Low-temperature dynamics of the classical antiferromagnetic Heisenberg chain in an external field

B. De Raedt
Institut für Festkörperforschung der Kernforschungsanlage Jülich, Postfach 1913, D-5170 Jülich, West Germany

H. De Raedt and J. Fivez
Departement Natuurkunde, Universitaire Instelling Antwerpen, B-2610 Wilrijk, Belgium
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The influence of the nonlinearity of the spin equations of motion on the dynamic structure factor of the antiferromagnetic Heisenberg chain in an external field is calculated in the low-temperature limit. The memory functions for the longitudinal and transverse spin component are calculated rigorously to lowest nonvanishing order in the temperature. In this limit, only two-magnon processes contribute to the memory functions. Divergencies in the two-magnon density of states give rise to resonances in addition to the usual spin-wave peak in the dynamic structure factor.

I. INTRODUCTION

One-dimensional magnetic systems have received much interest in the last few years for several reasons. First of all, many materials are known in which the magnetic interactions are quasi one dimensional, and much experimental work has been done on these systems, which show very interesting dynamical properties.[1-5] The most interesting phenomenon is the existence of well defined excitations, to be called spin waves, despite the lack of long-range order at nonzero temperature. This is due to the short-range order, which can support spin waves in a large temperature range in one-dimensional magnets.

Theoretical studies have been mainly concerned with classical spin chains. The static properties of the Heisenberg chain can be calculated analytically,[6,7] but the evaluation of dynamic correlation functions is far from trivial. Therefore, most of the theoretical work is based on the knowledge of the low-order frequency moments, and especially continued-fraction methods turned out to be extremely useful.[7-11] Conventional mode-coupling theories are inadequate to describe the low-temperature region because they lead to erroneous static correlations and consequently to nonvanishing linewidths at finite temperatures.[12] Recently, Reiter and Sjölander put forward a theory which gives the dynamic correlation function exact up to lowest nontrivial order in temperature.[13] Their treatment is based on a low-temperature expansion of the memory function. The first nonvanishing term of the memory function could be calculated analytically for both the ferro- and antiferromagnetic Heisenberg chain in zero field. Almost all the work mentioned above is restricted to the zero-field case. Recently, however, it was found that in applying an external field the dynamics are changed drastically. From computer simulations Loveluck *et al.* found a second resonance besides the usual magnon peak, in the spectrum of the z component of a ferromagnetic Heisenberg chain in a magnetic field.[14] They argued that this is due to the coupling between the longitudinal spin component and the energy density fluctuations.[15] However, following Reiter *et al.*, the second resonance should be a consequence of a divergence in the two-magnon density of states.[16] The antiferromagnetic chain in an external magnetic field was examined by the present authors.[17] The six lowest frequency moments were calculated with the transfer-operator method,[18] for both the longitudinal and transverse components. The dynamic correlation functions were estimated by means of the continued-fraction technique.[19] The most surprising result was, that for both spin components, the spectrum showed resonances in addition to the usual spin-wave peak, even at fairly low temperature. For the longitudinal component, it could be ruled out that the coupling with the energy density fluctuations is solely responsible for this phenomenon. Therefore we concluded that many-spin-wave resonances show up in the spectrum, even at relatively low temperatures. A complete physical understanding of the observed features is still lacking at this moment.

A different starting point to treat classical spin chains is the continuum description. In the continuum limit it is possible to take the nonlinearity of the equations of motion fully into account. Fogedby showed that the spectrum of the continuum Heisenberg ferromagnet is exhausted by spin waves and solitary waves.[20] For other systems like the planar spin chain[21] and the Heisenberg chain in an external field,[22] solitary solutions exist. The influence of nonlinear solutions on the dynamic structure factor however is not clear. At this moment, no satisfactory treatment of the thermodynamics of these solitary modes exists. Nevertheless some experimentalists claim to have found soliton contributions in the spectra of one-dimensional magnets at low temperatures.[23,24] In our opinion, a reliable description of the effects of the nonlinearity of the equations of motion on the dynamic structure factor at low temperatures, must start from a spin-wave picture for two reasons. At zero temperature, only spin waves contribute to the spectrum, and consequently the nonlinearity is completely negligible. Furthermore, nonlinear solutions of the equations of motion can be represented as multimagnon states. Obviously, at low temperatures two-magnon processes will be more important than three-magnon processes, and therefore, instead of taking the complete nonlinearity of certain solutions into account, one should rather consider all two-magnon processes, because it is this part of the nonlinearity which will affect the spectrum at low temperatures. The method of Ref. 13 seems to be most suited for this purpose. In the case of the Heisenberg chain in zero field, the main influence of the two-magnon processes, consists in a broadening of the spin-wave excitation. As we will show, this is not the case anymore when a field is present, because apart from a broadening of the spin wave, the two-magnon processes give rise to additional resonances in the spectrum. The weight of these resonances seems large enough in a number of cases to be able to be detected experimentally.

In the present work we investigate the same system from a different starting point. In order to calculate the memory functions we follow the method, proposed by Reiter and Sjölander,[13] and extend it to the nonzero field case. In this way, two-spin-wave processes are taken into account, and these should be responsible, at least in part, for the unusual behavior of the dynamic correlation functions.

In Sec. II we discuss the harmonic approximation. For the direct evaluation of the dynamic correlation functions, the harmonic approximation is not very useful, because it only gives the trivial $T=0$ limit correctly. However, we need the harmonic approximation for the calculation of the memory function. In Sec. III we use Mori's projection operator formalism in order to express the dynamic correlation functions in terms of their second frequency moments and their memory functions, and we discuss the low-temperature expansion of the memory functions. In Sec. IV we present the results for the longitudinal and transverse components and we make a comparison with our previous work. The conclusions are summarized in Sec. V. The explicit calculation of the memory function is given in an Appendix because it is rather technical.

II. HARMONIC APPROXIMATION

The Hamiltonian of an antiferromagnetic Heisenberg chain in an external magnetic field is given by

$$H=J\sum_{n=1}^{N}(\vec{S}_n\cdot\vec{S}_{n+1}-hS_n^z)\ , \tag{2.1}$$

with $J>0$, and where $h=g\mu_B B/J$ is the magnetic field in reduced units. In the following we put $J=1$ and we always consider the range of magnetic fields for which $0\leq h\leq 4$. Keeping in mind that we only consider classical spins of unit length, we may introduce spherical coordinates

$$S_n^x=(-1)^n\sin\theta_n\cos\xi_n\ , \tag{2.2a}$$

$$S_n^y=(-1)^n\sin\theta_n\sin\xi_n\ , \tag{2.2b}$$

$$S_n^z=\cos\theta_n\ . \tag{2.2c}$$

We can now minimize the Hamiltonian, and we find that the ground state is determined by

$$\theta_n=\theta=\cos^{-1}(h/4);\quad n=1,\ldots,N\ , \tag{2.3a}$$

$$\xi_n=\xi;\quad n=1,\ldots,N\ , \tag{2.3b}$$

where ξ may be chosen arbitrarily. It is important to note that in the ground state, the longitudinal spin components order ferromagnetically, whereas the transverse components order antiferromagnetically. To obtain the Hamiltonian in the harmonic approximation, we introduce the new coordinates

$$\psi_n=\theta_n-\theta,\quad \phi_n=\sin\theta(\xi_n-\xi)\ , \tag{2.4}$$

and we can write

$$H=E_0+\tfrac{1}{2}\sum_n[\psi_n^2-2\cos(2\theta)\psi_n\psi_{n+1}+\psi_{n+1}^2+(\phi_n-\phi_{n+1})^2]\ , \tag{2.5a}$$

$$E_0=-N(1+2\cos^2\theta)=-N(1+\tfrac{1}{8}h^2)\ . \tag{2.5b}$$

After Fourier transformation, the Hamiltonian reads

$$H=\tfrac{1}{2}\sum_k[a(k)\psi_k\psi_{-k}+b(k)\phi_k\phi_{-k}]\ , \tag{2.6a}$$

$$a(k)=2[1-\cos(2\theta)\cos k]\ , \tag{2.6b}$$

$$b(k)=2(1-\cos k)\ , \tag{2.6c}$$

where we have omitted the ground-state energy and we rewrite Eq. (2.6) as

$$H=\tfrac{1}{2}\sum_k[\psi_k\psi_{-k}/m(k)+m(k)\omega^2(k)\phi_k\phi_{-k}]\ , \tag{2.7a}$$

$$m(k)=1/a(k)\ , \tag{2.7b}$$

$$\omega^2(k)=a(k)b(k)\ . \tag{2.7c}$$

We also want to examine the spin equations of motion. In their exact form, these look like

$$\dot{S}_n^z = S_{n-1}^x S_n^y - S_{n-1}^y S_n^x - S_n^x S_{n+1}^y + S_n^y S_{n+1}^x \ , \tag{2.8a}$$

$$\dot{S}_n^+ = i(-hS_n^+ + S_{n-1}^z S_n^+ - S_{n-1}^+ S_n^z - S_n^z S_{n+1}^+ + S_n^+ S_{n+1}^z) \ , \tag{2.8b}$$

where we used $S_n^+ = S_n^x + iS_n^y$. Combining Eqs. (2.2) and (2.8) we obtain

$$\dot{\theta}_n = -[\sin\theta_{n-1}\sin(\xi_{n-1}-\xi_n) - \sin\theta_{n+1}\sin(\xi_n - \xi_{n+1})] \ , \tag{2.9a}$$

$$\dot{\xi}_n = -h + \cos\theta_{n-1} + \cos\theta_{n+1} + \cot\theta_n[\sin\theta_{n-1}\cos(\xi_{n-1}-\xi_n) + \sin\theta_{n+1}\cos(\xi_n - \xi_{n+1})] \ . \tag{2.9b}$$

We can expand $\dot{\theta}_n$ and $\dot{\xi}_n$ to lowest order in ψ_n and ϕ_n, and if we then perform a Fourier transformation, we get the linearized equations of motion

$$\dot{\psi}_k = b(k)\phi_k; \quad \dot{\phi}_k = -a(k)\psi_k \ . \tag{2.10}$$

We are interested in the dynamic spin-correlation functions

$$C_k^\perp(t) \equiv \langle S_k^x(t) S_{-k}^x(0)\rangle + \langle S_k^y(t) S_{-k}^y(0)\rangle \ , \tag{2.11a}$$

$$C_k^z(t) \equiv \langle S_k^z(t) S_{-k}^z(0)\rangle - N\langle S^z\rangle^2 \delta_{k,0} \ . \tag{2.11b}$$

We obtain these correlation functions in the harmonic approximation by using Eqs. (2.2)–(2.4), expanding Eqs. (2.11) up to lowest order in ψ and ϕ, and by using the well-known results

$$\langle \psi_k \psi_{-k}\rangle = 1/\beta a(k) \ , \tag{2.12a}$$

$$\langle \phi_k \phi_{-k}\rangle = 1/\beta b(k) \ , \tag{2.12b}$$

where $\beta = 1/T (k_B = 1)$. We then have

$$C_k^\perp(t) = C_k^\perp(0)\cos[\Omega(k)t] \ , \tag{2.13a}$$

$$C_k^z(t) = C_k^z(0)\cos[\omega(k)t] \ , \tag{2.13b}$$

with

$$C_k^\perp(0) = \frac{1}{\beta}[\cos^2\theta/a(k^*) + 1/b(k^*)] \ , \tag{2.14a}$$

$$C_k^z(0) = \sin^2\theta/\beta a(k) \ , \tag{2.14b}$$

$$\Omega(k^*) = \omega(k) \ , \tag{2.15}$$

$$k^* = \pi - k \ . \tag{2.16}$$

The main results of this section can be summarized as follows. In the limit $T \to 0$, the longitudinal spin component S_k^z performs a harmonic oscillation with frequency given by $\omega(k)$. This is easily understood, because at low temperature S_k^z reduces to the normal coordinate ψ_k as can be seen from Eqs. (2.2c) and (2.4a). Although the transverse component depends in a more complicated manner on both normal coordinates ϕ_k and $\psi_{k'}$, it also oscillates harmonically for $T \to 0$. Its frequency $\Omega(k)$ is not identical to the harmonic frequency $\omega(k)$ but it is shifted over a wave vector π. Physically speaking this is a consequence of the antiferromagnetic ordering of the transverse component in the ground state. Mathematically it is expressed in the transformation Eq. (2.2) by the factor $(-1)^n$, which leads to a shift over π after Fourier transforming. In Fig. 1 we have plotted $\omega(k)$ for different magnetic fields.

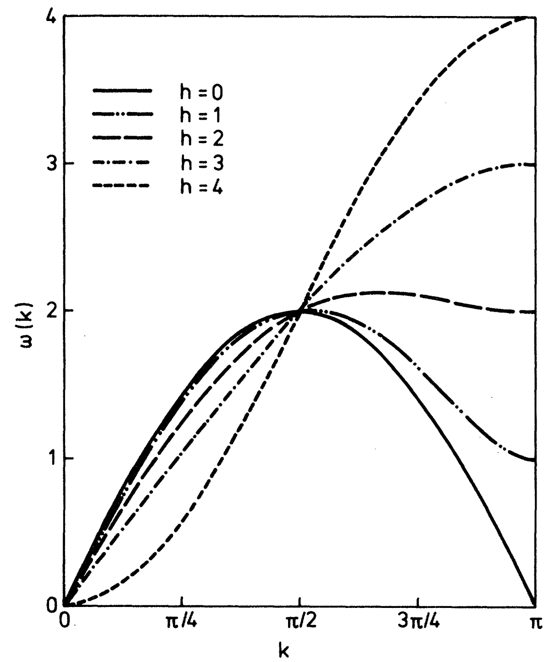

FIG. 1. Dispersion relations for the longitudinal spin component for different magnetic fields. The dispersions for the transverse component are obtained by replacing k by $k^* = \pi - k$.

III. DYNAMIC CORRELATION FUNCTIONS

For the calculation of the dynamic correlation functions we use Mori's projection operator technique as a starting point.[19] We start with some basic definitions. The time evolution of a dynamic variable A is determined by the Liouville operator L in the following way:

$$LA = -i\dot{A} = -i\{A,H\} \ , \tag{3.1}$$

where the curly brackets are the Poisson brackets. It is convenient to define

$$(A,B) = \langle A^+ B\rangle - \langle A^+\rangle\langle B\rangle \ . \tag{3.2}$$

The time-dependent correlation functions can then be written as

$$C_q^\alpha(t)=(S_q^\alpha(t),S_q^\alpha(0))=(S_q^\alpha,e^{-iLt}S_q^\alpha) \quad (3.3)$$

and after Laplace transforming we find

$$C_q^\alpha(z)=-i\int_0^\infty dt e^{izt}C_q^\alpha(t)$$
$$=(S_q^\alpha,(z-L)^{-1}S_q^\alpha);\ z=\omega+i\epsilon \ . \quad (3.4)$$

Following Mori we can write $C_q^\alpha(z)$ as

$$C_q^\alpha(z)=C_q^\alpha(t=0)\frac{z+\Sigma_q^\alpha(z)}{z^2+z\Sigma_q^\alpha(z)-\langle\omega^2\rangle_q^\alpha} \ , \quad (3.5)$$

where $\langle\omega^2\rangle_q^\alpha$ is the second frequency moment and $\Sigma_q^\alpha(z)$ is the memory function and the explicit expressions read

$$\langle\omega^2\rangle_q^\alpha=\frac{(LS_q^\alpha,LS_q^\alpha)}{(S_q^\alpha,S_q^\alpha)}=\frac{(\dot S_q^\alpha,\dot S_q^\alpha)}{(S_q^\alpha,S_q^\alpha)} \ , \quad (3.6)$$

$$\Sigma_q^\alpha(z)=-(LS_q^\alpha,LS_q^\alpha)^{-1}$$
$$\times(QL^2S_q^\alpha,(z-QLQ)^{-1}QL^2S_q^\alpha) \ . \quad (3.7)$$

The second frequency moments can be calculated rigorously for all temperatures and wave vectors by means of the transfer operator method.[17,18] The main problem in the evaluation of the memory function is that its time evolution is determined by QLQ, where Q is the projection operator which projects on the nonsecular variables and which is defined by

$$QA_q=A_q-\frac{(LS_q^\alpha,A_q)}{(LS_q^\alpha,LS_q^\alpha)}LS_q^\alpha-\frac{(S_q^\alpha,A_q)}{(S_q^\alpha,S_q^\alpha)}S_q^\alpha \ . \quad (3.8)$$

Let us write the memory function in time space as

$$\Sigma_q^\alpha(t)=-(LS_q^\alpha,LS_q^\alpha)^{-1}M_q^\alpha(t) \ , \quad (3.9a)$$

$$M_q^\alpha(t)=(QL^2S_q^\alpha,e^{-iQLQt}QL^2S_q^\alpha) \ . \quad (3.9b)$$

We can show (see Appendix) that in the limit of low temperatures $(LS_q^\alpha,LS_q^\alpha)\propto T$ and $M_q^\alpha(0)\propto T^2$. Consequently $\Sigma_q^\alpha(t)$ vanishes proportional to T. For nonzero times we write the Taylor expansion of $M_q^\alpha(t)$:

$$M_q^\alpha(t)=\sum_{n=0}^\infty\frac{(-it)^{2n}}{(2n)!}(QL^2S_q^\alpha,(QLQ)^{2n}QL^2S_q^\alpha) \ . \quad (3.10)$$

Now each term can be expanded in a temperature series, and to lowest order it must be quadratic in T. It can be shown that keeping only the lowest-order contribution is equivalent to omitting the Q operators in the time evolution. Thus we have

$$(QL^2S_q^\alpha,(QLQ)^{2n}QL^2S_q^\alpha)=(QL^2S_q^\alpha,L^{2n}QL^2S_q^\alpha)$$
$$+O(T^3) \ , \quad (3.11)$$

or, equivalently,

$$M_q^\alpha(t)=(QL^2S_q^\alpha,e^{-iLt}QL^2S_q^\alpha)+O(T^3) \ . \quad (3.12)$$

In Ref. 13 a detailed proof of Eq. (3.12) is given in the case of a Heisenberg chain in zero field. The advantage of Eq. (3.12) is that we can use the harmonic approximation for the evaluation of the lowest-order term. The explicit evaluation of $M_q^\alpha(t)$ is given in the Appendix. It is worth noting that the relation (3.12) can be obtained directly in frequency space. Defining

$$\Lambda_q^\alpha(z)=-(LS_q^\alpha,LS_q^\alpha)^{-1}(QL^2S_q^\alpha,(z-L)^{-1}QL^2S_q^\alpha) \ , \quad (3.13)$$

one can derive the exact relation

$$(z^2-\langle\omega^2\rangle_q^\alpha)\Sigma_q^\alpha(z)=(z^2-\langle\omega^2\rangle_q^\alpha)\Lambda_q^\alpha(z)$$
$$+z\Lambda_q^\alpha(z)\Sigma_q^\alpha(z) \ . \quad (3.14)$$

Knowing that

$$\Sigma_q^\alpha(t=0)=\Lambda_q^\alpha(t=0)=\langle\omega^4\rangle_q^\alpha/\langle\omega^2\rangle_q^\alpha-\langle\omega^2\rangle_q^\alpha\propto T \quad (3.15)$$

and expanding in the temperature one finds

$$\Sigma_q^\alpha(z)=\Lambda_q^\alpha(z)+O(T^2);\ z=\omega+i\epsilon \ . \quad (3.16)$$

Strictly speaking, Eq. (3.16) only holds when the lowest-order term of $\Lambda_q^\alpha(z)$ is not too large. However, if there are points in frequency space where this term becomes very large or even divergent, an expansion is not allowed anymore and the Q operators in the time evolution should be taken into account. The results (3.12) and (3.16), can therefore be considered as being exact only in the following sense: The Taylor expansion of Eq. (3.12), or equivalently the asymptotic expansion of Eq. (3.16) gives all the expansion coefficients exact to lowest nonvanishing order in the temperature.

We now turn to the discussion of the result for $\Sigma_q^\alpha(z)$ for the special case $h=2\sqrt{2}$ for which we can give the analytic result. For this purpose we must take the Laplace transform of Eqs. (A29) and (A32) giving

$$\Sigma_{q^*}^{\perp}(z)=-4T\frac{(1+\cos q)}{3-\cos q}\Bigg[I(c,u)(z^2-\lambda_1^2)^{-1/2}+I(-c,u)(z^2-\lambda_2^2)^{-1/2}$$
$$-\frac{1}{z}\left[(3+4c^2-4c^4)u^2+2\frac{5-c^2-c^4}{1-c^2}u^4+2\frac{1+c^2}{(1-c^2)^2}u^6\right]\Bigg] \ , \quad (3.17)$$

$$\Sigma_q^z(z) = -2T(1-\cos q)\left[K(c,u)(z^2-\lambda_1^2)^{-1/2} + K(-c,u)(z^2-\lambda_2^2)^{-1/2} + \frac{1}{z}\left\{2(3+2c^2)u^2 - 2\frac{(1+6c^2+c^4)}{(1-c^2)^2}u^4\right\}\right] , \tag{3.18}$$

$$u = \frac{z}{h};\ \lambda_1 = 2h\left|\cos\frac{q}{4}\right|;\ \lambda_2 = 2h\left|\sin\frac{q}{4}\right|;\ c = \cos\frac{q}{2};\ h = 2\sqrt{2} , \tag{3.19}$$

$$I(c,u) = [(1+c)(1+c-c^2) - (2-c)u^2]^2 + \left(c - \frac{u^2}{1+c}\right)^2 u^2 , \tag{3.20}$$

$$K(c,u) = \left(2 - c^2 - \frac{1-c}{1+c}u^2\right)^2 . \tag{3.21}$$

We see that the memory functions diverge for $z = \lambda_1$ and for $z = \lambda_2$. This is a consequence of a divergence in the two-magnon density of states

$$n_\pm = \left|\frac{dk}{d\Omega_\pm}\right| , \tag{3.22}$$

where $\Omega_\pm$, given by Eq. (A26), now reduce to

$$\Omega_+(k,q) = \left|\lambda_1 \sin\frac{k}{2}\right|;\ \Omega_-(k,q) = \left|\lambda_2 \cos\frac{k}{2}\right| . \tag{3.23}$$

The quantity of interest however is the dynamic structure factor $C_q^\alpha(\omega)$, which is the imaginary part of $C_q^\alpha(z)$ and which is given by

$$C_q^\alpha(\omega) = C_q^\alpha(t=0)\frac{\langle\omega^2\rangle_q^\alpha \Sigma_q^{\alpha''}(\omega)}{[\omega^2 - \langle\omega^2\rangle + \omega\Sigma_q^{\alpha'}(\omega)]^2 + [\omega\Sigma_q^{\alpha''}(\omega)]^2} , \tag{3.24}$$

where $\Sigma_q^{\alpha'}(\omega)$ and $\Sigma_q^{\alpha''}(\omega)$ are the real and imaginary part of $\Sigma_q^\alpha(z)$. In Fig. 2 we have plotted $\Sigma_q^{\perp''}(\omega)$ and $C_q^\perp(\omega)$ for $q = 3\pi/4$ and $h = 2\sqrt{2}$. From Eq. (3.24) we see that $C_q^\perp(\omega)$ is zero when $\Sigma_q^{\perp''}(\omega)$ diverges and in this way a second resonance near $\omega = \lambda_2(q^*)$ originates. The effect of the divergency at $\omega = \lambda_1(q^*)$ on $C_q^\perp(\omega)$ is too small to be seen on the same scale.

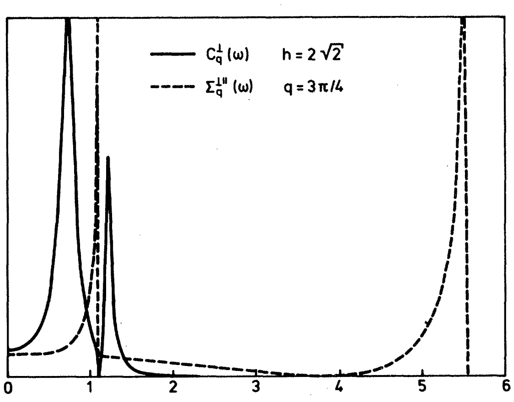

FIG. 2. Dynamic correlation function and imaginary part of the memory function of the transverse component for $h = 2\sqrt{2}$ and $q = 3\pi/4$. The ordinate differs for both curves.

For $h = 2\sqrt{2}$ we have $\omega(q) = h|\sin(q/2)|$, and therefore we have for all wave vectors

$$\omega(q) \le \lambda_1(q);\ \omega(q) \le \lambda_2(q);\ h = 2\sqrt{2} . \tag{3.25}$$

Consequently, for $h = 2\sqrt{2}$ only the high-frequency part of the spectrum is affected by the divergencies in the memory function. For $h = 0$ we reobtain the result of Reiter and Sjölander[13] for the antiferromagnet

$$\Sigma_q(z) = -\frac{T}{2}[z - (z^2 - \Omega_1^2)^{1/2} + \tfrac{1}{16}\Omega_2^4(z^2 - \Omega_2^2)^{-1/2}] \tag{3.26}$$

with Ω_1 and Ω_2 given by Eq. (A28). We now have

$$\omega(q) = 2|\sin q|;\ \omega(q) \le \Omega_1(q) ;$$
$$\omega(q) \le \Omega_2(q);\ h = 0 . \tag{3.27}$$

For a detailed discussion of this case we refer to Ref. 13. We only remark that as for $h = 2\sqrt{2}$ divergencies in the memory function only occur at frequencies larger than the spin-wave frequency.

IV. NUMERICAL RESULTS

In Eqs. (A17) and (A18) the results for the memory functions in time space are presented. The integration over the Brillouin zone can easily be done numerically by replacing the integral by a sum over discrete points $k_n = 2\pi n/N$. In this way $\Sigma_q^\alpha(t)$ can be determined to any desired precision. As an example we have plotted $\Sigma_q^z(t)/\Sigma_q^z(0)$ for $q=\pi/2$ and $h=1$ in Fig. 3. However, for the calculation of frequency-dependent quantities, another numerical technique is required. If we take the Laplace transform of Eqs. (A17) and (A18) we end up with integrals of the form

$$G(z)=\frac{1}{\pi}\int_0^\pi dk A(k)\frac{1}{z\pm\Omega(k)};\quad z=\omega+i\epsilon\ . \tag{4.1}$$

Using the well-known identity

$$\lim_{\epsilon\to 0^+}\frac{1}{\omega\pm\Omega(k)+i\epsilon}=P\frac{1}{\omega\pm\Omega(k)}-i\pi\delta(\omega\pm\Omega(k))\ , \tag{4.2}$$

where P stands for principal value, we can write $G(z)=R(\omega)+iI(\omega)$ with

$$R(\omega)=\frac{1}{\pi}P\int_0^\pi dkA(k)\frac{1}{\omega+\Omega(k)}\ , \tag{4.3}$$

$$I(\omega)=-\int_0^\pi dkA(k)\delta(\omega+\Omega(k))\ . \tag{4.4}$$

In order to integrate Eqs. (4.3) and (4.4) numerically we used the linear analytic method as proposed by Gilat.[25] The method essentially consists in dividing the Brillouin zone in small intervals. In each interval $A(k)$ and $\Omega(k)$ are replaced by a linear approximation and the integral over the interval is evaluated analytically. The advantage of the linear analytic method is that it allows a simultaneous high-accuracy calculation of $R(\omega)$ and $I(\omega)$.

We now turn to a discussion of the results for $h=1$. Let us first reconsider Eq. (4.4). If we change the integration variable from k to $\Omega(k)$ we see that $I(\omega)$ diverges whenever $d\Omega(k)/dk=0$, or equivalently whenever the density of states $|dk/d\Omega(k)|$ diverges. Let us demonstrate this effect by a specific example. In Fig. 4 we have plotted Ω_+ and Ω_- as defined by Eq. (A26) for $q=3\pi/4$ and as a function of k. If we define the two-magnon density of states by

$$n_\pm=\left|\frac{dk}{d\Omega_\pm(k)}\right|\ , \tag{4.5}$$

we see from Fig. 4 that n_- diverges for $\omega\simeq 0.6$ and $\omega\simeq 3.6$ and n_+ diverges for $\omega\simeq 3.8$. Figure 5 shows $\Sigma_q^{z\prime\prime}(\omega)$ for the same parameters. Besides the three divergencies we observe a broad resonance around $\omega=2$. This is due to a decrease of the slope of Ω_+ in

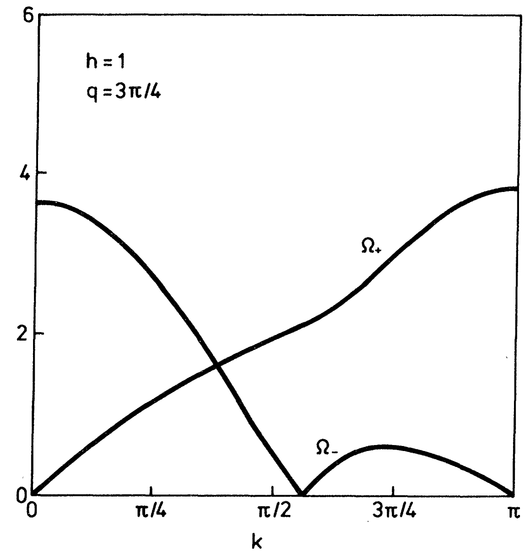

FIG. 4. Two-magnon dispersions $\Omega_\pm(k,q)$ for $h=1$ and $q=3\pi/4$ and as a function of the wave vector k.

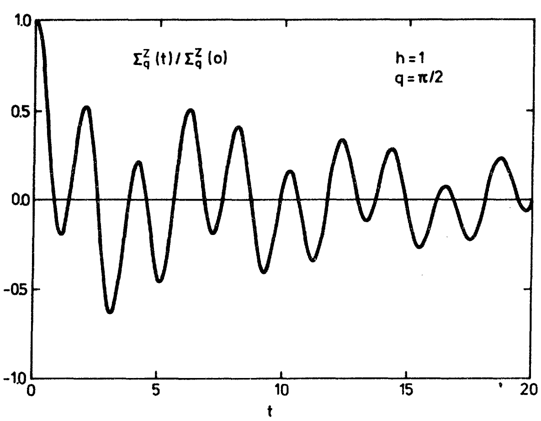

FIG. 3. Time-dependent memory function of the longitudinal component for $h=1$ and $q=\pi/2$.

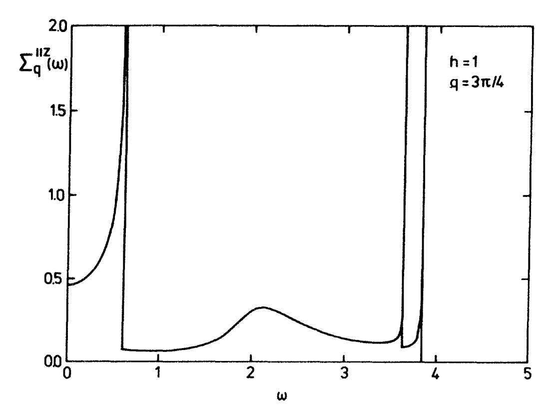

FIG. 5. Imaginary part of the memory function of the longitudinal component for $h=1$ and $q=3\pi/4$.

this frequency region. We conclude that we can understand the main behavior of $\Sigma_q^{\alpha''}(\omega)$ from the two-magnon dispersions $\Omega_{\pm}$, although for an explicit evaluation the knowledge of the functions P, Q, R, S, F, and G, Eqs. (A20)–(A25), is required. In Figs. 6–9 the dynamic structure factor for both components is shown for $T=0.1$ and $T=0.2$ and for $q=15\pi/16$, $q=7\pi/8$, and $q=3\pi/4$. We observe that divergencies in the two-magnon density of states give rise to additional peaks, besides the usual spin-wave resonance. It is important to note that two-magnon resonances also show up in the low-frequency part of the spectrum. This is not the case for $h=0$ and $h=2\sqrt{2}$ as already discussed in Sec. III. Resonances that occur at frequencies lower than the spin-wave frequency have generally more weight than resonances that occur at higher frequencies and therefore they should be more accessible for experimental measurement. Another important point is the fact that the two-magnon resonances are present at any nonzero temperature, although the spin-wave resonance gains more weight when the temperature is lowered.

In Ref. 17 we examined the antiferromagnetic Heisenberg chain in an applied field, starting from the continued-fraction method and the exact knowledge of the six lowest-frequency moments for both the longitudinal and transverse component. We found second resonances for both components in the low-frequency range even at fairly low temperatures. Obviously these peaks correspond to the two-magnon resonances at the left of the spin-wave peak in Figs. 6–9. The quantitative agreement between the two approaches is very poor because the continued-fraction method gives only a rather crude approximation for the dynamic structure factor and fine details are smeared out in this approach. On the other hand, the continued-fraction method is not restricted to low temperatures because it also takes many-spin-wave processes into account. The present theory contains only two-spin-wave processes and probably the sharp two-magnon resonances will be rounded by higher-spin-wave processes.

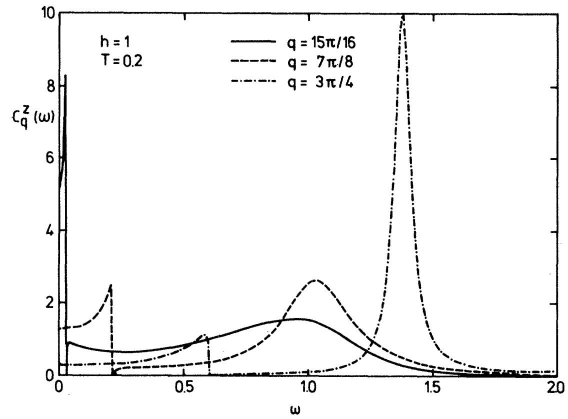

FIG. 7. See Fig. 6 but now $T=0.2$.

We have restricted the numerical results to one particular value of the magnetic field ($h=1$), because as far as we know there exist no experimental studies of the dynamics of a one-dimensional isotropic antiferromagnet in an external field at the moment. A substance for which such experiments are possible in principle is TMMC [$(CD_3)_4NMnCl_3$]. This one-dimensional antiferromagnet has a spin $S=\frac{5}{2}$ and an exchange parameter $J \simeq 13$ K. As the spin value is rather large, one may expect a classical approach to give good results. The reduced magnetic field is defined by

$$h = g\mu_B H / J\sqrt{S(S+1)} \ , \tag{4.6}$$

and a simple calculation shows us that $h=1$ corre-

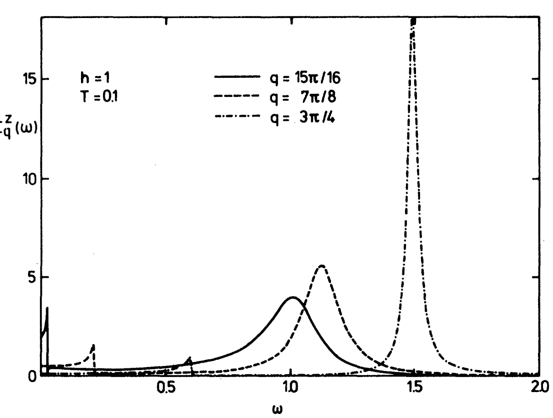

FIG. 6. Normalized dynamic structure factor of the z component for $h=1$, $T=0.1$ and for different wave vectors.

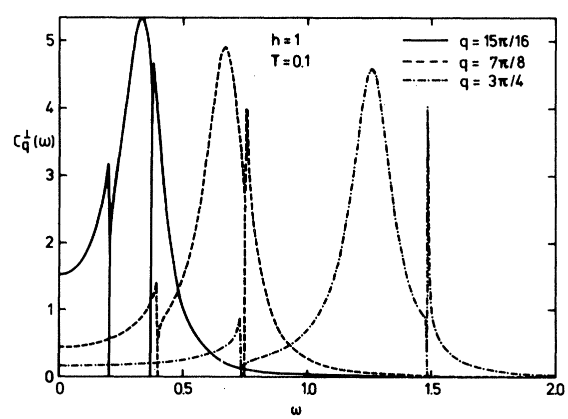

FIG. 8. Normalized dynamic structure factor of the transverse component for $h=1$, $T=0.1$ and for different wave vectors.

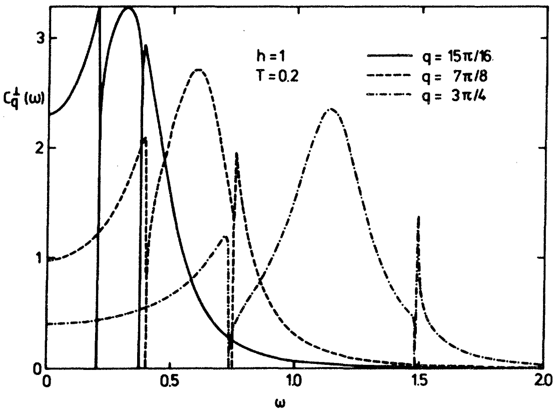

FIG. 9. See Fig. 8 but now $T=0.2$.

sponds to a magnetic field $H \simeq 250$ kG. As the experimental limit lies around 50 kG or maybe 100 kG, we should consider values of $h=0.2$ or $h=0.4$, to get results that are also accessible experimentally. One can however easily discuss any h value more closely, by examining the two-magnon dispersions $\Omega_{\pm}(k,q)$, as given by Eq. (A26). Indeed, the location of two-magnon resonances in the spectrum, is simply given by those frequencies for which $\partial\Omega_{\pm}(k,q)/\partial k=0$. Obviously, two-magnon resonances will occur in the spectrum at frequencies lower than the spin-wave value for any nonzero magnetic field, although the effects will be more prominent for larger magnetic fields.

V. CONCLUSIONS

We have presented a detailed account on the low-temperature dynamics of the antiferromagnetic Heisenberg chain in an external field. At zero temperature the longitudinal, as well as the transverse spin component exhibits undamped oscillations, because the memory function, which is responsible for damping effects, vanishes proportional to the temperature for $T\rightarrow 0$. More explicitly, to lowest nonvanishing order in the temperature, only two-spin-wave processes contribute to the memory function. Two-magnon resonances show up in the spectra at those frequencies where the two-magnon density of states diverges. Mostly, the weight of these resonances is rather small. However when they occur at frequencies, smaller than the spin-wave frequency, which is the case for moderate magnetic fields, their weight may be large enough to be detected experimentally.

It is obvious that other one-dimensional magnetic systems can be treated with the same method, which was first employed by Reiter and Sjölander for the zero-field case.[13] In particular, any correlation function, oscillating undamped at zero temperature, can be calculated with this method and the result is exact to the extent that it gives all the frequency moments exact up to lowest nontrivial order in the temperature. Therefore, the following conclusions are not restricted to one particular system. Our results clearly show that one should try to understand nonlinear effects, showing up in the dynamic structure factor, in terms of two- (or multi-) magnon processes, in particular if these effects are observed at low temperatures. A theory starting from a continuum description and some particular nonlinear solutions of the equations of motion (like solitons or breathers), cannot give a reliable estimate of the influence of these specific solutions on the dynamic structure factor.[26] Figure 7 demonstrates that one should be careful in interpreting quasielastic peaks as being due to solitons.

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APPENDIX

In Sec. III we found that the functions

$$M_q^\alpha(t)=(QL^2S_q^\alpha,e^{-iLt}QL^2S_q^\alpha);\quad \alpha=\perp,z\ , \tag{A1}$$

need to be evaluated to lowest nonvanishing order in the temperature. This means that the time evolution $\exp(-iLt)$ may be replaced by the harmonic time evolution, which is determined by Eq. (2.10), and that the expectation value may be evaluated with the harmonic Hamiltonian (2.6a). In $QL^2S_q^\alpha$ we must take the full Liouville operator L into account, but we can expand $QL^2S_q^\alpha$ in the normal coordinates ψ_k and ϕ_k and we must only retain the lowest-order terms, which are quadratic in the normal coordinates. The projection operator Q can be worked out explicitly

$$QL^2S_q^\alpha=L^2S_q^\alpha-\langle\omega^2\rangle_q^\alpha S_q^\alpha \tag{A2}$$

and the second moment $\langle\omega^2\rangle_q^\alpha$ can be replaced by its zero-temperature value, because the first correction is proportional to T, which leads to higher-order terms. Thus we have, consistently to lowest order

$$QL^2S_{q^*}^\alpha=L^2S_{q^*}^\alpha-\omega^2(q)S_{q^*}^\alpha;\quad \alpha=x,y;\quad q^*=\pi-q\ , \tag{A3}$$

$$QL^2S_q^z=L^2S_q^z-\omega^2(q)S_q^z\ . \tag{A4}$$

To get the explicit expressions we need the equations of motion (2.10) to be extended to second order in the normal coordinates. The results read

$$L\psi_q = -ib(q)\phi_q + 2i\cot\theta\frac{1}{\sqrt{N}}\sum_k\left[\cos q - \cos\left(k+\frac{q}{2}\right)\right]\psi_{k+q/2}\phi_{-k+q/2} , \tag{A5}$$

$$L\phi_q = ia(q)\psi_q - 2i\cot\theta\frac{1}{\sqrt{N}}\sum_k\left[\cos\frac{q}{2}\left(\cos k - \cos\frac{q}{2}\right)\phi_{k+q/2}\phi_{-k+q/2} + \left(1-\cos\frac{q}{2}\cos k - \sin^2\theta\cos q\right)\psi_{k+q/2}\psi_{-k+q/2}\right] . \tag{A6}$$

With Eqs. (A3)–(A6) it is straightforward, although very tedious, to obtain

$$QL^2S^x_{q^*} = \frac{1}{\sqrt{N}}\sum_k [u(k,q)\phi_{k+q/2}\phi_{-k+q/2} + v(k,q)\psi_{k+q/2}\psi_{-k+q/2}] , \tag{A7}$$

$$QL^2S^y_{q^*} = \frac{1}{\sqrt{N}}\sum_k w(k,q)\psi_{k+q/2}\phi_{-k+q/2} , \tag{A8}$$

$$QL^2S^z_q = \frac{1}{\sqrt{N}}\sum_k [f(k,q)\phi_{k+q/2}\phi_{-k+q/2} + g(k,q)\psi_{k+q/2}\psi_{-k+q/2}] , \tag{A9}$$

with

$$u(k,q) = 8\sin\theta\cos\frac{q}{2}\left(\cos\frac{q}{2} - \cos k\right)\left[1 - \cos\frac{q}{2}\left(\cos\frac{q}{2} + \cos k\right)\right] , \tag{A10}$$

$$v(k,q) = 8\sin\theta\cos\frac{q}{2}\left[\cos\frac{q}{2}\sin^2\frac{q}{2} + \left(1 + 2\cos^2\theta\sin^2\frac{q}{2}\right)\cos k - \cos(2\theta)\cos\frac{q}{2}\cos^2 k\right] , \tag{A11}$$

$$w(k,q) = 16\sin(2\theta)\cos\frac{q}{2}\sin\left(\frac{k}{2}-\frac{q}{4}\right)\left[\sin\left(\frac{k}{2}-\frac{q}{4}\right)\cos k - \sin\frac{q}{2}\cos\left(\frac{k}{2}-\frac{q}{4}\right)\right] , \tag{A12}$$

$$f(k,q) = 4\cos\theta(1-\cos q)\left(\cos^2 k - \cos^2\frac{q}{2}\right) , \tag{A13}$$

$$g(k,q) = 4\cos\theta(1-\cos q)\left[2\sin^2\theta - \left(\cos\frac{q}{2} + \cos k\right)\left(\cos\frac{q}{2} - \cos(2\theta)\cos k\right)\right] . \tag{A14}$$

The reader might be surprised that in combining Eq. (A1) with Eqs. (A7) and (A8) one finds $M^x_q(t) \neq M^y_q(t)$. This is because Eqs. (A7) and (A8) were obtained by expanding around $\phi=0$. Evidently we might have chosen any other direction in the xy plane, because of the continuous symmetry in this plane. Therefore one can use Eqs. (A7 and A8) only for the calculation of quantities that are fully symmetric themselves, such as $M^\perp_q(t) = M^x_q(t) + M^y_q(t)$. In order to get the memory functions completely we see from Eq. (3.9) that we also need the quantities $(LS^\alpha_q, LS^\alpha_q)$. To lowest order in the temperature one finds

$$(LS^\perp_{q^*}, LS^\perp_{q^*}) = 2T[\cos^2\theta(1-\cos q) + 1 - \cos(2\theta)\cos q] , \tag{A15}$$

$$(LS^z_q, LS^z_q) = 2T\sin^2\theta(1-\cos q) . \tag{A16}$$

Using Eqs. (A7)–(A16) and the results of Sec. II we finally obtain for the memory functions (3.9)

$$\Sigma^\perp_{q^*}(t) = -C_\perp\frac{1}{N}\sum_k \{[(P-Q)^2 + (R+S)^2]\cos(\Omega_+ t) + [(P+Q)^2 + (R-S)^2]\cos(\Omega_- t)\} , \tag{A17}$$

$$\Sigma^z_q(t) = -C_z\frac{1}{N}\sum_k [(F+G)^2\cos(\Omega_+ t) + (F-G)^2\cos(\Omega_- t)] , \tag{A18}$$

with $C_\perp$, C_z, P, Q, R, S, F, G, and $\Omega_\pm$ given by

$$C_\perp = 4T\sin^2\theta(1+\cos q)/[\cos^2\theta(1-\cos q)+1-\cos(2\theta)\cos q] \ , \tag{A19a}$$

$$C_z = 2T\cot^2\theta(1-\cos q) \ , \tag{A19b}$$

$$P = 1-\cos\frac{q}{2}\left(\cos\frac{q}{2}+\cos k\right) \ , \tag{A20}$$

$$Q = 2\left[\cos\frac{q}{2}\sin^2\frac{q}{2}+\left(1+2\cos^2\theta\sin^2\frac{q}{2}\right)\cos k-\cos(2\theta)\cos\frac{q}{2}\cos^2 k\right]\Big/\left[a\left(k+\frac{q}{2}\right)a\left(k-\frac{q}{2}\right)\right]^{1/2} \ , \tag{A21}$$

$$R = 2\cos\theta\left[\sin\left(\frac{k}{2}-\frac{q}{4}\right)\cos k-\sin\frac{q}{2}\cos\left(\frac{k}{2}-\frac{q}{4}\right)\right]\Big/\left[a\left(k+\frac{q}{2}\right)\right]^{1/2} \ , \tag{A22}$$

$$S = 2\cos\theta\left[\sin\left(\frac{k}{2}+\frac{q}{4}\right)\cos k+\sin\frac{q}{2}\cos\left(\frac{k}{2}+\frac{q}{4}\right)\right]\Big/\left[a\left(k-\frac{q}{2}\right)\right]^{1/2} \ , \tag{A23}$$

$$F = \cos\frac{q}{2}+\cos k \ , \tag{A24}$$

$$G = 2\left\{2\sin^2\theta-\left(\cos\frac{q}{2}+\cos k\right)\left[\cos\frac{q}{2}-\cos(2\theta)\cos k\right]\right\}\Big/\left[a\left(k+\frac{q}{2}\right)a\left(k-\frac{q}{2}\right)\right]^{1/2} \ , \tag{A25}$$

$$\Omega_\pm = 2\left|\sin\left(\frac{k}{2}+\frac{q}{4}\right)\left[a\left(k+\frac{q}{2}\right)\right]^{1/2}\pm\sin\left(\frac{k}{2}-\frac{q}{4}\right)\left[a\left(k-\frac{q}{2}\right)\right]^{1/2}\right| \ . \tag{A26}$$

In two special cases the integrals in Eqs. (A17) and (A18) can be done analytically. In the zero-field case $h=0$ we obtain

$$\Sigma_q(t) = -8T\left(\sin^2\frac{q}{2}\frac{J_1(\Omega_1 t)}{\Omega_1 t}+\cos^4\frac{q}{2}J_0(\Omega_2 t)\right) \ , \tag{A27}$$

$$\Omega_1 = 4\left|\sin\frac{q}{2}\right|;\quad \Omega_2 = 4\left|\cos\frac{q}{2}\right| \ , \tag{A28}$$

where $\Sigma = (M^\perp + M^z)/[(LS^\perp, LS^\perp)+(LS^z, LS^z)]$, and where J_0 and J_1 are the Bessel functions. This is the result of Reiter and Sjölander, which they have discussed extensively.[13] For $h=2\sqrt{2}$, the spins make an angle $\frac{1}{4}\pi$ with the z axis in the ground state. For the transverse component we have

$$\Sigma^\perp_{q^*}(t) = -4T(1+\cos q)[I(c,u)+I(-c,v)]/(3-\cos q) \ , \tag{A29}$$

$$I(c,u) = \left[\left((1+c)(1+c-c^2)+2(1+c)(2-c)\frac{d^2}{du^2}\right)^2-2(1+c)\left(c+2\frac{d^2}{du^2}\right)^2\frac{d^2}{du^2}\right]J_0(u) \ , \tag{A30}$$

$$c = \cos\frac{q}{2};\quad u = \left(2h\cos\frac{q}{4}\right)t;\quad v = \left(2h\sin\frac{q}{4}\right)t;\quad h = 2\sqrt{2} \ , \tag{A31}$$

and the longitudinal component reads

$$\Sigma^z_q(t) = -2T(1-\cos q)[K(c,u)+K(-c,v)] \ , \tag{A32}$$

$$K(c,u) = \left(2-c^2+2(1-c)\frac{d^2}{du^2}\right)^2 J_0(u) \ . \tag{A33}$$

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