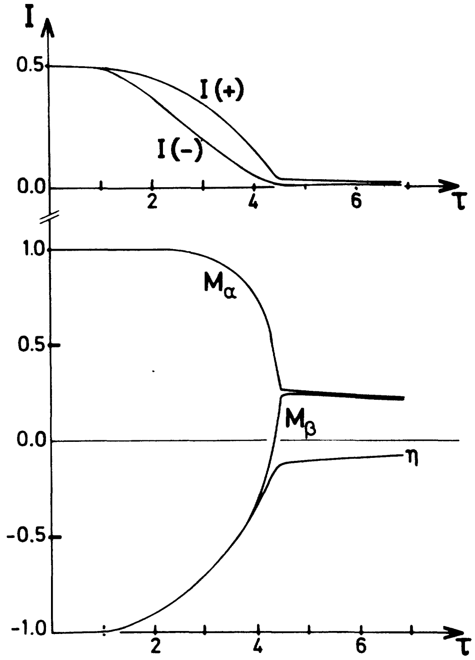

FIG. 4. Same graph for an antiferromagnetic system with coordination number $z=6$. Notation for the order parameters is the same as the one used in Fig. 3. The $I(+)$ and $I(-)$ intensities are given as function of temperature for a particular value of the field ($h=3.0 < h_c = 6.0$). This picture illustrates the low-field regime, with antiferromagnetic ordering below the critical temperature. Resonance lines displayed in the upper part of the figure are the only ones not vanishing at zero temperature. The calculation has been done using the Bethe-Peierls approximation (within this scheme the crystal structure is only taken into account through the coordination number).

ACKNOWLEDGMENTS

The authors are grateful to the Brazilian agencies Conselho Nacional de Desenvolvimento Científico e Tecnológico and Fundação de Amparo à Pesquisa do Estado de São Paulo for partial financial support. One of us (G.G.C.) wants to acknowledge the hospitality of the Solid State Physics Laboratory at Orsay, where part of the present calculation was done.

---

*Present address.

[1]L. Onsager, Phys. Rev. **65**, 117 (1944).

[2]See, for example, J. B. Kogut, Rev. Mod. Phys. **51**, 659 (1979).

[3]R. J. Glauber, J. Math. Phys. **4**, 294 (1963).

[4]G. G. Cabrera and Alzira M. Stein-Barana, Phys. Rev. B **29**, 433 (1984).

[5]R. Kubo, J. Phys. Soc. Jpn. **9**, 935 (1954).

[6]M. E. Fisher, J. Math. Phys. **4**, 124 (1963); G. A. T. Allan and D. D. Betts, Can. J. Phys. **46**, 15 (1968); A. K. Rajagopal and G. S. Grest, J. Math. Phys. **15**, 583 (1974); **15**, 589 (1974); E. J. van Dongen and H. W. Capel, Physica A **84**, 285 (1976); G. O. Berim and A. R. Kessel, Physica B **96**, 71 (1979).

[7]Alzira M. Stein-Barana, G. G. Cabrera, and L. M. Falicov, Phys. Rev. B **23**, 4391 (1981).

[8]C. Domb, Adv. Phys. **9**, 245 (1960).

[9]G. O. Berim, M. M. Zaripov, and A. R. Kessel, Zh. Eksp. Teor. Fiz. **66**, 734 (1974) [Sov. Phys. JETP **39**, 355 (1974)].

[10]D. N. Zubarev, Usp. Fiz. Nauk. **71**, 71 (1960) [Sov. Phys. Usp. **3**, 320 (1960)].