Theory of exchange narrowing in one and two dimensions

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We show that the theoretical approximations in the Kubo and Tomita theory of electron paramagnetic resonance are such as to make its application to one- and two-dimensional systems dubious. A self-consistent theory based upon the independent-mode approximation, that does not have the defects of the Kubo and Tomita theory, is described, and line shapes are calculated for the strongly narrowed limit. Both theories are in significant disagreement with the experimental results on tetramethyl manganese chloride.

I. INTRODUCTION

Recent EPR experiments[1] in one-dimensional exchange-coupled systems have been interpreted in terms of the Kubo and Tomita[2] (KT) theory of exchange narrowing, which has been applied successfully for many years to three-dimensional systems. We show here that the KT theory is fundamentally inadequate to describe these experiments, and discuss an alternative theory. The difficulty arises from the fact that in one dimension the dominant contribution to the "mass" operator (or "correlation function of the fluctuating torques") in the spectral density of the $q=0$ magnetization mode comes from modes with wave vectors in the neighborhood of $q=0$, where the dipole interaction strongly modifies the spectrum of the fluctuations. The KT prescription for calculating the spectral density is based upon second-order perturbation theory, and treats the fluctuations responsible for the line broadening as though they were unaffected by the dipolar interaction. Specifically, the KT theory represents the correlation function for the $q=0$ mode, at infinite temperature,[3] as

$$\Phi(t)=\langle S^-(0,t)S^+(0,0)\rangle = \exp\left(i\omega_0 t-\int_0^t (t-\tau)\Psi(\tau)\,d\tau\right) \times\langle S^-(0,0)S^+(0,0)\rangle\ , \quad (1)$$

where $\langle A\rangle \equiv \mathrm{Tr}A$, ω_0 is the resonance frequency in the absence of a perturbation leading to broadening, and $\Psi(\tau)$ is a correlation function describing the effects of the perturbation. Equation (1) is completely general as long as $\Psi(\tau)$ is not specified. The KT prescription for obtaining $\Psi(\tau)$ is to expand the right- and left-hand sides of (1) in powers of the perturbation, and identify Ψ with the second-order perturbation term. For the Heisenberg Hamiltonian H, where the perturbation is due to the dipole Hamiltonian H_D, $\Psi(\tau)$ is given by

$$\Psi(\tau)=\langle[H_D(\tau),S^-(0,0)] \times[H_D,S^+(0,0)]\rangle/\tfrac{2}{3}S(S+1)\ , \quad (2a)$$

where $H_D(t)=\exp[(i/\hbar)(H+H_Z)t]H_D\exp[-(i/\hbar)(H+H_Z)t]$, H_Z is the Zeeman Hamiltonian. In the case that H_D commutes with H_Z, i.e., neglecting the nonsecular part of H_D, (2a) can be written as

$$\Psi(\tau)=\langle[H_D,S^-(0,0)]e^{-i\mathcal{L}\tau} \times[H_D,S^+(0,0)]\rangle/\tfrac{2}{3}S(S+1) \quad (2b)$$

where $\mathcal{L}$ is the Liouville operator for H, $\mathcal{L}[A]=(1/\hbar)[H,A]$. Equation (1) is equivalent to (3):

$$\frac{d\Phi(t)}{dt}=i\omega_0\Phi(t)-\int_0^t \Psi(\tau)d\tau\,\Phi(t)\ . \quad (3)$$

An exact expression for the generalization of Φ to nonzero q, $\langle S^-(q,t)S^+(-q,0)\rangle$, which we will denote by $\Sigma^+(q,t)$ (Ref. 4), can be derived, that is similar to (3)

$$\frac{d\Sigma^+(q,t)}{dt}=i\omega_0\Sigma^+(q,t) - \int_0^t \tilde{\Psi}_q^+(t-\tau)\Sigma^+(q\tau)\,d\tau\ , \quad (4)$$

where

$$\tilde{\Psi}_q^+(\tau)=\langle[H+H_D,S^-(q,0)]e^{-i\mathcal{L}'t} \times[H+H_D,S^+(-q,0)]\rangle/\tfrac{2}{3}S(S+1)\ , \quad (5)$$

$\mathcal{L}'$ is a modified Liouville operator[4] for H_Z+H+H_D, and H_Z is the Zeeman Hamiltonian. Equation (5) is valid for the full dipole Hamiltonian, but we will only consider the effect of the secular terms in this work. The lowest-order expansion of $e^{-i\omega_0 t}\tilde{\Psi}(t)$ in powers of H_D agrees with $\Psi(t)$, since $[H,S^+(0,0)]=0$. This expansion is not uniform in the time variable, since for any nonzero value of ω_D, the long-

time behavior of $\tilde{\Psi}_0(t)$ is very different from the long-time behavior of $\Psi(t)$. The difference arises from the presence of the dipole Hamiltonian in the propagator in Eq. (5), which yields a finite lifetime for the modes in the vicinity of $q=0$. In three dimensions, this difference is unimportant, since the relative weight of the modes in the vicinity of $q=0$ is negligible. $\Psi(t)$ decays essentially to zero in a time that is characteristic of the short-wavelength fluctuations, i.e., $1/\omega_{ex}=\phi(t)$, $[\Sigma(0,t)]$ decays in a time $\omega_{ex}^2/\omega_D \gg 1/\omega_{ex}$, where ω_D is a characteristic frequency for the dipole Hamiltonian. In the limit that $\omega_{ex}/\omega_D \to \infty$, we can neglect the variation of $\Sigma(t)$ in the integral in Eq. (4) and the equation becomes identical to Eq. (3), with the linewidth given by $\int_0^\infty \Psi(\tau)\,d\tau$ and $\int_0^\infty e^{-i\omega_0\tau}\tilde{\Psi}(\tau)\,d\tau$ in Eqs. (3) and (4), respectively.

For finite ω_{ex}/ω_D these numbers are not the same but they do approach each other in the limit that this ratio diverges. In one and two dimensions, the contribution of the modes near $q=0$ is more important, dominating the decay completely in one dimension. The time scales of Φ and Ψ are comparable so that the two formulations are no longer equivalent. Equation (5) remains exact, but there is no reason to believe that Eq. (3) remains valid. The higher-order cumulants that were neglected in defining $\Psi(\tau)$ can give contributions that are comparable (in powers of ω_D/ω_E) to the terms that are formally of lowest order in ω_D. This is a general feature of the motional narrowing phenomenon.

If we consider the physical situation as the decay time of $\Psi(\tau)$ increases, with $\Psi(0)$ a constant, it is natural to divide the problem into a "slow" and a "fast" regime according to whether the decay time, τ_c of Ψ is smaller or larger than the decay time of Φ. The inadequacy of the KT theory in the slow regime is best illustrated by the fact that in the extreme slow limit, i.e., the random fields responsible for the linewidth are static, it yields a Gaussian line shape independent of the distribution of the fields, whereas in fact, the line shape depends on the probability distribution of the fields and in the simplest case is simply the Fourier transform of that distribution.[5] This inadequacy appears to have been appreciated in Ref. 2. The details of the line shape in this regime are reflected in the higher-order cumulants, which have been neglected.

In the extreme fast regime, the line shape will always be Lorentzian. The higher-order cumulants give negligible corrections, since they contain additional time integrations that reduce the contribution of these terms by factors of $\tau_c\Psi(0)^{1/2}$. The case that the fluctuations are produced by the modes of the Heisenberg system is in the fast regime for three dimensions, and as we shall see also for two dimensions. In one dimension, the resultant linewidth, Γ, satisfies $\Gamma\tau_c \cong 1$ and hence this case is on the boundary between the slow and fast regime.

II. ONE DIMENSION

Neither (2) nor (5) is exactly calculable, but both may be calculated approximately using the kinetic theory developed by Reiter,[6] and Barreto and Reiter.[5] This will be done in a later work. In order to obtain relatively simple qualitative results that will make clear the points above, we will make an independent-mode approximation to evaluate (2b) and (5). In one dimension, we will consider only the case that the magnetic field is along the chain axis, in which case only the secular terms appear in H_D. The operator $[H_D, S^-(0)]$ is of the form $\sum F(q)S^z(q)S^-(-q)$. The independent-mode approximation for the correlation function appearing in (2) and (5) is shown in Eq. (6) (see Ref. 6 for a discussion of the limitations of this approximation). [We note that although the time dependence of the correlation function on the left of Eq. (6) is determined by $\mathcal{L}'$, the time dependence of the correlation functions on the right of Eq. (6) is properly taken to arise from $\mathcal{L}$.]

$$\langle S^z(q,t)S^-(-q,t)S^z(q',0)S^+(-q',0)\rangle \cong \delta_{qq'}\langle S^z(q,t)S^z(-q,0)\rangle\langle S^-(q,t)S^+(-q,0)\rangle\,. \tag{6}$$

We will use the long-wavelength approximations for the dependence of $\langle S^\alpha(q,t)S^\alpha(-q)\rangle$ throughout the zone in evaluating Ψ and $\tilde{\Psi}$. This approximation is asymptotically correct as the ratio of ω_D/ω_E approaches zero, in the sense that the correction to the spectral density at any finite but fixed multiple of the line width coming from a region in the zone outside of some fixed and arbitrarily small interval about $q=0$ can be made vanishingly small. In (2) the long wavelength behavior is diffusive, so that

$$\langle S^z(q,t)S^z(-q)\rangle = \tfrac{1}{2}\langle S^-(q,t)S^+(-q)\rangle = \tfrac{1}{3}S(S+1)\,e^{-Dq^2t}\,.$$

In (5), because of the inclusion of the dipole terms in the propagator the long wavelength behavior is more complicated. However we can show from (4) that the form of $\Sigma^\alpha(q,z) \equiv \int_0^\infty e^{izt}\Sigma^\alpha(q,t)$ is

$$\Sigma^\alpha(q,z) = i\tfrac{2}{3}S(S+1)[z+(\alpha\omega_0)+i\Gamma^\alpha(q,z)]^{-1}\,, \tag{7}$$

where

$$\Gamma^\alpha(q,z) = \int_0^\infty e^{izt}\tilde{\Psi}_q^\alpha(t)\,dt \tag{8}$$

and $\alpha = \pm$ or 0. $\Psi_q^0(t)$ differs from (5) only in the replacement of $S^+(q,0)$ by $S^z(q,0)$. From Eq. (5) it follows that for small q,

$$\Gamma^\alpha(q,z) = \Gamma^\alpha(0,z) + D^\alpha(z)q^2\,, \tag{9}$$

the first term being the result one would obtain if the Heisenberg Hamiltonian were omitted from the

commutator in Eq. (5). $D^{\alpha}(z)$ differs for $\alpha=\pm,0$, but the difference vanishes linearly as $\omega_0/\omega_E \to 0$ so that we will, with negligible error in the case of tetramethyl manganese chloride (TMMC), replace $D^{\alpha}(z)$ by $D(z)$, its value in the Heisenberg system. For the frequencies of interest, i.e., $\omega-\omega_0 \cong \Gamma(0,0)$, there will be negligible variation of $D(z)$ and we may replace it by its value at zero frequency, so that we have finally

$$\Gamma^{\pm}(q,z)=\Gamma^{\pm}(0,z)+Dq^2\ ,$$
$$\Gamma^{0}(q,z)=Dq^2\ . \tag{10}$$

The approximations that were made in obtaining (10) are all strictly valid in the limit $\omega_E \to \infty$, ω_0, and ω_D finite. Using the independent-mode approximation, $\Gamma^{\pm}(0,z)$ can be determined self-consistently. This is the only approximation needed to solve the problem in the above limit. We have, using (6) in Eqs. (2a) and (5), and taking the long-wavelength limit for the correlation functions,

$$\Psi(\tau)=\tfrac{3}{4}S(S+1)\omega_D^2\sum_q F(q)^2 e^{-2Dq^2t}\ , \tag{11a}$$

$$\tilde{\Psi}_0(\tau)=\tfrac{9}{8}\omega_0^2\sum_q F(q)^2 e^{-Dq^2t}\Sigma^{+}(q,t)\ , \tag{11b}$$

where

$$F(q)=\sum_j \cos(qr_{ij})(1-3\cos^2\theta_{ij})(r_{ij}/a)^3,\quad \omega_D=g\mu_B a^{-3}.$$

We can approximate $F(q)$ by $F(0)$, again with a vanishing error as $\omega_E \to \infty$, and replace the upper limit on the sums in Eqs. (11) by infinity. From Eqs. (11a) and (1) we obtain

$$\Phi(t)=\tfrac{2}{3}S(S+1)\,e^{i\omega_0 t}\,e^{-(t/t_0)^{3/2}}\ , \tag{12}$$

where $1/t_0=\Omega(2\Omega a^2/9\pi D)^{1/3}$, $\Omega^2=\tfrac{3}{4}S(S+1)\omega_D^2F(0)^2$. From Eqs. (7)–(9) and (11b) we obtain, in terms of the function $\Gamma^*(z)\equiv\Gamma^{+}(0,z-\omega_0)$, an equation that determines Γ^{α} self-consistently:

$$\Gamma^*(z)=i\Omega^2\sum_q [z+2iDq^2+i\Gamma^*(z+iDq^2)]^{-1}\ . \tag{13}$$

The solution of this integral equation [Eq. (13)] provides an exact solution for $\Sigma^{+}(0,z)$ within the independent-mode approximation, i.e.,

$$\Sigma^{+}(0,z)=i\tfrac{2}{3}S(S+1)[z+\omega_0+i\Gamma^*(z+\omega_0)]^{-1}\ . \tag{14}$$

We shall not solve this equation exactly, but will obtain an approximate solution by ignoring the q dependence of Γ^* within the summation on the right-hand side of Eq. (13). In this case, the sum in Eq. (13) can be done explicitly and we obtain the result that $\Gamma^*(z)$ must satisfy

$$\Gamma^*(z)=\Omega^2(2Dq_0^2)^{-1/2}[-iz+\Gamma^*(z)]^{-1/2}$$
$$\times\tan^{-1}\left(1+\frac{2iDq_0^2}{z+i\Gamma^*(z)}\right), \tag{15}$$

where $q_0=\pi/a$. For values of z on the order of $\Gamma^*(0)$, the argument of $\tan^{-1}$ in Eq. (15) diverges as $\omega_E/\omega_D \to \infty$, and hence the equation simplifies to

$$\Gamma^*(z)=\Omega^2(\pi/2)(2Dq_0^2)^{-1/2}[-iz+\Gamma^*(z)]^{-1/2}\ . \tag{16}$$

The frequency scale for the spectral density, Eq. (14), is set by $\Gamma^*(0)$ which is

$$\Gamma^*(0)=\Omega(\Omega a^2/8D)^{1/3}\ , \tag{17}$$

It should be noted from (15) that $\tilde{\Psi}_0(\tau)$ will decay on a time scale of $\Gamma^*(0)$, which is the same time scale as the response function, $\Sigma_0(t)$, as previously noted. In terms of the dimensionless quantity

$$\rho(z)=\Gamma^*(z)/\Gamma^*(0),\quad z'=z/\Gamma^*(0)\ , \tag{18}$$

we have

$$\rho(z)^2[-iz'+\rho(z)]=1\ . \tag{19}$$

The curve shown in Fig. 1 was obtained by solving Eq. (19) numerically and substituting the result for $\rho(z)$ [$=\Gamma^*(z)$ when $\Gamma^*(0)=1$] into Eq. (14). We have compared it with the KT result for the same value of the physical parameters. The value of $1/t_0$ in the case that $\Gamma(0)=1$ is $(16/9\pi)^{1/3}=0.825$. The corrections to this approximation may be estimated by using the Taylor series expansion of $\rho(z)$,

$$\rho(z)=1-\tfrac{1}{3}iz-\tfrac{1}{9}z^2+\cdots\ . \tag{20}$$

From (14), we expect that about half the contribution to the integral in Eq. (13) will come from values of q such that $D(q^2)<\Gamma$, so that it is reasonable to use Eq. (20) in Eq. (12). If we keep only the linear term in Eq. (20) we see that the sole effect of keeping the dependence upon q in the integration for $z=0$ is to replace $2D$ by $\frac{7}{3}D$. Since D

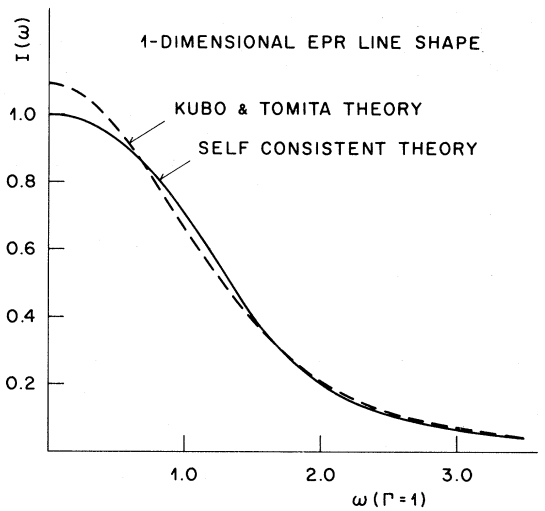

FIG. 1. Comparison of theories for the same value of the physical parameters.

enters Eq. (16) to the $\frac{1}{3}$ power, this correction would decrease $\Gamma^*(0)$ by about 6%. For larger values of z, Γ^* is less important in the integral, which becomes dominated by the iDq^2 term, and we would expect the corrections to be even less. We note that approximating $\Gamma^*(z)$ by $\Gamma^*(0)$ under the integral yields an unphysical (i. e. unobserved) maxima at finite frequencies for the line-shape function, $\mathrm{Re}\Sigma(0, \omega + i\epsilon)$.

It can be seen from Fig. 1 that the difference between the self-consistent theory and the KT theory is small (< 10%) even at zero frequency. The corrections to the approximate solution are in a direction that improves the agreement. At high frequencies, both theories yield

$$\mathrm{Re}\Sigma(0, \omega + i\epsilon) = \Gamma^*(0)/\sqrt{2}\,\omega^{5/2} + O(1/\omega^4) \tag{21}$$

the differences between the two results appearing in the coefficient of the $1/\omega^4$ term in the high-frequency expansion. The KT theory, despite the theoretical objections to its validity in the one-dimensional case, gives a result that agrees well with the self-consistent theory (within the independent-mode approximation). The major discrepancy between the two theories is in the long-time behavior, since the KT theory predicts

$$\lim_{t\to\infty} \Sigma^+(0, t) \propto e^{-(t/t_0)^{3/2}}, \tag{22}$$

whereas the self-consistent theory yields

$$\lim_{t\to\infty} \Sigma^+(0, t) \propto e^{-\Gamma^*(0)t}. \tag{23}$$

It is evident that the inadequacies of the KT theory, the truncation of the cumulant expansion, and the inclusion of the time dependence of the fluctuations in the form given in Eq. (3) rather than Eq. (4), have compensated each other to a large extent.

Neither theory gives a satisfactory description of the experiments on tetramethyl manganese chloride. In Table I we have compared the experimental and theoretical values for the full width at half-maximum. The lattice parameter ($c = 6.49$ Å)[8] and hence Ω are known precisely, so that the major uncertainty in the theoretical expressions is the value of D for the one-dimensional Heisenberg chain. The exchange constants are known accurately [$H = -\frac{1}{2}\sum_{i,j} V_{ij}\vec{S}_i \cdot \vec{S}_j$, $V_{i,i+1} = V = (-14.2 \pm 1.2)$ °K][8] so that the major uncertainty in the calculation of $\Gamma^*(0)$ is the relationship between D and V, which we take to be $D = 1.33SVa^2$, the value inferred from numerical simulations.[9] There is considerable uncertainty as to the value of the constant appearing in this relationship, and theoretical calculations differ by a factor of 2 between themselves.[10] We will take this factor of 2 to be the uncertainty in the value of D, and it is this factor which produces the theoretical uncertainties shown in Table I. It is evident that the experimental and measured

TABLE I. Comparison of theoretical and experimental linewidths. Experimental value is the result of recent high-sensitivity measurements by J. P. Boucher and differs slightly from the value cited in earlier work (1360 G) (Ref. 1).

	Experimental	Self-consistent	Kubo and Tomita
Linewidth (G)	1300	2400 ± 380	2220 ± 350

values for the line width differ by far more than the uncertainty in the theory, and hence that some of the assumptions are questionable. We have assumed that an accurate model for the system was an isotropic Heisenberg linear chain with dipolar interactions, and we have made some approximations, given that model, to obtain a solution. Since the experimental line width is smaller than the theoretical value, there are strong restrictions on any corrections to the model. Interchain exchange would lower the line width by increasing the effective bandwidth for the fluctuations. The ratio of the intra-chain (J) to inter-chain (J_1) coupling constants can be estimated from the magnitude of the ordering temperature.[11] It is found that $(J_1/J)^{1/2} \simeq 0.63(KT_n/KT_\omega)$ where KT_n is the actual ordering temperature (1.1 °K) and KT_ω the Weiss Néel temperature (78 °K), so that $J_1/J \leq 2\times 10^{-3}$. This is far too small to reduce the theoretical line width significantly. This is further demonstrated by the line shape itself. If the inter-chain coupling were significant, the line shape would tend toward being Lorentzian. In Fig. 2 we have compared the theoretical line shapes, with the width as an adjustable parameter, to the experimental data. The fit is rather good, and furthermore, the lineshape is far from being Lorentzian.

Additional terms in the Hamiltonian that did not commute with $S^z(0)$ could reduce the line width if they were of such a sign as to effectively cancel part of the dipole interaction. However, the dipole interaction has a characteristic dependence upon the angle of the magnetic field with the crystal axis. If only the secular terms in the Hamiltonian are considered the line width varies as $\omega_D^{4/3}|1 - 3\cos^2\theta|^{4/3}$, where θ is the angle between the chain axis and the magnetic field. If there are additional terms, the variation would be $|\omega_D(1 - 3\cos^2\theta) - A|^{4/3}$ where A is some effective interaction strength. To lower the value of the line width by the factor of 1.7 needed to get agreement with the experiment at $\theta = 0$ would require such a large value of A that the angular dependence would be drastically modified. The experiments show, however,[1] that the variation in line width is very close to $|1 - 3\cos^2\theta|^{4/3}$. It seems extremely unlikely that a narrowing mechanism can be found that would modify the model consistent with the constraints imposed by the

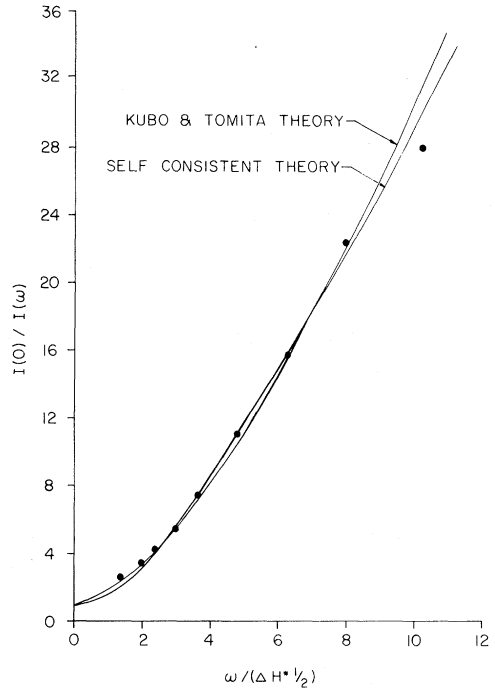

FIG. 2. Comparison of line shapes with adjusted value for linewidth. t_0 adjusted to give experimental half-width, Γ adjusted to give overall fit. ΔH is experimental half-width, $\Delta H = 1.15\,\Gamma$ for value of Γ used to fit data.

existing data.

The approximations made in obtaining a solution from the model are the independent-mode approximation, and the approximation needed to solve Eq. (13). As we have indicated, the latter approximation produces only a small error (< 10%). The errors made by making the independent-mode approximation are unknown, but could easily be of the order of magnitude needed to give agreement with the theory. We regard this as the most likely source of error in the present calculation. A calculation based upon a solution of the kinetic equations[6,7] for the four-point correlation functions is required to resolve this question. We also point out that there is no reason to suspect that the corrections to the independent-mode approximation are not significant in two and three dimensions also, although numerical calculations of the EPR width within this approximation are in good agreement with the experiments,[12] in three dimensions.

III. TWO DIMENSIONS

In two dimensions, the nonsecular terms are present in the Hamiltonian for all orientations of the field, and there are significant contributions to the linewidth from fluctuation modes throughout the zone, even in the limit that $\omega_E \to \infty$. As a consequence, a calculation of the line width in a two-dimensional system requires that the terms $F(q)$ be kept under the summation to achieve accurate results, and that the spectral density of the fluctuations be known for all q, i.e., the spectrum cannot be simply characterized by a diffusion constant. We are primarily interested in a comparison between the KT and self-consistent theories and will not attempt an accurate calculation of the linewidth. In addition to the independent-mode approximation we will assume that the diffusive form of the correlation functions can be used throughout the entire zone, that the zone can be replaced by a circle of the same area, and that $F(q)$ can be replaced by $F(0)$. It is also the case that if $\ln(\omega_E/\omega_0) \ll \ln(\omega_E/\omega_D)$, which we will assume, then the nonsecular terms are negligible compared to the secular terms and can be neglected. With these simplifying assumptions, proceeding exactly as for the one-dimensional case, we obtain, for times such that $8\pi Dt/a^2 \gg 1$, that the KT theory predicts

$$\Phi(t) = e^{i\omega_0 t}\, e\left(\frac{\Omega^2 a^2}{8\pi D}\right)\left(\frac{t\ln t}{t_0} + \frac{a^2}{8\pi D}\right), \tag{24}$$

where

$$\Omega^2 = \tfrac{3}{4} S(S+1)\omega_D^2 F(0)^2$$

and

$$\ln t_0 = 1 + \int_0^\infty e^{-t} \ln t\, dt + \ln a^2/8\pi D\ .$$

If we define R as $8\pi D/\Omega a^2$, then as $R \to \infty$, the line shape that results from (24) is asymptotically Lorentzian, with a linewidth, $1/t^* = \Omega^2 a^2/8\pi D \ln R^2$. It should be noted that the factor $\Omega^2 a^2/8\pi D$ is just the result one would expect in the fast limit, so that the effect of the long wavelength fluctuation is entirely included in the factor $\ln R^2$.

The self-consistent theory is simpler in two dimensions than in one dimension because the result is less sensitive to the frequency dependence of $\Gamma^*(q,z)$. We will therefore assume that

$$\begin{aligned} \langle S^-(q,t) S^+(-q)\rangle &= \tfrac{2}{3} S(S+1_0) e^{-(\Gamma + Dq^2)t} e^{i\omega_0 t}\ , \\ \langle S^z(q,t) S^z(-q)\rangle &= \tfrac{1}{3} S(S+1) e^{-Dq^2 t}\ , \end{aligned} \tag{25}$$

for the purpose of calculating $\tilde{\Psi}_0(t)$. We find, using Eqs. (5), (6), (8), and (25), that

$$\Gamma^*(\omega) = (\Omega^2 a^2/8\pi D)\ln(1 + 4\pi D/a^2(\Gamma - i\omega))\ , \tag{26}$$

Identifying $\Gamma^*(0)$ with Γ, we obtain the self-consistent equation for Γ

$$\Gamma = \lambda \Omega^2 a^2/8\pi D\ , \tag{27a}$$

$$\lambda = \ln(1 + R^2/2\lambda)\ . \tag{27b}$$

In terms of the dimensionless frequency $\omega' = (\omega + \omega_D)/\Gamma$,

$$\Gamma\Sigma^+(0,\omega) = i\tfrac{2}{3}S(S+1) \times[\omega' + i - i\lambda^{-1}\ln(1 - i\omega')]^{-1} . \quad (28)$$

We have assumed $R^2 \gg 1$ in order to simplify (28) slightly.

The solution of Eq. (27a) for λ, as $R^2 \to \infty$ is $\lambda = \ln R^2$. From (28) we see that the line shape becomes a Lorentzian in this limit, and that Γ agrees with the KT result, i.e., $\Gamma = 1/t^*$. For finite values of R, neither line shape will be Lorentzian, nor will the linewidths agree. We have compared the two theories for finite values of R in Fig. 3.

Richards and Salomon[13] have used the KT theory, together with a phenomenological form for $\Psi(t)$, to calculate the linewidths at $T = \infty$, in K_2MnF_4, a two-dimensional antiferromagnet. They obtain good agreement with both the linewidth and its angular variation. For the values in $\ln R^2$ that are appropriate for that system there would be significant differences with the self-consistent theory. The neglect of the variation of $F(q)$ in the present work precludes any direct comparison with experiments as does the neglect of the nonsecular terms. The differences in the line width predicted by the two theories will be much smaller than the changes produced by including these effects in the calculation. We regard the accuracy of the fit by Richards and Salomon to be due to a judicious choice of the phenomenological form for $\Psi(t)$. Their work does not resolve the question of whether a model that includes only dipolar and exchange interactions gives a satisfactory description of the experimental results, within the independent-mode approximation.

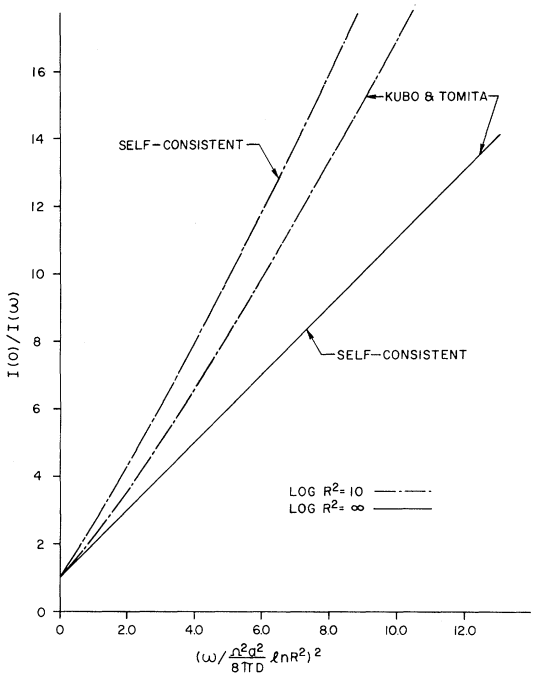

FIG. 3. Comparison of simplified model line shapes for two-dimensional systems. $\ln R^2 \to \infty$ is equivalent to $\omega_D/\omega_E \to 0$.

IV. CONCLUSIONS

The criteria for the validity of the KT theory are that (a) the fluctuation modes producing the narrowing decay rapidly compared to the linewidth of the mode being considered, and (b) the fluctuation modes are relatively unaffected by the interaction that couples them to the mode of interest. In two dimensions, this is the case, since the modes with wave vector q such that $Dq^2 \gg \Gamma$ dominate the contribution to $\tilde{\Psi}_0(t)$. As a consequence, the line shape is Lorentzian, the effect of the system being two dimensional showing up only in the linewidth varying as $(\omega_D^2/\omega_E)\ln(\omega_E/\omega_D)$ rather than ω_D^2/ω_E, as in three dimensions.

In one dimension, the dominant contribution to $\tilde{\Psi}_0(t)$ arises from modes for which $Dq^2 \lesssim \Gamma$, and hence both criteria fail. That the KT theory is not grossly in error, within the independent-mode approximation, even for this case, indicates that the errors introduced by violating (a) and (b) have canceled each other to a large extent.

The only approximation needed to reduce the exchange-narrowing problem to a closed-form solution Eq. (13), in the strongly narrowed limit, i.e. $\omega_E \to \infty$, ω_D, and ω_0 fixed, is the independent-mode approximation. Since the discrepancy between our self-consistent approximate solution and the exact self-consistent solution based upon the independent-mode approximation is much smaller than the discrepancy with the experimental linewidth in tetramethyl manganese chloride, we conclude that the independent-mode approximation is unable to provide a quantitative microscopic calculation of the linewidth, and is the primary source of error in the existing theory. This may also be the case in two and three dimensions.

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