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**Autor:** Erdös, Paul

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## Dynamical Behavior of Exchange-Coupled Spins<sup>1)</sup>

by Paul Erdös

International Business Machines Corporation Research Laboratory, Zurich  
Säumerstrasse 4, 8803 Rüschlikon-ZH

(19. I. 66)

*Abstract.* The magnetic resonance conditions and dynamic susceptibilities of a two-spin system are derived. The spins are coupled through a combination of isotropic and asymmetric exchanges. This type of interaction occurs in weak ferromagnets.

A method is developed which permits the summation of the perturbative series for the wave function in the case of resonance.

### I. Exchange-Coupled Pairs of Spin

It has been observed [1]<sup>2)</sup> in solids doped with magnetic ions, that besides the well-known Heisenberg type isotropic exchange interaction

$$\mathcal{H}_0 = J \mathbf{S}_1 \cdot \mathbf{S}_2, \quad (I.1)$$

an asymmetric, or DZIALOSHINSKY [2]-MORIYA [3] type interaction

$$\mathcal{H}_1 = \mathbf{D} \cdot [\mathbf{S}_1 \times \mathbf{S}_2] \quad (I.2)$$

may occur. Whereas the former tends to align the two spins antiparallel (or parallel), the second tends to direct the spins perpendicular to the vector  $\mathbf{D}$  and upon one another. The direction of the vector  $\mathbf{D}$  is determined by a crystallographic axis of the host solid. In the case of a large antiferromagnetic coupling ( $J$  positive), a small  $\mathbf{D}$  has the effect of slightly canting the antiparallel spins by an angle  $\alpha$ , which is given by

$$\alpha \simeq \frac{D}{4J}, \quad (I.3)$$

where  $D$  is the modulus of  $\mathbf{D}$ .

<sup>1)</sup> This paper describes the mathematical treatment of the problem. The discussion of the physical aspects will be published elsewhere.

<sup>2)</sup> Numbers in brackets refer to References, page 163.

We shall study two spins  $S_1 = S_2 = 1/2$ , coupled by

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1 \quad (I.4)$$

and in a static external magnetic field  $H_z$  parallel to the direction of the vector  $\mathbf{D}$ , which is also taken as the  $z$ -direction. An additional alternating magnetic field will be introduced in the next section.

Since the square of the total spin  $S$  and its  $z$ -component  $M$  commute with  $\mathcal{H}_0$ , we take the four eigenstates  $|SM\rangle$  as a basis for future calculations, and label them with a single index  $\nu$ , according to

$S M \rightarrow \nu$		
0	0	1
1	0	2
1-1		3
1	1	4.

The Hamiltonian then takes the form

$$\mathcal{H} = \begin{bmatrix} 0 & \frac{i}{4}D & 0 & 0 \\ -\frac{i}{4}D & J & 0 & 0 \\ 0 & 0 & J + \mu H_z & 0 \\ 0 & 0 & 0 & J - \mu H_z \end{bmatrix} \quad (I.5)$$

with the eigenvalues

$$E_{1,2} = \frac{1}{2} J \left( 1 \mp \sqrt{1 + D^2/4 - J^2} \right) \quad E_{3,4} = J \pm \mu H_z. \quad (I.6)$$

Here  $\mu$  denotes the Bohr magneton (Fig. 1).

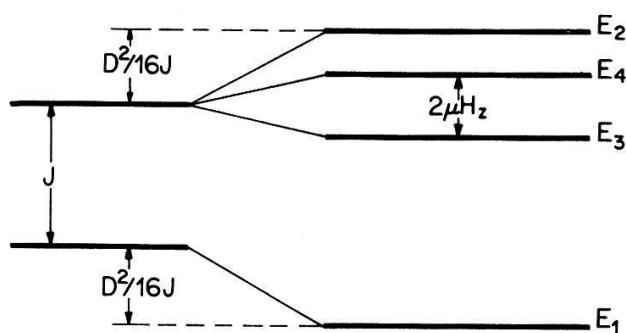


Figure 1

Splitting of the antiferromagnetic exchange doublet under the influence of the asymmetric exchange and the external magnetic field.  $J$  = isotropic exchange constant.  $D$  = asymmetric exchange constant.  $H_z$  = external magnetic field.

The exact eigenstates, whose energies are  $E_i$ , shall be denoted by  $|i\rangle$ ,  $i = 1, 2, 3, 4$ . They are obtained from the basis  $|\nu\rangle$  by the unitary transformation

$$U = \begin{bmatrix} \cos\alpha' & -i\sin\alpha' & 0 & 0 \\ -i\sin\alpha' & \cos\alpha' & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix} \quad (I.7)$$

with

$$\operatorname{tg}\alpha' = \frac{4E_1}{D}. \quad (I.8)$$

In first approximation

$$\operatorname{tg}\alpha' \simeq \frac{D}{4J} = \operatorname{tg}\alpha, \quad (I.9)$$

i.e., the angle  $\alpha'$  is the same as the canting angle defined by (I.3). Therefore, we shall drop the prime from  $\alpha$  bearing in mind that in the following, the exact relation (I.8) defines this angle.

The approximation  $\operatorname{tg}\alpha = D/4J$  is adequate when  $D/J \ll 1$ . However, a serious error would be introduced into the solution of the time-dependent problem by the replacement of  $\cos\alpha$  by 1, and  $\sin\alpha$  by  $\alpha$  in  $U$ ; this would destroy the unitarity of  $U$ , and non-periodic solutions of the Schrödinger-equation would appear.

## II. Dynamic Behavior of the Coupled Spin System

The *dynamic susceptibility* of the system determines its behavior in an alternating magnetic field, which varies with frequency  $\omega$ . *Magnetic resonance* occurs when the frequency of the field coincides with the energy difference divided by  $\hbar$ , between two levels of the system, and, in addition, the selection rules for the transition are fulfilled.

The time-dependent magnetic field is taken in the  $x$ -direction. It is sufficient to consider one Fourier-component  $H_\omega$  only, which contributes to the Hamiltonian the term

$$\mathcal{H}_3 = -\mu H_\omega \cos\omega t \mathbf{S}_x. \quad (II.1)$$

In the  $|\nu\rangle$ -representation,  $\mathbf{S}_x$  and  $\mathbf{S}_z$  are given by

$$\mathbf{S}_x = \frac{\sqrt{2}}{2} \begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 1 \\ 0 & 1 & 0 & 0 \\ 0 & 1 & 0 & 0 \end{bmatrix} \text{ and } \mathbf{S}_z = \begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix}. \quad (II.2)$$

[As can be seen by comparison with (I.5)]. In the  $|i\rangle$  representation we have

$$\mathcal{H}_3 = -\mu H_\omega \cos\omega t \mathbf{U}^{-1} \mathbf{S}_x \mathbf{U}. \quad (II.3)$$

To study the behavior of the system under the time-dependent perturbation  $\mathcal{H}_3$  we shall use the interaction representation, or *S*-matrix method, well-known in quantum field theory. Its usefulness in this context is based on the fact that the system has a relatively small number (4) of stationary states. This will permit us – in case of resonance – to sum the series of perturbation theory to all orders. Off resonance, the method will enable us to determine the linear and quadratic response of the system with ease. In contrast to this, the Kubo-formalism [4] and the method of rotating coordinate systems [5] only yield the linear response.

Let us denote the time-dependent wave functions of the unperturbed system by

$$\psi_i(t) = e^{-i\omega_i \cdot t} |i\rangle, \quad \text{with } \omega_i = \hbar^{-1} E_i, \quad i = 1, 2, 3, 4, \quad (\text{II.4})$$

and  $E_i$  given by (I.6). The wave function of the perturbed system will be written as

$$\psi(t) = \sum_i \Phi_i(t) \psi_i(t). \quad (\text{II.5})$$

The four coefficients  $\Phi_i(t)$  shall be condensed to a four-component wave function  $\Phi(t)$ , which will be called the wave function of the system in the *interaction representation*. Indeed, if  $\mathcal{H}_3$  vanishes,  $\Phi$  is constant. Inserting (II.4) and (II.5) into the time-dependent Schrödinger equation for  $\psi(t)$  yields,

$$i \hbar \frac{d\Phi}{dt} = \mathcal{H}_{int}(t) \Phi, \quad (\text{II.6})$$

where  $\mathcal{H}_{int}$  is the Hamiltonian in the interaction representation.

Its matrix elements are

$$(\mathcal{H}_{int})_{ij} = \langle i | \mathcal{H}_3(t) | j \rangle e^{-i\omega_j \cdot i t}, \quad (\text{II.7})$$

with

$$\omega_{j,i} = \omega_j - \omega_i, \quad (\text{II.8})$$

and  $\mathcal{H}_3(t)$  is given by (II.3).

The Equation (II.6) may now be integrated to yield

$$\Phi(t) = P \exp \left( - \frac{i}{\hbar} \int_{t_0}^t \mathcal{H}_{int}(t') dt' \right) \Phi^0. \quad (\text{II.9})$$

In this equation,  $P$  is the chronological operator which prescribes that operators at earlier times first act on  $\Phi^0$ . This prescription is very important here. The exponential which is obtained from (II.6) is really meant as the limit, as  $n$  tends to  $\infty$ , of the product of  $n$  operators which depend on different times and which differ only infinitesimally from unity [6]. Since  $\mathcal{H}_{int}(t_1)$  does not commute with  $\mathcal{H}_{int}(t_2)$ , in the following series expansion the time ordering has to be taken care of.

The initial conditions are specified by stating that the system be in its ground state  $\Phi^0 = (1, 0, 0, 0)$  at the time  $t = t_0$ .

The usefulness of the formal solution (II.9) will depend on whether we are able to evaluate the exponential operator multiplying  $\Phi^0$ . To do so, we first write out explicitly  $\mathcal{H}_{int}$ , using (II.7), and the matrices  $\mathbf{S}_x$  and  $\mathbf{U}$  as given by (II.2) and (I.7), respectively,

$$\mathcal{H}_{int}(t') = -\mu H_\omega \cos \omega t' \mathbf{Z}_x(t'), \quad (\text{II.10})$$

where

$$\mathbf{Z}_x = \frac{\sqrt{2}}{2} \begin{bmatrix} O & V \\ V^+ & O \end{bmatrix}, \text{ and } V = \begin{bmatrix} i \sin \alpha e^{-i\omega_{31}t'} & i \sin \alpha e^{-i\omega_{41}t'} \\ \cos \alpha e^{-i\omega_{32}t'} & \cos \alpha e^{-i\omega_{42}t'} \end{bmatrix}. \quad (\text{II.11})$$

In  $\mathbf{Z}_x$  the  $O$  denotes the two-by-two zero matrix. By virtue of its construction, the matrix  $\mathbf{Z}_x$  has the same algebraic property as  $\mathbf{S}_x$ ,  $\mathbf{S}_y$ , and  $\mathbf{S}_z$ , namely<sup>3)</sup>:

$$Z_x^3 = Z_x. \quad (\text{II.12})$$

Next, we develop the exponential operator occurring in (II.9) into a Taylor series

$$\Phi = P \{1 + i \hbar^{-1} \mu H_\omega \mathbf{A}(t) - (2! \hbar)^{-1} (\mu H_\omega)^2 \mathbf{A}^2(t) - + \dots\} \Phi^0 \quad (\text{II.13})$$

where

$$\mathbf{A}(t) = \int_{t_0}^t \cos \omega t' \mathbf{Z}_x(t') dt'. \quad (\text{II.14})$$

This is the perturbation series for the time-dependent wave function.

### III. Resonance

When the time integration is carried out in  $\mathbf{A}(t)$  (II.14), energy denominators of the form  $\omega_{ij} - \omega$  appear in each matrix element. If the frequency  $\omega$  of the perturbation is close to one of the transition frequencies  $\omega_{ij}$ , the corresponding matrix element becomes large, and one may neglect all other matrix elements in each term of the perturbation series (II.13).

For definiteness, suppose

$$\omega = \omega_{31} - \varepsilon, \text{ with } \varepsilon \ll \omega_{31}. \quad (\text{III.1})$$

As initial condition let us take  $\Phi^0 = \Phi(t=0)$ . From (II.14) we obtain

$$\mathbf{A}(t) = a \mathbf{Q} \quad \mathbf{A}^2(t) = a^2 \mathbf{Q}^2, \quad (\text{III.2})$$

where  $\mathbf{Q}$  and  $\mathbf{Q}^2$  are matrices whose only nonvanishing elements are

$$Q_{13} = Q_{31}^* = e^{i\varepsilon t/2} \text{ and } (Q^2)_{11} = (Q^2)_{33} = 1, \quad (\text{III.3})$$

and

$$a = -i \frac{\sqrt{2}}{2} \mu H_\omega \sin \alpha \varepsilon^{-1} \sin \left( \frac{1}{2} \varepsilon t \right). \quad (\text{III.4})$$

According to (II.13) we need the time-ordered powers of the operator  $\mathbf{A}(t)$ . For example:

$$\begin{aligned} P \mathbf{A}^2(t) &= \int_0^t dt_1 \int_0^t dt_2 P \{ \mathcal{H}_{int}(t_1) \mathcal{H}_{int}(t_2) \} = \int_0^t dt_1 \left\{ \int_0^{t_1} dt_2 \mathcal{H}_{int}(t_1) \mathcal{H}_{int}(t_2) \right. \\ &\quad \left. + \int_{t_1}^t dt_2 \mathcal{H}_{int}(t_2) \mathcal{H}_{int}(t_1) \right\} = \mathbf{A}^2(t) + \mathbf{C}(\cdot), \end{aligned} \quad (\text{III.5})$$

<sup>3)</sup> This property follows from the fact that every matrix fulfills its own minimal equation. Since the eigenvalues of the angular momentum operators in our representation are 0, 0, -1, and +1, the minimal equation is  $\lambda(\lambda+1)(\lambda-1) = 0$ .

where

$$\begin{aligned} \mathbf{C}(t) &= \int_0^t dt_1 [\mathcal{H}(0), \mathbf{A}(t_1)] \\ &= -\frac{1}{4} i \mu^2 H_\omega^2 \sin^2 \alpha \varepsilon^{-1} \left( t - \frac{1}{\varepsilon} \sin \varepsilon t \right) \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix}. \end{aligned} \quad (\text{III.6})$$

$\mathbf{C}(t)$  tends to zero as  $\varepsilon$ . In an analogous fashion

$$P \mathbf{A}^n(t) = \mathbf{A}^n(t) + O(\varepsilon). \quad (\text{III.7})$$

By virtue of the property (II.12),  $\mathbf{A}^3(t) \equiv \mathbf{A}(t)$ , and the series (II.13) may be summed up at exact resonance  $\varepsilon = 0$  to give

$$\begin{aligned} &= \left\{ 1 + i \sin \left( \frac{1}{4} \hbar^{-1} \mu H_\omega t \sin \alpha \right) \mathbf{Q} (\varepsilon = 0) \right. \\ &\quad \left. - \left[ 1 - \cos \left( \frac{1}{4} \hbar^{-1} \mu H_\omega t \sin \alpha \right) \right] \mathbf{Q}^2 (\varepsilon = 0) \right\} \Phi^0. \end{aligned} \quad (\text{III.8})$$

What has been done for  $\omega = \omega_{31}$ , also carries through for the other resonances. The general result being that for the resonance  $\omega = \omega_{jk}$ , the only nonvanishing matrix elements of  $\mathbf{Q}$  and  $\mathbf{Q}^2$  are

$$Q_{jk} = Q_{kj}^* = e^{-\varepsilon t/2} \text{ and } (Q^2)_{jj} = (Q^2)_{kk} = 1. \quad (\text{III.9})$$

For example, the wave function for exact resonance  $\omega - \omega_{31} = 0$  is

$$\begin{aligned} \Phi_1 &= \cos \left( \frac{1}{4} \hbar^{-1} \mu H_\omega t \sin \alpha \right), \\ \Phi_2 &= 0, \\ \Phi_3 &= i \sin \left( \frac{1}{4} \hbar^{-1} \mu H_\omega t \sin \alpha \right), \\ \Phi_4 &= 0. \end{aligned} \quad (\text{III.10})$$

The system oscillates between the states  $|1\rangle$  and  $|3\rangle$  with frequency  $1/4 \hbar^{-1} \mu H_\omega \sin \alpha$ . For a sufficiently weak exciting field  $H_\omega$  the transition from the initial state  $|1\rangle$  to the excited state is slow, and we define the transition rates as proportional to the squared modulus of the wave function in the excited state

$$W_{1 \rightarrow 3} = t^{-2} |\Phi_3|^2. \quad (\text{III.11})$$

Combining the results for all resonances, we obtain the following results for the transition rates:

$$\begin{aligned} \omega = \omega_{12}: \quad &W_{1 \rightarrow 2} = 0, \\ \omega = \omega_{34}: \quad &W_{3 \rightarrow 4} = 0, \\ \omega = \omega_{13}: \quad &W_{1 \rightarrow 3} = (16 \hbar)^{-1} \mu^2 H_\omega^2 \sin^2 \alpha \simeq \hbar^{-1} (\mu H_\omega D / 16 J^2), \\ \omega = \omega_{14}: \quad &W_{1 \rightarrow 4} = W_{1 \rightarrow 3}, \\ \omega = \omega_{23}: \quad &W_{2 \rightarrow 3} = (16 \hbar)^{-1} \mu^2 H_\omega^2 \cos^2 \alpha \simeq \hbar^{-1} (\mu H_\omega / 4)^2, \\ \omega = \omega_{24}: \quad &W_{2 \rightarrow 4} = W_{2 \rightarrow 3}. \end{aligned} \quad (\text{III.12})$$

The transitions between the levels  $2 \leftrightarrow 3$  and  $2 \leftrightarrow 4$  are ordinary Zeeman transitions and are only in fourth order dependent on  $D/J$ . The transitions  $1 \leftrightarrow 3$  and  $1 \leftrightarrow 4$  are proportional to  $D/J$ . Hence the ratio of certain transition intensities, such as

$$\frac{W_{3-1}}{W_{3-2}} = \operatorname{tg}^2 \alpha \simeq \left( \frac{D}{4J} \right)^2, \quad (\text{III.13})$$

is proportional to the ratio of the asymmetric to the isotropic exchange constants. Its observation requires the preparation of the system in an excited state.

The frequency of the above transitions can be tuned by varying the static magnetic field  $H_z$ .

It should be noted that the behavior near resonance, i. e., the line shape<sup>4)</sup> may be obtained by a somewhat more elaborate analysis of (V.17). Successive commutators of the type (VI.6) involve successively higher powers of the frequency mismatch  $\epsilon$ . Keeping only the first nonvanishing terms in  $\epsilon$ , one may derive a formula similar to (VI.8) which contains the information necessary for the study of the line shape.

#### IV. Dynamic Susceptibility. Linear and Quadratic Response

When the frequency  $\omega$  of the external magnetic field is not close to any of the transition frequencies  $\omega_{ij}$ , the matrices  $A^n(t)$  which determine the time-dependent wave function contain large energy denominators, and the series (II.13) converges rapidly. The *linear response* of the system is obtained, if in calculating the expectation values of different quantities we keep only terms linear in  $H_\omega$ . *Quadratic* and higher order responses are obtained correspondingly.

The exact expression for the expectation value of the magnetic moment  $\mathbf{M}(t)$  is

$$\mathbf{M}(t) = \mu \langle \Phi(t) | \mathbf{S}(t) | \Phi(t) \rangle, \quad (\text{IV.1})$$

where the operator  $\mathbf{S}(t)$  and the wave function  $\Phi(t)$  are in the interaction representation.

For linear response, we represent  $\Phi(t)$  merely by the first two terms in (II.13), and obtain

$$\mathbf{M}(t) = i \hbar^{-1} \mu^2 H_\omega \langle \Phi^0 | \mathbf{S}(t) \mathbf{A}(t) - \mathbf{A}(t) \mathbf{S}(t) | \Phi^0 \rangle. \quad (\text{IV.2})$$

At this point, a remark on the initial conditions is in order.

In the case of resonance, we specified the initial wave function  $\Phi^0$  as the unperturbed ground state at  $t = 0$ , and the external field acted indefinitely. This was correct, because the system passes through its unperturbed ground state any number of times under the action of the perturbation. Hence, without loss of generality,  $t = 0$  could be assigned to one such passage.

Far from resonance the situation is different: If before the action of the perturbation, the system were in its ground state, it might never pass through it again. Hence, we assume  $\Phi^0$  to be the unperturbed ground state at  $t = -\infty$ , and multiply the interaction Hamiltonian (II.11) by  $e^{st}$ , where  $s$  is a small positive number. This corresponds to an adiabatic switching on of the perturbation<sup>5)</sup>. It starts with zero at

<sup>4)</sup> This may be called the 'natural line shape' of the resonance, in contrast to the line shape due to relaxation processes.

<sup>5)</sup> Some authors prefer to introduce the factor  $e^{st}$  by adding a small imaginary part to the frequency. That procedure is less illuminating, but equally effective.

$t = -\infty$  and reaches its full strength at  $t = 0$ . The expectation values thus calculated are made independent of  $s$  by going to the limit  $s \rightarrow 0$ .

With the initial conditions so specified,  $\Phi^0 = (1, 0, 0, 0)$ , and the problem is reduced to the determination of the single matrix element  $[\mathbf{S}(t), \mathbf{A}(t)]_{11}$  in the interaction representation.

As explained in Section II, in the interaction representation the  $x$ -component of the spin has the form  $Z_x$ , given by (V.11), whereas the  $z$ -component  $\mathbf{Z}_z$  is the same as  $\mathbf{S}_z$ , as given in (II.2). Using

$$\mathbf{A}(t) = \int_{-\infty}^t e^{s t'} \cos \omega t' \mathbf{Z}_x(t') dt' \quad (\text{IV.3})$$

(cf. (II.4)), one obtains in the linear response approximation, (that is, neglecting terms proportional to  $H_\omega^2$ )

$$\begin{aligned} M^x(t) = \hbar^{-1} \mu^2 H_\omega \sin^2 \alpha \cos \omega t & \left[ \mathcal{P} \frac{\omega_{31}}{\omega_{31}^2 - \omega^2} + \mathcal{P} \frac{\omega_{41}}{\omega_{41}^2 - \omega^2} \right. \\ & \left. - 2\pi i \delta(\omega_{31} - \omega) - 2\pi i (\omega_{41} - \omega) \right], \text{ and } M^z(t) = 0. \end{aligned} \quad (\text{IV.4})$$

The symbol  $\mathcal{P}$  means, that the Cauchy principal value is to be taken, whenever an integral of this expression over the frequency  $\omega$  is required.  $\mathcal{P}$  may be omitted for  $\omega$  off resonance.

Inserting

$$\sin \alpha \simeq \frac{D}{4J}, \quad \omega_{31} \simeq \frac{J + \mu H_z + D^2/16J}{\hbar}, \quad \omega_{41} \simeq \frac{J - \mu H_z + D^2/16J}{\hbar}, \quad (\text{IV.5})$$

we obtain the static susceptibility

$$\frac{M_x^0}{H_\omega \cos \omega t} = \chi_{xx} = \frac{\mu^2 D^2}{8J^3} \quad (\text{IV.6})$$

for  $\omega = 0$ . The Kramers-Kronig relations are also fulfilled.

Since the first nonvanishing term of the  $z$ -component of the magnetic moment is quadratic in  $H_\omega$ , we proceed to calculate the quadratic response. Using (II.13) and (III.5) to take cognizance of the chronological ordering, the magnetic moment is again given by the matrix element (1, 1) of a sum of operators:

$$\begin{aligned} \mathbf{M}(t) = i \hbar^{-1} \mu^2 H_\omega & [\mathbf{S}^z, \mathbf{A}]_{11} + \mu^3 \hbar^{-2} H_\omega^2 (\mathbf{A} \mathbf{S} \mathbf{A})_{11} \\ & - (2\hbar)^{-1} \mu^3 H_\omega^2 (\mathbf{S} \mathbf{A}^2 + \mathbf{A}^2 \mathbf{S} + \mathbf{S} \mathbf{C} + \mathbf{C} \mathbf{S})_{11}. \end{aligned} \quad (\text{IV.7})$$

Here

$$C = (\mu H_\omega)^{-1} \int_0^t dt_1 [\mathcal{H}_{int}(t_1), \mathbf{A}(t_1)]. \quad (\text{IV.8})$$

The first term in (IV.7) is the linear response already calculated. Each summand of the third term of (IV.7) may be shown to vanish. For the  $x$ -component  $\mathbf{S}_x$ , the second term vanishes, too. Hence  $M^x(t)$  contains no quadratic term in  $H_\omega$ . There remains only

$$M^z(t) = \mu^3 \hbar^{-2} H_\omega^2 (\mathbf{A}(t) \mathbf{S}^z \mathbf{A}(t))_{11} = \mu^3 \hbar^{-2} H_\omega^2 (|A_{14}|^2 - |A_{13}|^2). \quad (\text{IV.9})$$

Evaluation of the matrix elements  $A_{14}$  and  $A_{13}$  yields

$$M^z(t) = \mu^3 \hbar^{-2} H_\omega^2 \sin^2 \alpha \left\{ \frac{1}{(\omega_{41}-\omega)^2} + \frac{1}{(\omega_{41}+\omega)^2} + \pi^2 \delta(\omega_{41}-\omega) \right. \\ + \mathcal{P} \frac{2}{\omega_{41}^2 - \omega^2} \cos 2\omega t - \frac{2}{\omega_{41} + \omega} \delta(\omega_{41}-\omega) \sin 2\omega t - \frac{1}{(\omega_{31}-\omega)^2} - \frac{1}{(\omega_{31}+\omega)^2} \\ \left. - \pi^2 \delta(\omega_{31}-\omega) - \mathcal{P} \frac{2}{\omega_{31}^2 - \omega^2} \cos 2\omega t + \frac{2}{\omega_{31} + \omega} \delta(\omega_{31}-\omega) \sin 2\omega t \right\}. \quad (\text{IV.10})$$

For  $\hbar\omega \ll J$  this expression reduces to

$$M^z(t) = \frac{1}{16} \frac{\mu^4 D^2}{J^5} H_\omega^2 (1 + \cos 2\omega t). \quad (\text{IV.11})$$

As expected, the magnetization contains, besides the static limit, a second harmonic component. Resonances occur for  $\omega = \omega_{31}$  and  $\omega = \omega_{41}$ .

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