Thermal-neutron double scattering: Critical magnetic scattering from ferromagnets

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A quantum-mechanical formulation for multiple scattering of thermal neutrons is presented and applied to double scattering. The contribution of double scattering to the cross section for quasielastic magnetic scattering of neutrons from ferromagnets slightly above their critical temperature is calculated for relevant experimental parameters. The effect is to alter the usual Lorentzian line-shape dependence on neutron wave-vector transfer. Comparison with corresponding deviations in line shape resulting from the use of a modified form of Ornstein-Zernike spin correlations within the framework of single-scattering theory leads to values for the parameter η of the modified correlations which reproduce the effect of double scattering.

I. INTRODUCTION

Thermal neutron scattering is one of the most powerful experimental tools for investigating the atomic structure and dynamics of materials. The primary quantity of interest in slow neutron scattering by macroscopic materials is the dynamic structure factor $S(\vec{k},\omega)$ which gives, within a normalization factor, the first-Born-approximation cross section for a neutron to be scattered with momentum and energy transfers to the material of $\hbar\vec{k}$ and $\hbar\omega$, respectively. For magnetic scattering of neutrons from a magnetic material, $S(\vec{k},\omega)$ is the Fourier transform of the space- and time-dependent spin-spin correlations of the scattering system. The dynamic structure factor can be obtained theoretically from microscopic models of the scattering system.

Experimentally observed scattering intensity is linearly proportional to $S(\vec{k},\omega)$, but only provided that the sample is small enough and neutron wavelength short enough that multiple scatterings are negligible. However, while multiple scatterings may be small, they are often not negligible, and the observed scattering intensity is not simply proportional to $S(\vec{k},\omega)$.

In this paper, we present a general quantum-mechanical formulation for the thermal-neutron-scattering cross section which depends on higher-order correlations between target variables beyond the bilinear correlations involved in $S(\vec{k},\omega)$. For experimental conditions such that double scattering is a small, but significant effect, and higher-order scattering is negligible, the scattered intensity depends on quadrilinear correlations between target variables, which can be well approximated by products of bilinear correlations. The double scattering can then be expressed in terms of products of two dynamic structure factors. Under the special conditions of small angle, thermal-neutron scattering slightly above the critical temperature, this decomposition becomes virtually exact. We consider the case of quasielastic magnetic scattering from ferromagnets slightly above their Curie temperature in detail, and derive a cross section including double scattering in a form useful for analysis of experimental data.

Conventionally, experiments in this area have usually been interpreted by assuming multiple scattering effects to be negligible, implying that the observed scattering is proportional to $S(\vec{k},\omega)$. Observed discrepancies between experimental results so interpreted and theoretical predictions for $S(\vec{k},\omega)$ have sometimes been taken as indicating a failure of the Ornstein-Zernike (OZ) theory[1] of spin-spin correlations. A modified theory[2] has been proposed to remove this discrepancy. The modified OZ theory of spin correlations, inserted in the first Born approximation for scattering, produces a deviation from the usual Lorentzian line shape of quasielastically scattered neutrons as a function of wave-vector transfer. A similar qualitative effect is produced with double scattering included, but with no modification of the OZ theory. So the presence of even a small amount of double scattering in an experiment, when interpreted within the framework of a single-scattering theory, can appear to require a modified OZ theory. We quantitatively estimate this effect for typical experimental parameters.

In Sec. II we present a general expression for the transition probability for a probe particle interacting with a target, which contains target dynamical variables only in the form of averages over initial target states. The first term in a series expansion of the general expression is shown to yield the time-dependent correlation form for the single-scattering cross section first given by Van Hove.[3]

Section III treats the next relevant term of the

general expression derived in Sec. II. This term contains the double-scattering contribution as well as negligible refraction and extinction effects, and contains the average over target initial states of products of four target variables. Approximating this quadrilinear correlation by products of bilinear correlations, the magnetic double-scattering cross section is expressed in terms of products of dynamic structure factors, Eq. (3.24).

Section IV considers the double-scattering contribution to neutron magnetic scattering from ferromagnets slightly above their Curie temperature, and results in a simple formula for the cross section, Eq. (4.42). Numerical estimates of the double-scattering contribution are made, using relevant experimental parameters. Values are given for the parameter η of the modified OZ theory which, when used in a first-Born-scattering approximation, reproduce the effect of double scattering.

II. GENERAL THEORY

In this section a general expression for transition probabilities is obtained, Eq. (2.17), that can be applied to the scattering of a probe by a target. The practical utility of the formulation given here is that target variables appear only in the form of averages over initial target states. Connection is made with Van Hove's expression for the first Born approximation for scattering.

Consider a system R, with Hamiltonian H_R, representing the target, which we will take as a ferromagnetic crystal in Sec. IV, in contact with another system S, with Hamiltonian H_S, representing the probe, the neutron, so that $R+S$ form a closed system. The total Hamiltonian is given by

$$H = H_R + H_S + \phi(A, B), \tag{2.1}$$

where ϕ represents the interaction between R and S, A represents some dynamical operators of S, and B represents some dynamical operators of R. The density operator for the total system at time t is determined in terms of the density operator at time t_0 and is given by

$$\rho(t) = e^{-iH(t-t_0)/\hbar}\rho(t_0)e^{iH(t-t_0)/\hbar} . \tag{2.2}$$

The reduced density operator for S is obtained from Eq. (2.2) by tracing over R states,

$$\rho_S(t) = \mathrm{Tr}_R\rho(t) \tag{2.3}$$

Now, assuming that the interaction is turned on at t_0, we can write the total density operator at t_0 as

$$\rho(t_0) = \rho_R(t_0)\rho_S(t_0) \tag{2.4}$$

For simplicity, we assume that S is initially in an eigenstate, $|S_i\rangle$, of H_S,

$$\rho_S(t_0) = |S_i\rangle\langle S_i| \tag{2.5}$$

while in terms of eigenstates, $|R_i\rangle$, of H_R,

$$\rho_R(t_0) = \sum_{R_i} P_{R_i} |R_i\rangle\langle R_i| , \tag{2.6}$$

where P_{R_i} is the probability of the occurrence of $|R_i\rangle$. Therefore, the reduced density operator, Eq. (2.3), can be written as

$$\rho_S(t) = \sum_{R_f}\sum_{R_i} P_{R_i}\langle R_f|e^{-iH(t-t_0)/\hbar}|R_i\rangle|S_i\rangle\langle S_i|\langle R_i|e^{iH(t-t_0)/\hbar}|R_f\rangle . \tag{2.7}$$

Using the appropriate time ordering operators, we can write

$$e^{-iH(t-t_0)/\hbar} = e^{-i(H_R+H_S)t/\hbar}\left[T_+ \exp\left(-\frac{i}{\hbar}\int_{t_0}^{t} dt'\,\phi(t')\right)\right]e^{i(H_R+H_S)t_0/\hbar} , \tag{2.8}$$

$$e^{iH(t-t_0)/\hbar} = e^{-i(H_R+H_S)t_0/\hbar}\left[T_- \exp\left(\frac{i}{\hbar}\int_{t_0}^{t} dt'\,\phi(t')\right)\right]e^{i(H_R+H_S)t/\hbar} , \tag{2.9}$$

where $\phi(t)$ is in the interaction picture and the operator T_+ (T_-) time orders products of the $\phi(t)$'s with the earlier (later) time to the right. Now, the probability of finding S in eigenstate $|S_f\rangle$ at time t is given by $\langle S_f|\rho_S(t)|S_f\rangle$, therefore from Eqs. (2.7), (2.8), and (2.9) we have,

$$\langle S_f|\rho_s(t)|S_f\rangle = \sum_{R_i} P_{R_i}\langle S_i|\langle R_i|T_-\exp\left(\frac{i}{\hbar}\int_{t_0}^{t} dt'\,\phi(t')\right)|S_f\rangle\langle S_f|T_+\exp\left(-\frac{i}{\hbar}\int_{t_0}^{t} dt'\,\phi(t')\right)|R_i\rangle|S_i\rangle \tag{2.10}$$

where closure has been used:

$$\sum_{R_f}|R_f\rangle\langle R_f| = 1.$$

To proceed further in obtaining the reduced density matrix elements in terms of target averages, we introduce the subscripts 1, 2 on operators and state vectors of S, write Eq. (2.10) as

$$\langle S_{f_2}|\rho_s(t)|S_{f_1}\rangle=\sum_{R_i}P_{R_i}\langle S_{i_1}|\langle R_i|T_-\exp\left(\frac{i}{\hbar}\int_{t_0}^{t}dt'\,\phi_1(t')\right)|S_{f_1}\rangle\langle S_{f_2}|T_+\exp\left(-\frac{i}{\hbar}\int_{t_0}^{t}dt'\,\phi_2(t')\right)|R_i\rangle|S_{i_2}\rangle \tag{2.11}$$

and introduce a new position ordering operator, P_{12}, which moves all operators and state vectors with subscript 1 to the left. Introducing this operator into Eq. (2.11) allows us to freely move all operators and state vectors with different subscripts past each other since P_{12} will restore the correct order:

$$\langle S_{f_2}|\rho_s(t)|S_{f_1}\rangle=P_{12}\langle S_{f_2}|\langle S_{i_1}|\left\langle T_-\exp\left(\frac{i}{\hbar}\int_{t_0}^{t}dt'\,\phi_1(t')\right)T_+\exp\left(-\frac{i}{\hbar}\int_{t_0}^{t}dt'\,\phi_2(t')\right)\right\rangle|S_{i_2}\rangle|S_{f_1}\rangle, \tag{2.12}$$

where

$$\langle\cdots\rangle=\sum_{R_i}P_{R_i}\langle R_i|\cdots|R_i\rangle, \tag{2.13}$$

$$\phi_1(t)=\phi[A_1(t),B(t)], \tag{2.14}$$

and

$$A_1(t)=\exp(iH_{S_1}t/\hbar)A_1\exp(-iH_{S_1}t/\hbar). \tag{2.15}$$

We now assume a specific form for $\phi(t)$, namely,

$$\phi(t)=A(t)B(t). \tag{2.16}$$

This form for the interaction is found in many processes, including magnetic scattering of neutrons [see Eq. (3.11)]. Inserting it into Eq. (2.12) and setting $T_+=T_+^{A_2}T_+^{B}$, $T_-=T_-^{A_1}T_-^{B}$, the transition probability is

$$\langle S_{f_2}|\rho_S(t)|S_{f_1}\rangle=P_{12}\langle S_{f_2}|\langle S_{i_1}|T_-^{A_1}T_+^{A_2}\left\langle T_-^{B}\exp\left(\frac{i}{\hbar}\int_{t_0}^{t}dt'A_1(t')B(t')\right)T_+^{B}\exp\left(-\frac{i}{\hbar}\int_{t_0}^{t}dt'A_2(t')B(t')\right)\right\rangle|S_{i_2}\rangle|S_{f_1}\rangle. \tag{2.17}$$

In order to obtain a perturbative expansion,[4] we expand Eq. (2.17) in a Taylor's series, i.e.,

$$\Gamma(\lambda)\equiv\left\langle T_-^{B}\exp\left(\lambda\frac{i}{\hbar}\int_{t_0}^{t}dt'A_1(t')B(t')\right)T_+^{B}\exp\left(-\lambda\frac{i}{\hbar}\int_{t_0}^{t}dt'A_2(t')B(t')\right)\right\rangle=\sum_{n=0}^{\infty}\frac{\Gamma_n}{n!}\lambda^n \tag{2.18}$$

where

$$\Gamma_n=\left.\frac{d^n\Gamma(\lambda)}{d\lambda^n}\right|_{\lambda=0}, \tag{2.19}$$

and where finally, λ is set equal to unity. The first three terms of the expansion are

$$\Gamma_0=\Gamma(0)=1, \tag{2.20}$$

$$\Gamma_1=\frac{i}{\hbar}\int_{t_0}^{t}dt_1[A_1(t_1)-A_2(t_1)]\langle B(t_1)\rangle, \tag{2.21}$$

$$\frac{\Gamma_2}{2!}=-\frac{1}{\hbar^2}\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2A_1(t_1)A_1(t_2)\langle B(t_2)B(t_1)\rangle+\frac{1}{\hbar^2}\int_{t_0}^{t}dt_1\int_{t_0}^{t}dt_2A_1(t_1)A_2(t_2)\langle B(t_1)B(t_2)\rangle$$
$$-\frac{1}{\hbar^2}\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2A_2(t_1)A_2(t_2)\langle B(t_1)B(t_2)\rangle. \tag{2.22}$$

It is easily shown from Eq. (2.17) that all terms in Γ_n that contain only A_1 operators or only A_2 operators do not cause a transition in S, i.e., $|S_i\rangle=|S_f\rangle$ for such terms. In terms of scattering, these would represent forward scattering terms. The lowest-order term capable of causing a transition is the middle term in Eq. (2.22) and retaining only this term in the expansion, we will obtain a first-order perturbation theory. From Eq. (2.17), we get for the lowest-order transition probability

$$\langle S_{f_2}|\rho_S(t)|S_{f_1}\rangle^{(1)} = P_{12}\frac{1}{\hbar^2}\langle S_{f_2}|\langle S_{i_1}|\int_{t_0}^{t} dt_1 \int_{t_0}^{t} dt_2 A_1(t_1)A_2(t_2)\langle B(t_1)B(t_2)\rangle|S_{i_2}\rangle|S_{f_1}\rangle. \tag{2.23}$$

Equation (2.23) can be transcribed into Van Hove's form for the first-Born-scattering approximation containing the target dynamic structure factor. For an interaction depending on distance only, we have

$$\phi(t) = \sum_j \phi[\vec{r}(t)] = \sum_j \sum_{\vec{k}_a} g(\vec{k}_a) e^{i\vec{k}_a\cdot[\vec{r}_N(t)-\vec{r}_j(t)]}, \tag{2.24}$$

where $\vec{r}(t) = \vec{r}_N(t) - \vec{r}_j(t)$, $\vec{r}_N(t)$ being the neutron position vector, $\vec{r}_j(t)$ being the position vector of the jth target atom and $g(\vec{k}_a)$ is the Fourier transform of $\phi(\vec{r})$, i.e.,

$$g(\vec{k}_a) = \int d^3r\, e^{-i\vec{k}_a\cdot\vec{r}}\phi(\vec{r}), \tag{2.25}$$

where we have set the quantization volume equal to unity, a convention we will follow henceforth. Thus, the operators $A(t)$ and $B(t)$ are represented by

$$A(t) \to e^{i\vec{k}_a\cdot\vec{r}_N(t)}, \quad B(t) \to \sum_j e^{-i\vec{k}_a\cdot\vec{r}_j(t)} = n^{\dagger}_{\vec{k}_a}(t), \tag{2.26}$$

where $n^{\dagger}_{\vec{k}_a}(t)$ creates a density fluctuation in the target of wavelength $\lambda = 2\pi/k_a$. With initial and final plane-wave states for the neutron, $|S_{i,f}\rangle \to |\vec{k}_{i,f}\rangle$, and with $t_0 = 0$, Eq. (2.23) becomes

$$\langle\vec{k}_f|\rho_S(t)|\vec{k}_f\rangle^{(1)} = \frac{1}{\hbar^2}\sum_{\vec{k}_a}\sum_{\vec{k}_b}\int_0^t dt_1\int_0^t dt_2\, g(\vec{k}_a)g(\vec{k}_b)\langle\vec{k}_i|e^{i\vec{k}_a\cdot\vec{r}_N(t_1)}|\vec{k}_f\rangle\langle\vec{k}_f|e^{i\vec{k}_b\cdot\vec{r}_N(t_2)}|\vec{k}_i\rangle\langle n^{\dagger}_{\vec{k}_a}(t_1)n^{\dagger}_{\vec{k}_b}(t_2)\rangle. \tag{2.27}$$

The neutron matrix elements are

$$\begin{aligned}\langle\vec{k}_i|e^{i\vec{k}_a\cdot\vec{r}_N(t_1)}|\vec{k}_f\rangle &= e^{i\omega t_1}\delta_{\vec{k},\vec{k}_a},\\ \langle\vec{k}_f|e^{i\vec{k}_b\cdot\vec{r}_N(t_2)}|\vec{k}_i\rangle &= e^{-i\omega t_2}\delta_{-\vec{k},\vec{k}_b},\end{aligned} \tag{2.28}$$

where $\hbar\omega = E_i - E_f$ is the neutron energy loss and $\hbar\vec{k} = \hbar(\vec{k}_i - \vec{k}_f)$ is the neutron momentum transfer. Using Eqs. (2.28) in Eq. (2.27), the probability for a transition from $|\vec{k}_i\rangle$ to $|\vec{k}_f\rangle$ is

$$\begin{aligned}&\langle\vec{k}_f|\rho_S(t)|\vec{k}_f\rangle^{(1)}\\ &= \frac{1}{\hbar^2}|g(\vec{k})|^2\int_0^t dt_1\int_0^t dt_2\, e^{i\omega(t_1-t_2)}\\ &\quad\times\langle n^{\dagger}_{\vec{k}}(t_1)n_{-\vec{k}}(t_2)\rangle.\end{aligned} \tag{2.29}$$

The target density-density correlation function can be expressed in terms of the dynamic structure factor $S(\vec{k},\omega')$ in the form

$$\langle n^{\dagger}_{\vec{k}}(t_1)n_{-\vec{k}}(t_2)\rangle = \int d\omega' e^{i\omega'(t_2-t_1)}S(\vec{k},\omega') \tag{2.30}$$

or

$$S(\vec{k},\omega') = \frac{1}{2\pi}\int d\tau\, e^{-i\omega'\tau}\langle n^{\dagger}_{\vec{k}}(0)n_{-\vec{k}}(\tau)\rangle \tag{2.31}$$

where $\tau = t_2 - t_1$. Inserting Eq. (2.30) into Eq. (2.29) gives, in the limit $t\to\infty$,

$$\frac{d}{dt}\langle\vec{k}_f|\rho_S(t)|\vec{k}_f\rangle^{(1)} = \frac{2\pi}{\hbar^2}|g(\vec{k})|^2 S(\vec{k},\omega). \tag{2.32}$$

Converting this probability per second into a cross section by multiplying by the density of final neutron states, $\rho(E_f)dE_f$, and dividing by the incoming neutron current j_{in},

$$\rho(E_f)\,dE_f j_{in}^{-1} = \frac{m_N^2}{(2\pi\hbar)^3}\frac{k_f}{k_i}\,d\Omega\,dE_f \tag{2.33}$$

where m_N is the neutron mass, yields the well-known form for the scattering cross section first given by Van Hove[5]:

$$\frac{d^2\sigma^{(1)}}{d\Omega\,dE_f} = \frac{m_N^2}{4\pi^2\hbar^5}\frac{k_f}{k_i}|g(\vec{k})|^2 S(\vec{k},\omega). \tag{2.34}$$

It is clear from the perturbative expansion, Eq. (2.18), that higher-order scattering terms beyond Γ_2 will involve target averages of products of increasing numbers of target operators $B(t)$ beyond the bilinear product involved in Γ_2. The present development thus seems to be a natural extension of Van Hove's first Born formulation to higher orders. Although the problem of precisely evaluating higher-order time-dependent correlations of target operators is a formidable one, these higher-order target correlations can be well approximated by products of lower-order correlations and such a decomposition is utilized in Sec. III.

III. THERMAL-NEUTRON DOUBLE SCATTERING

In Sec. II, we showed that the probability of finding S in eigenstate $|S_f\rangle$ at time t after having interacted with R from time t_0 is

$$\langle S_{f_2}|\rho_S(t)|S_{f_1}\rangle = P_{12}\langle S_{f_2}|\langle S_{i_1}|T^{A_1}_{-}T^{A_2}_{+}\sum_{n=0}^{\infty}\frac{\Gamma_n}{n!}|S_{i_2}\rangle|S_{f_1}\rangle \tag{3.1}$$

with Γ_n determined by Eqs. (2.18) and (2.19). The Γ_2 term was shown to reproduce the first-Born-approximation scattering. This section treats the Γ_4 term which contains the double scattering and is shown to result in a simple formula for the cross section, Eq. (3.24), which is bilinear in dynamic structure factors.

To obtain a multiple (double) scattering theory, we consider higher-order terms in Eq. (3.1). If multiple scattering is to be a small effect, we can, to a good approximation, neglect the interference term Γ_3. In fact, when we specialize to small-angle thermal-neutron magnetic scattering in Sec. IV we will show [see discussion following Eq. (4.15)] that all Γ_n, where n is odd, are zero if R is a ferromagnetic crystal just above its Curie temperature. Therefore, we now consider the Γ_4 term. Explicitly,

$$\begin{aligned}\frac{\Gamma_4}{4!} &= \frac{1}{\hbar^4}\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2\int_{t_0}^{t_2}dt_3\int_{t_0}^{t_3}dt_4 A_1(t_1)A_1(t_2)A_1(t_3)A_1(t_4)\langle B(t_4)B(t_3)B(t_2)B(t_1)\rangle \\ &- \frac{1}{\hbar^4}\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2\int_{t_0}^{t_2}dt_3\int_{t_0}^{t}dt_4 A_1(t_1)A_1(t_2)A_1(t_3)A_2(t_4)\langle B(t_3)B(t_2)B(t_1)B(t_4)\rangle \\ &+ \frac{1}{\hbar^4}\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2\int_{t_0}^{t}dt_3\int_{t_0}^{t_3}dt_4 A_1(t_1)A_1(t_2)A_2(t_3)A_2(t_4)\langle B(t_2)B(t_1)B(t_3)B(t_4)\rangle \\ &- \frac{1}{\hbar^4}\int_{t_0}^{t}dt_1\int_{t_0}^{t}dt_2\int_{t_0}^{t_2}dt_3\int_{t_0}^{t_3}dt_4 A_1(t_1)A_2(t_2)A_2(t_3)A_2(t_4)\langle B(t_1)B(t_2)B(t_3)B(t_4)\rangle \\ &+ \frac{1}{\hbar^4}\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2\int_{t_0}^{t_2}dt_3\int_{t_0}^{t_3}dt_4 A_2(t_1)A_2(t_2)A_2(t_3)A_2(t_4)\langle B(t_1)B(t_2)B(t_3)B(t_4)\rangle .\end{aligned} \tag{3.2}$$

The first and last terms in Eq. (3.2) contain only A_1 operators or only A_2 operators, and hence contribute only to forward scattering and are not treated here. Detailed investigation of the second and fourth terms in Eq. (3.2) indicates that they make small adjustments in the single scattering corresponding to the processes of refraction and extinction, which can be neglected for neutron scattering.[6] The third, or middle term in Eq. (3.2) represents the true double scattering, and will now be considered in detail. It gives a second-order correction, $\mathcal{P}^{(2)}$, to Eq. (2.23),

$$\begin{aligned}\mathcal{P}^{(2)} &\equiv \langle S_{f_2}|\rho_S(t)|S_{f_1}\rangle^{(2)} \\ &= P_{12}\frac{1}{\hbar^4}\langle S_{f_2}|\langle S_{i_1}|\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2\int_{t_0}^{t}dt_3\int_{t_0}^{t_3}dt_4 A_1(t_2)A_1(t_1)A_2(t_3)A_2(t_4) \\ &\qquad\times\langle B(t_2)B(t_1)B(t_3)B(t_4)\rangle|S_{i_2}\rangle|S_{f_1}\rangle\end{aligned} \tag{3.3}$$

containing the average over initial target states of the product of four target operators B. Again, if multiple scattering is to be a small effect, then to a good approximation, the four B operator correlation function in Eq. (3.3) can be decomposed into products of pair correlation functions:

$$\langle B(t_2)B(t_1)B(t_3)B(t_4)\rangle = \langle B(t_2)B(t_1)\rangle\langle B(t_3)B(t_4)\rangle + \langle B(t_2)B(t_3)\rangle\langle B(t_1)B(t_4)\rangle + \langle B(t_2)B(t_4)\rangle\langle B(t_1)B(t_3)\rangle . \tag{3.4}$$

For small-angle thermal-neutron scattering from a ferromagnetic crystal just above its Curie temperature, this decomposition is virtually exact [see discussion following Eq. (4.15)]. In fact the two approximations made thus far, i.e., setting all Γ_n, n odd, equal to zero and the decomposition in Eq. (3.4), become exact for targets composed of noninteracting bosons in thermal equilibrium if $\langle B(t)\rangle$ vanishes.[7] Thus, this formalism is fully applicable to neutrons magnetically scattered from low-temperature spin waves[8] as well as critical magnetic scattering, and should provide a good approximation for other types of multiple scattering, such as nuclear multiple scattering.

Of the three terms on the right-hand side of Eq. (3.4), only the last term is found to contribute significantly to double scattering and will be the only term retained. The first two terms are treated in Sec. IV for the case of critical magnetic scattering (see discussion preceeding Sec. IV A).

So far in the formalism, the interaction is suddenly turned on at t_0. To describe a scattering process, the initial probe state $|S_i\rangle$ should be localized in space, i.e., represented by a wave packet, to reflect laboratory conditions. To circumvent using a wave packet for $|S_i\rangle$ and use, instead, plane-wave initial states which are mathematically more tractable, we adopt the following well-known ansatz.[9] Representing the

initial probe state by a plane wave, we let $t_0 \to -\infty$ and adiabatically turn on the interaction, i.e., $\phi(t)$ is modified thusly,

$$\phi(t) \to \phi(t)e^{\epsilon t} \quad (\epsilon > 0) \tag{3.5}$$

so that $\lim_{t\to-\infty}\phi(t)=0$. Finally, we let $\epsilon \to 0$. Therefore, from Eq. (3.4), Eq. (3.3) becomes,

$$\mathcal{P}^{(2)} = \lim_{\epsilon\to 0}\frac{1}{\hbar^4}\int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t} dt_3 \int_{-\infty}^{t_3} dt_4 e^{\epsilon(t_1+t_2+t_3+t_4)} \times \langle S_i|A(t_2)A(t_1)|S_f\rangle\langle S_f|A(t_3)A(t_4)|S_i\rangle\langle B(t_2)B(t_4)\rangle\langle B(t_1)B(t_3)\rangle . \tag{3.6}$$

We now specify a form for the operators A and B in the interaction $\phi = AB$ that will encompass the case of neutron magnetic scattering. The specific details of the magnetic interaction are considered in Sec. IV. The interaction ϕ is written as

$$\phi = \sum_j \phi_j[\vec{r}(t), \vec{\mu}_N, \vec{\mu}_j(t)], \tag{3.7}$$

where $\vec{r}(t) = \vec{r}_N(t) - \vec{r}_j(t)$ is the relative neutron–target-electron separation, $\vec{r}_N$ is the neutron position vector, $\vec{r}_j$ is the position vector of the jth target electron, $\vec{\mu}_N$ is the neutron magnetic moment, and $\vec{\mu}_j$ is the magnetic moment of the jth target electron. The Fourier transform, $\hat{\phi}_j$, of ϕ_j is defined as

$$\hat{\phi}_j(\vec{k},t) = \int d^3r e^{-i\vec{k}\cdot\vec{r}}\phi_j[\vec{r}(t), \vec{\mu}_N, \vec{\mu}_j(t)] . \tag{3.8}$$

Thus,

$$\phi = \sum_j \sum_{\vec{k}} e^{i\vec{k}\cdot[\vec{r}_N(t)-\vec{r}_j(t)]}\hat{\phi}_j(\vec{k},t) . \tag{3.9}$$

Because of the simple form of the magnetic dipole interaction $\phi_j(\vec{r}, \vec{\mu}_N, \vec{\mu}_j)$ [see Eq. (4.2)], $\hat{\phi}_j(\vec{k},t)$ can be written

$$\hat{\phi}_j(\vec{k},t) = \vec{a}(\vec{k})\cdot\vec{b}_j(\vec{k},t) = \sum_\alpha a^\alpha(\vec{k})b_j^\alpha(\vec{k},t) , \tag{3.10}$$

where $\vec{a}$ contains the neutron spin operator, $\vec{b}_j$ contains the spin operator of the $j$the target electron [see Eqs. (4.7) and (4.8)], and $\alpha = x,y,z$ refer to rectangular coordinates. For the case of neutron nuclear scattering, the sum over j in Eq. (3.7) refers to target nuclei and $\phi_j = \text{const} = 2\pi b\hbar^2/m_N$, b representing the S-wave scattering length.

Inserting Eq. (3.10) for ϕ_j in Eq. (3.9) for ϕ,

$$\phi(t) = \sum_\alpha \sum_{\vec{k}} A^\alpha(\vec{k},t)B^\alpha(\vec{k},t) , \tag{3.11}$$

where

$$A^\alpha(\vec{k},t) = e^{i\vec{k}\cdot\vec{r}_N(t)}a^\alpha(\vec{k}) , \tag{3.12}$$

$$B^\alpha(\vec{k},t) = \sum_j e^{-i\vec{k}\cdot\vec{r}_j(t)}b_j^\alpha(\vec{k},t) \tag{3.13}$$

Making these identifications in Eq. (3.6) and writing $|S_{i,f}\rangle = |\vec{k}_{i,f}\rangle|\sigma_{i,f}\rangle$, i.e., a direct product of neutron spatial and spin states, there results,

$$\mathcal{P}^{(2)} = \lim_{\epsilon\to 0}\frac{1}{\hbar^4}\int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t} dt_3 \int_{-\infty}^{t_3} dt_4 e^{\epsilon(t_1+t_2+t_3+t_4)} \sum_{\alpha\beta\gamma\delta}\sum_{\vec{k}_1\vec{k}_2\vec{k}_3\vec{k}_4} M_N^r M_N^\sigma M_T , \tag{3.14}$$

where M_N^r stands for the following product of neutron spatial matrix elements,

$$M_N^r = \langle\vec{k}_i|e^{i\vec{k}_2\cdot\vec{r}_N(t_2)}e^{i\vec{k}_1\cdot\vec{r}_N(t_1)}|\vec{k}_f\rangle\langle\vec{k}_f|e^{i\vec{k}_3\cdot\vec{r}_N(t_3)}e^{i\vec{k}_4\cdot\vec{r}_N(t_4)}|\vec{k}_i\rangle; \tag{3.15}$$

M_N^σ stands for the product of neutron spin matrix elements,

$$M_N^\sigma = \langle\sigma_i|a^\alpha(\vec{k}_2)a^\beta(\vec{k}_1)|\sigma_f\rangle\langle\sigma_f|a^\gamma(\vec{k}_3)a^\delta(\vec{k}_4)|\sigma_i\rangle; \tag{3.16}$$

and M_T stands for the target space and spin average,

$$M_T = \langle B^\alpha(\vec{k}_2, t_2) B^\delta(\vec{k}_4, t_4)\rangle \langle B^\beta(\vec{k}_1, t_1) B^\gamma(\vec{k}_3, t_3)\rangle . \tag{3.17}$$

The neutron spatial matrix elements in M_N^r of Eq. (3.15) are expressed in terms of intermediate neutron states with energies E_a, E_b and wave vectors $\vec{k}_a, \vec{k}_b$ in the following form:

$$M_N^r = \sum_{\vec{k}_a, \vec{k}_b} e^{i(E_i - E_a)t_2/\hbar} e^{i(E_a - E_f)t_1/\hbar} e^{i(E_f - E_b)t_3/\hbar} e^{i(E_b - E_i)t_4/\hbar} \delta_{\vec{k}_i, \vec{k}_2 + \vec{k}_a} \delta_{\vec{k}_a, \vec{k}_1 + \vec{k}_f} \delta_{\vec{k}_f, \vec{k}_3 + \vec{k}_b} \delta_{\vec{k}_b, \vec{k}_4 + \vec{k}_i} . \tag{3.18}$$

Following Van Hove,[10] we define a dynamic structure factor[11] $S_{\alpha\delta}(\vec{k}_2, \vec{k}_4, \omega')$

$$\langle B^\alpha(\vec{k}_2, t_2) B^\delta(\vec{k}_4, t_4)\rangle \equiv \int d\omega' S_{\alpha\delta}(\vec{k}_2, \vec{k}_4, \omega') e^{i\omega'(t_4 - t_2)} \tag{3.19}$$

or

$$S_{\alpha\delta}(\vec{k}_2, \vec{k}_4, \omega') = \frac{1}{2\pi} \int d\tau \langle B^\alpha(\vec{k}_2, 0) B^\delta(\vec{k}_4, \tau)\rangle e^{-i\omega'\tau} . \tag{3.20}$$

Therefore, from Eq. (3.17)

$$M_T = \int d\omega' \int d\tilde{\omega} S_{\alpha\delta}(\vec{k}_2, \vec{k}_4, \omega') S_{\beta\gamma}(\vec{k}_1, \vec{k}_3, \tilde{\omega}) e^{i\omega'(t_4 - t_2)} e^{i\tilde{\omega}(t_3 - t_1)} . \tag{3.21}$$

Using Eqs. (3.18) and (3.21) we can now perform the time integrations in Eq. (3.14). In anticipation of the limit $\epsilon \to 0$, we obtain

$$\begin{aligned} \frac{d}{dt} \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t} dt_3 \int_{-\infty}^{t_3} dt_4 e^{\epsilon(t_1 + t_2 + t_3 + t_4)} e^{i(E_i - E_a - \hbar\omega')t_2/\hbar} \\ \times e^{i(E_a - E_f - \hbar\tilde{\omega})t_1/\hbar} e^{i(E_f - E_b + \hbar\tilde{\omega})t_3/\hbar} e^{i(E_b - E_i + \hbar\omega')t_4/\hbar} \\ = 2\pi\hbar^3 [(E_a - E_i + \hbar\omega' + i\hbar\epsilon)(E_b - E_i + \hbar\omega' - i\hbar\epsilon)]^{-1} \delta(E_i - E_f - \hbar\omega' - \hbar\tilde{\omega}) . \end{aligned} \tag{3.22}$$

From Eqs. (3.18), (3.21), and (3.22), Eq. (3.14) is expressed in the form

$$\begin{aligned} \frac{d\mathcal{P}^{(2)}}{dt} = C' \lim_{\epsilon \to 0} \sum_{\alpha\beta\gamma\delta} \sum_{\vec{k}_a, \vec{k}_b} M_N^\alpha \int d\omega' S_{\alpha\delta}(\vec{k}_i - \vec{k}_a, \vec{k}_b - \vec{k}_i, \omega') S_{\beta\gamma}(\vec{k}_a - \vec{k}_f, \vec{k}_f - \vec{k}_b, \omega - \omega') \\ \times (k_a^2 - k_i^2 + 2m_N\omega'/\hbar + i\epsilon)^{-1} (k_b^2 - k_i^2 + 2m_N\omega'/\hbar - i\epsilon)^{-1} , \end{aligned} \tag{3.23}$$

where

$$C' = 8\pi m_N^2/\hbar^6 , \quad \omega = (1/\hbar)(E_i - E_f) .$$

We now let the sums over $\vec{k}_a, \vec{k}_b$ go over to integrals and multiply Eq. (3.23) by the density of final neutron states and divide it by the incoming current, as given in Eq. (2.33), thereby obtaining the general double scattering cross section

$$\begin{aligned} \frac{d^2\sigma^{(2)}}{d\Omega\, d\omega} = C \lim_{\epsilon \to 0} \frac{k_f}{k_i} \sum_{\alpha\beta\gamma\delta} \int d^3k_a \int d^3k_b \int d\omega' M_N^\alpha S_{\alpha\delta}(\vec{k}_i - \vec{k}_a, \vec{k}_b - \vec{k}_i, \omega') \\ \times S_{\beta\gamma}(\vec{k}_a - \vec{k}_f, \vec{k}_f - \vec{k}_b, \omega - \omega')(k_a^2 - k_i^2 + 2m_N\omega'/\hbar + i\epsilon)^{-1} \\ \times (k_b^2 - k_i^2 + 2m_N\omega'/\hbar - i\epsilon)^{-1} , \end{aligned} \tag{3.24}$$

$$C = \left[\frac{\hbar m_N^2}{(2\pi\hbar)^3 (2\pi)^6}\right] C' = \frac{4m_N^4}{(2\pi\hbar)^8} ,$$

which is bilinear in dynamic structure factors.

IV. CRITICAL MAGNETIC DOUBLE SCATTERING FROM FERROMAGNETIC CRYSTALS

The results of Sec. III are now applied to the case of critical magnetic scattering.

We let our target be a ferromagnetic crystal just above its Curie temperature and specialize to the case of magnetic scattering, where the spin of the incoming neutron couples to the spin of the unpaired target electrons. We only consider tar-

gets where the orbital contribution to the magnetic moment is negligible or can be taken account of by simply adjusting the electronic spin quantum number. The interaction ϕ_j between the magnetic moment of the neutron $\vec{\mu}_N$ at $\vec{r}_N$, and the magnetic moment of the jth electron $\vec{\mu}_j$ at $\vec{r}_j$ is[12]

$$\phi_j = -\vec{\mu}_j \cdot \vec{H}_N \tag{4.1}$$

where

$$\vec{H}_N = \vec{\nabla} \times \frac{\vec{\mu}_N \times \vec{r}}{r^3} = -\vec{\nabla} \times \vec{\mu}_N \times \vec{\nabla}(1/r)$$

and

$$\vec{r} = \vec{r}_N - \vec{r}_j .$$

By standard vector algebra,

$$\phi_j = -\vec{\mu}_j \cdot \vec{\nabla}[\vec{\mu}_N \cdot \vec{\nabla}(1/r)] - 4\pi \vec{\mu}_j \cdot \vec{\mu}_N \delta(\vec{r}). \tag{4.2}$$

Therefore, from Eq. (3.8) we have

$$\hat{\phi}_j(\vec{k}, t) = -4\pi \vec{\mu}_N \cdot \{\vec{\mu}_j(t) - \hat{k}[\vec{\mu}_j(t) \cdot \hat{k}]\} \tag{4.3}$$

where $\hat{k} = \vec{k}/k$. The operators $\vec{a}, \vec{b}_j$ appearing in Eq. (3.10) become

$$\vec{a}(\vec{k}) = -4\pi \vec{\mu}_N, \tag{4.4}$$

$$\vec{b}_j(\vec{k}, t) = \vec{\mu}_j(t) - \hat{k}[\vec{\mu}_j(t) \cdot \hat{k}]. \tag{4.5}$$

The magnetic moments are related to neutron $\vec{s}_N$ and electron $\vec{s}_j$ spins by

$$\vec{\mu}_N = -g(e\hbar/m_N c)\vec{s}_N, \qquad \vec{\mu}_j = -(e\hbar/m_e c)\vec{s}_j, \tag{4.6}$$

where e and m_e are the electron charge and mass and $g = 1.91$. Replacement of these moments in Eqs. (4.4) and (4.5) gives

$$\vec{a}(\vec{k}) = 4\pi g(e\hbar/m_N c)\vec{s}_N, \tag{4.7}$$

$$\vec{b}_j(\vec{k}, t) = -(e\hbar/m_e c)\{\vec{s}_j(t) - \hat{k}[\vec{s}_j(t) \cdot \hat{k}]\}. \tag{4.8}$$

From Eqs. (3.13) and (4.8), the dynamic structure factor defined by Eq. (3.20) can be written as

$$S_{\alpha\beta}(\vec{k}_2, \vec{k}_4, \omega') = \frac{1}{2\pi} \int d\tau \left\langle \sum_j e^{-i\vec{k}_2 \cdot \vec{r}_j(0)} b_j^\alpha(\vec{k}_2, 0) \sum_n e^{-i\vec{k}_4 \cdot \vec{r}_n(\tau)} b_n^\beta(\vec{k}_4, \tau) \right\rangle e^{-i\omega'\tau}, \tag{4.9}$$

$$S_{\beta\gamma}(\vec{k}_1, \vec{k}_3, \tilde{\omega}) = \frac{1}{2\pi} \int d\tau \left\langle \sum_l e^{-i\vec{k}_1 \cdot \vec{r}_l(0)} b_l^\beta(\vec{k}_1, 0) \sum_m e^{-i\vec{k}_3 \cdot \vec{r}_m(\tau)} b_m^\gamma(\vec{k}_3, \tau) \right\rangle e^{-i\tilde{\omega}\tau}, \tag{4.10}$$

where b_j^α is the α component $\vec{b}_j$, given by Eq. (4.8).

We now assume the target to be a crystal made up of identical atoms whose nuclear thermal motions can be neglected. Writing the electron position vector $\vec{r}_j(\tau)$ as $\vec{r}_{jj'}(\tau) = \vec{R}_j(\tau) + \vec{u}_{j'}(\tau)$, describing the position of the j'th electron of the jth atom, these assumptions imply that the jth atom's position vector $\vec{R}_j(\tau) \cong \vec{R}_j(0)$, a fixed "$c$ number." Inserting a complete set of electronic spatial states into the target averages in Eqs. (4.9) and (4.10), we neglect all matrix elements that involve transitions to excited electronic states, since thermal neutrons have insufficient energies to cause such transitions. Now we assume that the electronic spin states are adequately descibed by the Heisenberg model, which ascribes to each atom an effective spin operator $\vec{S}$ of fixed length, and that the crystal lattice is a Bravais lattice with one atom per cell. Equations (4.9) and (4.10) then become

$$S_{\alpha\beta}(\vec{k}_2, \vec{k}_4, \omega') = \frac{1}{2\pi} F(k_2) F(k_4) \sum_{j,n} e^{-i(\vec{k}_2 \cdot \vec{R}_j + \vec{k}_4 \cdot \vec{R}_n)} \times \int d\tau \langle B_j^\alpha(\vec{k}_2, 0) B_n^\beta(\vec{k}_4, \tau) \rangle e^{-i\omega'\tau}, \tag{4.11}$$

$$S_{\beta\gamma}(\vec{k}_1, \vec{k}_3, \tilde{\omega}) = \frac{1}{2\pi} F(k_1) F(k_3) \sum_{l,m} e^{-i(\vec{k}_1 \cdot \vec{R}_l + \vec{k}_3 \cdot \vec{R}_m)} \times \int d\tau \langle B_l^\beta(\vec{k}_1, 0) B_m^\gamma(\vec{k}_3, \tau) \rangle e^{-i\tilde{\omega}\tau}, \tag{4.12}$$

where

$$B_j^\alpha(\vec{k}, \tau) = -(e\hbar/m_e c)\{\vec{S}_j(\tau) - \hat{k}[\vec{S}_j(\tau) \cdot \hat{k}]\}^\alpha. \tag{4.13}$$

Here $\vec{S}_j$ stands for the effective spin operator of the jth atom, and $F(k)$ is the magnetic atomic form factor, i.e.,

$$F(k) = \int d^3u \, e^{i\vec{k} \cdot \vec{u}} s(\vec{u}), \tag{4.14}$$

$s(u)$ being the normalized spin density. The average is over the target eigenstates of the Heisenberg Hamiltonian,

$$H = \sum_{jl} J_{jl} \vec{S}_j \cdot \vec{S}_l, \tag{4.15}$$

which determines the time evolvement of the Heisenberg operators $\vec{S}_j(\tau)$.

At this point, we now digress to show that the approximation of decomposing the four target operator correlation function into the sum of bilinear

products is virtually exact for the critical magnetic scattering treated here. If this approximation had not been made, we would have arrived at this point in the formalism with a correlation function containing four B operators, instead of a bilinear product of pair correlation functions. However, if we restrict ourselves to small-angle scattering, then for the very small neutron wave-vector transfers involved, the neutron is probing fluctuations in the spin operator $B_j^\alpha(\vec{k},\tau)$ over very large distances on the microscopic scale. Therefore, a classical description of the system in terms of these long-wavelength fluctuations is adequate for determining the correlation function. In terms of the fluctuations of B_j^α, the free energy of the system can be expressed as a quadratic form,[13] and since $\langle \vec{S}_j \rangle = 0$ above the Curie temperature, i.e., $\langle B_j^\alpha \rangle = 0$, it immediately follows that the correlation of any odd number of B_j^α's is zero. Similarly, the decomposition of the four $B_j^\alpha(\vec{k},\tau)$ operator correlation function becomes exact.

Since long-wavelength spin fluctuations are known to relax very slowly over times for the neutron to traverse an effective scattering volume $B_j^\alpha(\vec{k},\tau) \cong B_j^\alpha(\vec{k},0)$, i.e., the scattering is predominately quasielastic. Therefore from Eqs. (4.11) and (4.12) we have

$$S_{\alpha\delta}(\vec{k}_2,\vec{k}_4,\omega') = F(k_2)F(k_4)\sum_{j,n} e^{-i(\vec{k}_2\cdot\vec{R}_j+\vec{k}_4\cdot\vec{R}_n)} \times\langle B_j^\alpha(\vec{k}_2)B_n^\delta(\vec{k}_4)\rangle\delta(\omega'), \tag{4.16}$$

$$S_{\beta\gamma}(\vec{k}_1,\vec{k}_3,\tilde{\omega}) = F(k_1)F(k_3)\sum_{l,m} e^{-i(\vec{k}_1\cdot\vec{R}_l+\vec{k}_3\cdot\vec{R}_m)} \times\langle B^\beta{}_l(\vec{k}_1)B_m^\gamma(\vec{k}_3)\rangle\delta(\tilde{\omega}). \tag{4.17}$$

The neutron spin matrix element term M_N^σ given by Eq. (3.16) is evaluated for an unpolarized neutron beam. Summing over final neutron spin states and averaging over initial neutron spin states gives from Eq. (4.7),

$$M_N^\sigma = \frac{1}{2}\left(\frac{4\pi g e\hbar}{m_N c}\right)^4 \sum_{\sigma_i}\langle\sigma_i|s_N^\alpha s_N^\beta s_N^\gamma s_N^\delta|\sigma_i\rangle. \tag{4.18}$$

As previously remarked, we adopt the Heisenberg model for the spin system of the crystal, with Hamiltonian given by Eq. (4.15). For this Hamiltonian the spin system is invariant to simultaneous rotations of spins, i.e.,

$$\langle S_j^\alpha S_l^\beta\rangle = \tfrac{1}{3}\delta_{\alpha\beta}\langle\vec{S}_j\cdot\vec{S}_l\rangle. \tag{4.19}$$

The sum over spin components α, β, γ, and δ in Eq. (3.24) can now be performed. Using Eqs. (4.13)– (4.19), one obtains

$$\sum_{\alpha\beta\gamma\delta} M_N^\sigma\langle B_j^\alpha(\vec{k}_2)B_n^\delta(\vec{k}_4)\rangle\langle B_l^\beta(\vec{k}_1)B_m^\gamma(\vec{k}_3)\rangle = \left(\frac{4\pi g e^2\hbar^2}{m_N m_e c^2}\right)^4 f(\vec{k}_1,\vec{k}_2,\vec{k}_3,\vec{k}_4)\langle\vec{S}_j\cdot\vec{S}_n\rangle\langle\vec{S}_l\cdot\vec{S}_m\rangle, \tag{4.20}$$

where

$$f = \tfrac{1}{144}\{[1+(\hat{k}_2\cdot\hat{k}_4)^2][1+(\hat{k}_1\cdot\hat{k}_3)^2] - (\hat{k}_2\cdot\hat{k}_4)(\hat{k}_1\cdot\hat{k}_3)(\hat{k}_2\times\hat{k}_4)\cdot(\hat{k}_1\times\hat{k}_3)\}. \tag{4.21}$$

From Eqs. (4.16), (4.17), and (4.20), the cross section for double scattering, Eq. (3.24), becomes

$$\frac{d^2\sigma^{(2)}}{d\Omega\,d\omega} = C_1\lim_{\epsilon\to 0}\frac{k_f}{k_i}\int d^3k_a\int d^3k_b f(\vec{k}_a-\vec{k}_f,\vec{k}_i-\vec{k}_a,\vec{k}_f-\vec{k}_b,\vec{k}_b-\vec{k}_i)\,F(|\vec{k}_i-\vec{k}_a|)F(|\vec{k}_a-\vec{k}_f|)F(|\vec{k}_f-\vec{k}_b|)F(|\vec{k}_b-\vec{k}_i|) \times\sum_{jlmn} e^{-i(\vec{k}_i-\vec{k}_a)\cdot\vec{R}_j}e^{-i(\vec{k}_a-\vec{k}_f)\cdot\vec{R}_l}e^{-i(\vec{k}_f-\vec{k}_b)\cdot\vec{R}_m}e^{-i(\vec{k}_b-\vec{k}_i)\cdot\vec{R}_n} \times\langle\vec{S}_j\cdot\vec{S}_n\rangle\langle\vec{S}_l\cdot\vec{S}_m\rangle(k_a^2-k_i^2+i\epsilon)^{-1}(k_b^2-k_i^2-i\epsilon)^{-1}\delta(\omega), \tag{4.22}$$

where

$$C_1 = 4(ge^2/\pi m_e c^2)^4.$$

For the spin system, we assume a translationally invariant system, i.e., neglect boundary effects, so that $\langle\vec{S}_j\cdot\vec{S}_n\rangle$ will depend only on the relative separation between spins. The sums over target atoms in Eq. (4.22) then become

$$\sum_{j,n} e^{-i(\vec{k}_i-\vec{k}_a)\cdot\vec{R}_j}e^{-i(\vec{k}_b-\vec{k}_i)\cdot\vec{R}_n}\langle\vec{S}_j\cdot\vec{S}_n\rangle = \sum_{j,p} e^{-i(\vec{k}_i-\vec{k}_a)\cdot\vec{R}_j}e^{-i(\vec{k}_b-\vec{k}_i)\cdot(\vec{R}_p+\vec{R}_j)}\langle\vec{S}_0\cdot\vec{S}_{R_p}\rangle = \sum_{j,n} e^{i(\vec{k}_a-\vec{k}_b)\cdot\vec{R}_j}e^{-i(\vec{k}_b-\vec{k}_i)\cdot\vec{R}_n}\langle\vec{S}_0\cdot\vec{S}_{R_n}\rangle, \tag{4.23}$$

where the last equality is obtained by replacing p by n. Similarly,

$$\sum_{l,m} e^{-i(\vec{k}_a-\vec{k}_f)\cdot\vec{R}_l} e^{-i(\vec{k}_f-\vec{k}_b)\cdot\vec{R}_m}\langle\vec{S}_l\cdot\vec{S}_m\rangle = \sum_{l,m} e^{-i(\vec{k}_a-\vec{k}_b)\cdot\vec{R}_l} e^{i(\vec{k}_a-\vec{k}_f)\cdot\vec{R}_m}\langle\vec{S}_0\cdot\vec{S}_{R_m}\rangle. \tag{4.24}$$

At this point, the sum over j in Eq. (4.23) and the sum over l in Eq. (4.24) could be performed, the result of which for small-angle scattering from a macroscopic target would yield zero unless $|\vec{k}_a-\vec{k}_b| \lesssim l^{-1}$, where l represents a linear dimension of the target, i.e.,

$$\vec{k}_a \cong \vec{k}_b. \tag{4.25}$$

The exact equality, $\vec{k}_a=\vec{k}_b$, is rigorous only for a truly infinite spatial target, and using it, we would lose all target geometry which is inherent in multiple scattering from finite targets. Therefore, we will not perform the j, l sums at this stage, but rather perform the $\vec{k}_a, \vec{k}_b$ integrations first. Before we do and realizing that Eq. (4.25) must, none the less, be valid, we use it to simplify the factor f in Eq. (4.22), which is a much more slowly varying function of $\vec{k}_a$ and $\vec{k}_b$ than the exponential factors. Thus, from Eq. (4.21)

$$f(\vec{k}_a-\vec{k}_f, \vec{k}_i-\vec{k}_a, \vec{k}_f-\vec{k}_b, \vec{k}_b-\vec{k}_i) \cong f(\vec{k}_a-\vec{k}_f, \vec{k}_i-\vec{k}_a, \vec{k}_f-\vec{k}_a, \vec{k}_a-\vec{k}_i) = \tfrac{1}{36}. \tag{4.26}$$

Using Eqs. (4.14), (4.23), (4.24), and (4.26), we now perform the $\vec{k}_a, \vec{k}_b$ integrations in Eq. (4.22) and obtain for the cross section

$$\frac{d^2\sigma^{(2)}}{d\Omega\, d\omega} = \frac{4}{9}\left(\frac{ge^2}{m_e c^2}\right)^4 \frac{k_f}{k_i} \int d^3u_1 \int d^3u_2 \int d^3u_3 \int d^3u_4 e^{-i\vec{k}_f\cdot\vec{u}_1} e^{i\vec{k}_i\cdot\vec{u}_2} e^{i\vec{k}_f\cdot\vec{u}_3} e^{-i\vec{k}_i\cdot\vec{u}_4} \mathcal{S}(\vec{u}_1)\mathcal{S}(\vec{u}_2)\mathcal{S}(\vec{u}_3)\mathcal{S}(\vec{u}_4)$$
$$\times \sum_{jlmn} e^{i\vec{k}_l\cdot\vec{R}_n} e^{-i\vec{k}_f\cdot\vec{R}_m} \frac{e^{-ik_i|\vec{R}_l-\vec{R}_j-\vec{R}_m-\vec{u}_1+\vec{u}_2|}}{|\vec{R}_l-\vec{R}_j-\vec{R}_m-\vec{u}_1+\vec{u}_2|}$$
$$\times \frac{e^{ik_i|\vec{R}_l-\vec{R}_j-\vec{R}_n-\vec{u}_3+\vec{u}_4|}}{|\vec{R}_l-\vec{R}_j-\vec{R}_n-\vec{u}_3+\vec{u}_4|} \langle\vec{S}_0\cdot\vec{S}_{R_m}\rangle\langle\vec{S}_0\cdot\vec{S}_{R_n}\rangle \delta(\omega). \tag{4.27}$$

Since R_m and R_n appear in the spin correlation functions, they are restricted to microscopic distances by the spin correlation range, κ_1^{-1}, i.e., $|\vec{R}_m| \lesssim \kappa_1^{-1}$. Also, the $\vec{u}$ vectors are restricted by the normalized spin density functions $\mathcal{S}(\vec{u})$, which are of microscopic range. Therefore, since $\vec{R}_l$ and $\vec{R}_j$ are free to range over the entire target which is of macroscopic dimensions, the overwhelming contributions to the sums over j, l come when

$$|\vec{R}_l-\vec{R}_j| \gg |\vec{R}_m|, |\vec{R}_n|, |\vec{u}|.$$

Thus

$$|\vec{R}_l-\vec{R}_j-\vec{R}_m-\vec{u}_1+\vec{u}_2| \cong |\vec{R}_{lj}| - \hat{R}_{lj}\cdot(\vec{R}_m+\vec{u}_1-\vec{u}_2), \quad |\vec{R}_l-\vec{R}_j-\vec{R}_n-\vec{u}_3+\vec{u}_4| \cong |\vec{R}_{lj}| - \hat{R}_{lj}\cdot(\vec{R}+\vec{u}_3-\vec{u}_4), \tag{4.28}$$

where $\vec{R}_{lj}=\vec{R}_l-\vec{R}_j$. The exponential terms in Eq. (4.27) become

$$\frac{e^{-ik_i|\vec{R}_l-\vec{R}_j-\vec{R}_m-\vec{u}_1+\vec{u}_2|}}{|\vec{R}_l-\vec{R}_j-\vec{R}_m-\vec{u}_1+\vec{u}_2|} \cong |\vec{R}_{lj}|^{-1} e^{-ik_i|\vec{R}_{lj}|} e^{i\vec{q}\cdot(\vec{R}_m+\vec{u}_1-\vec{u}_2)}, \quad \frac{e^{ik_i|\vec{R}_l-\vec{R}_j-\vec{R}_n-\vec{u}_3+\vec{u}_4|}}{|\vec{R}_l-\vec{R}_j-\vec{R}_n-\vec{u}_3+\vec{u}_4|} \cong |\vec{R}_{lj}|^{-1} e^{ik_i|\vec{R}_{lj}|} e^{-i\vec{q}\cdot(\vec{R}_n+\vec{u}_3-\vec{u}_4)}, \tag{4.29}$$

where

$$\vec{q} = k_i\hat{R}_{lj} \tag{4.30}$$

represents the wave vector of magnitude k_i along the direction $\hat{R}_{lj}$. Identifying $\vec{R}_j$ as locating the first scattering event and $\vec{R}_l$ as locating the second scattering event, $\vec{q}$ is the intermediate wave vector of the neutron between scattering events.

Integrating Eq. (4.27) over ω, using Eqs. (4.29) and letting the sums over j, l, m, and n go over to integrals, retaining only the $\vec{\tau}=0$ reciprocal-lattice vector, we get for the double-scattering cross section,

$$\frac{d\sigma^{(2)}}{d\Omega} = \frac{4}{9}\left(\frac{ge^2}{v_0 m_e c^2}\right)^4 \int_V d^3R \int d^3R' R'^{-2} |F(|\vec{k}_i-\vec{q}|)|^2 |F(|\vec{q}-\vec{k}_f|)|^2 \hat{\gamma}(\vec{k}_i-\vec{q})\hat{\gamma}(\vec{q}-\vec{k}_f), \tag{4.31}$$

where

$$\hat{\gamma}(\vec{k}) = \int_{-\infty}^{\infty} d^3r \gamma(r) e^{i\vec{k}\cdot\vec{r}}, \tag{4.32}$$

$$\gamma(r) = \langle \vec{S}_0 \cdot \vec{S}_{\vec{r}} \rangle, \tag{4.33}$$

v_0 is the unit cell volume and we have let $\vec{R}_j \to \vec{R}$ and $\vec{R}_{ij} \to \vec{R}'$. The limits of integration on R, R' go over the target volume V. However, the limits for R' depend on R. We also have let the limits of integration in Eq. (4.32) extend to infinity with negligible error due to the short-range nature of $\gamma(r)$.

We use for $\gamma(r)$ the asymptotic expression for the spin correlation function first determined by Ornstein and Zernike[1] and given by

$$\gamma(r) = \frac{v_0 S(S+1)}{4\pi r_1^2} \frac{e^{-\kappa_1 r}}{r}, \tag{4.34}$$

where r_1 is a relatively temperature-insensitive microscopic length. Then Eq. (4.31) becomes

$$\frac{d\sigma^{(2)}}{d\Omega} = C_2 \int_V d^3R \int d^3R' R'^{-2} |F(|\vec{k}_i - \vec{q}|)|^2 |F(|\vec{q} - \vec{k}_f|)|^2 (|\vec{k}_i - \vec{q}|^2 + \kappa_1^2)^{-1} (|\vec{q} - \vec{k}_f|^2 + \kappa_1^2)^{-1} \tag{4.35}$$

where

$$C_2 = \frac{4}{9} \left(\frac{S(S+1)}{v_0 r_1^2} \right)^2 \left(\frac{e^2 g}{m_e c^2} \right)^4.$$

Remembering that $\vec{q}$ depends on $\hat{R}'$, it becomes evident that the double scattering depends on the target geometry. Now, using Eq. (4.30) for $\vec{q}$,

$$|\vec{k}_i - \vec{q}|^2 = 4k_i^2 \sin^2 \tfrac{1}{2}\theta_1, \quad |\vec{q} - \vec{k}_f|^2 = 4k_1^2 \sin^2 \tfrac{1}{2}\theta_2, \tag{4.36}$$

where the intermediate scattering angles θ_1 and θ_2 are depicted in Fig. 1. As the Curie temperature T_C is approached, $\kappa_1 \to 0$, the cross section, Eq. (4.35), is very sharply peaked around $\theta_1 = \theta_2 = 0$ and $\hat{R}'$ lies in a very small solid angle centered about $\vec{k}_i$ with vertex at $\vec{R}$. Therefore, defining $l(\vec{R})$ as the length from $\vec{R}$ in the direction of $\vec{k}_i$ to the boundary of the target (see Fig. 1), to a very good approximation, we may write

$$\frac{d\sigma^{(2)}}{d\Omega} = C_2 \int_V d^3R\, l(\vec{R}) \int_0^{2\pi} d\phi_1 \int_0^{\pi} d\theta_1 \sin\theta_1 (4k_i^2 \sin^2 \tfrac{1}{2}\theta_1 + \kappa_1^2)^{-1} (4k_i^2 \sin^2 \tfrac{1}{2}\theta_2 + \kappa_1^2)^{-1}$$

$$\equiv C_2 \int_V d^3R\, l(\vec{R}) G(k_i, \theta_S, \kappa_1). \tag{4.37}$$

The angle θ_2 will depend on ϕ_1, θ_1, and the scattering angle θ_S. We have also used the fact that form factors are slowly varying functions of their arguments and are normalized to unity, $F(0) = 1$. Defining

$$\langle l \rangle \equiv V^{-1} \int_V d^3R\, l(\vec{R}),$$

the critical magnetic double scattering cross section is

$$\frac{d\sigma^{(2)}}{d\Omega} = \tfrac{4}{9} V \langle l \rangle \left(\frac{S(S+1)}{v_0 r_1^2} \right)^2 \left(\frac{e^2 g}{m_e c^2} \right)^4 G(k_i, \theta_S, \kappa_1). \tag{4.38}$$

The length $\langle l \rangle$ will be of the order of magnitude of the linear dimension of the target. For a spherical target of radius R, $\langle l \rangle = \frac{3}{4}R$, and for a slab of thickness L, $\langle l \rangle = \frac{1}{2}L$. The integral $G(k_i, \theta_S, \kappa_1)$ is easily evaluated in general, but we choose to evaluate it in the small-angle approximation, i.e., $\sin\theta \cong \theta$, which gives

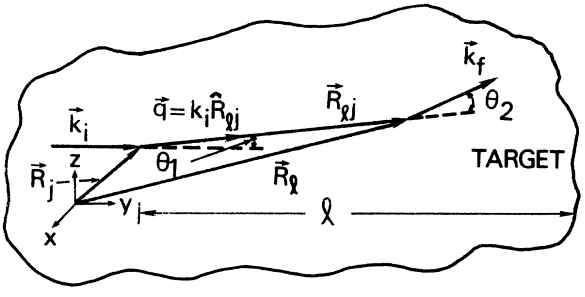

FIG. 1. Double-scattering geometry.

$$G=\frac{\pi}{k_i^2\kappa_1^2 x(x^2+4)^{1/2}}\ln\left(\frac{x^3+3x+(x^2+1)(x^2+4)^{1/2}}{(x^2+4)^{1/2}-x}\right), \tag{4.39}$$

where

$$x=k\kappa_1^{-1} \tag{4.40}$$

and $k=k_i\theta_S$, the wave-vector transfer.

With the same appropriate approximations made in this section, the first Born or single-scattering cross section is

$$\frac{d\sigma^{(1)}}{d\Omega}=N\frac{2}{3}\left(\frac{ge^2}{m_e c^2}\right)^2\frac{S(S+1)}{\kappa_1^2 r_1^2}(1+x^2)^{-1}. \tag{4.41}$$

Therefore, the combined scattering cross section, single plus double, from Eqs. (4.38) and (4.41) is given by

$$\frac{d\sigma}{d\Omega}=\frac{d\sigma^{(1)}}{d\Omega}+\frac{d\sigma^{(2)}}{d\Omega}=\frac{d\sigma^{(1)}}{d\Omega}[1+\beta H(x)], \tag{4.42}$$

where

$$\beta=\frac{2\pi\langle l\rangle\rho S(S+1)}{3k_i^2 r_1^2}\left(\frac{ge^2}{m_e c^2}\right)^2, \tag{4.43}$$

$$H=\frac{x^2+1}{x(x^2+4)^{1/2}}\ln\left(\frac{x^3+3x+(x^2+1)(x^2+4)^{1/2}}{(x^2+4)^{1/2}-x}\right), \tag{4.44}$$

and ρ is the density of target atoms. Since $H(0)=1$, β is the fraction of double scattering at $x=0$. Note that the double scattering will become more pronounced, i.e., β will increase as (i) ρ increases, (ii) the target size increases ($\langle l\rangle$ increases), and (iii) the neutron initial energy E_i decreases (k_i^2 decreases). In fact, double scattering will be more sensitive to the neutron's initial wave vector (β going like k_i^{-2}) than to target size.

We now return to the decomposition of the four-B-operator correlation function given in Eq. (3.4) and consider the first two terms which, up to now, have been neglected. The second term in Eq. (3.4) gives rise to a cross section for critical magnetic scattering that represents a quantum interference term and can be neglected for macroscopic targets. In terms of order of magnitude, this term goes like $(k_i l)^{-2}\, d\sigma^{(2)}/d\Omega$, where l represents a linear dimension of the target and $d\sigma^{(2)}/d\Omega$ is the cross section given by Eq. (4.38).

The first term in Eq. (3.4) gives rise to a cross section for critical magnetic scattering that represents a diffraction term and is essentially all forward scattering for macroscopic targets. This term is non-negligible only for scattering angles $\theta_S \lesssim (k_i l)^{-1}$.

A. Numerical results

Before any numerical results are possible, the magnitude of r_1, from Eq. (4.34) must be determined. In the derivation of Eq. (4.34) by Ornstein and Zernike, r_1 is given in terms of the second moment of the so-called "direct correlation function," a short-ranged function whose form depends on the Hamiltonian, Eq. (4.15). From mean-field theory, r_1 can be determined[14] and is given in terms of $J(\vec{R})$, the exchange energy in Eq. (4.15),

$$r_1^2=\frac{1}{6}\frac{\sum_{\vec{R}} R^2 J(\vec{R})}{\sum_{\vec{R}} J(\vec{R})}\ . \tag{4.45}$$

Thus, considering nearest-neighbor interactions only,

$$r_1\cong a_0/\sqrt{6}\ , \tag{4.46}$$

where a_0 is the nearest-neighbor distance.

Using Eq. (4.46) and relevant parameters from an experiment on Fe by Passell *et al.*,[15] the double-scattering cross section per target atom given by Eq. (4.38) is plotted in Fig. 2. Also plotted is the single-scattering cross section per target atom, Eq. (4.41).

It has been suggested[2] that the form of the OZ correlation function Eq. (4.34) should be modified by the introduction of a new critical exponent η,

$$\tilde{\gamma}(r)=A(\eta)\,e^{-\kappa_1 r}/r^{1+\eta}. \tag{4.47}$$

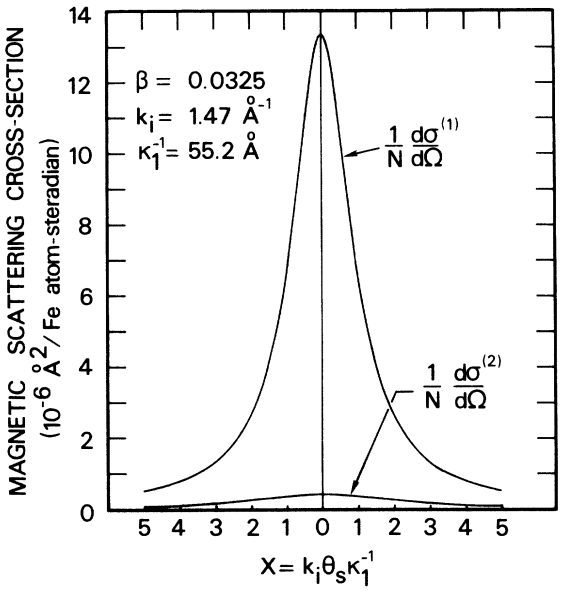

FIG. 2. Theoretical single- and double-scattering cross sections using relevant parameters from an experiment on Fe by Passell *et al*. Experimentally probed region corresponds to $2.1 \lesssim x \lesssim 3.9$.

This suggestion was motivated by the fact that an Ornstein-Zernike-Debye (OZD) plot of the reciprocal of the first-Born-scattering cross section, Eq. (4.41), vs x^2 should be linear, yet experimental plots for critical light scattering from liquids displayed a downward curvature for small x^2. The η-modified OZ correlation function $\tilde{\gamma}$ produces such a downward curvature. The possibility of an apparent η due to multiple (double) scattering effects has been investigated in connection with light scattering from liquids.[16] An OZD plot of Eq. (4.42), which includes both singly and doubly scattered neutrons, for Fe using experimental parameters of Ref. 15 is shown in Fig. 3, and a definite downward curvature is noticeable. This curvature is difficult to detect in experimental neutron work in ferromagnets owing to small-angle limitations. For example, the minimum value of x^2 in Fig. 3 attained by Passell *et al.* corresponds to $x^2 \cong 4.5$.

To obtain values of an apparent η due to double scattering effects, we proceed as follows. The coefficient $A(\eta)$ in Eq. (4.47) is determined by requiring that

$$\int d^3r\, \tilde{\gamma}(r) = \int d^3r\, \gamma(r). \tag{4.48}$$

Replacing $\gamma(r)$ by $\tilde{\gamma}(r)$ in the first-Born-scattering cross section, i.e.,

$$\frac{d\tilde{\sigma}}{d\Omega} = N\frac{2}{3}\left(\frac{ge^2}{m_e c^2}\right)^2 \frac{1}{v_0}\int d^3r\, \tilde{\gamma}(r) e^{-i\vec{k}\cdot\vec{r}}, \tag{4.49}$$

performing the integration, expanding in terms of η and retaining only terms linear in η, there results

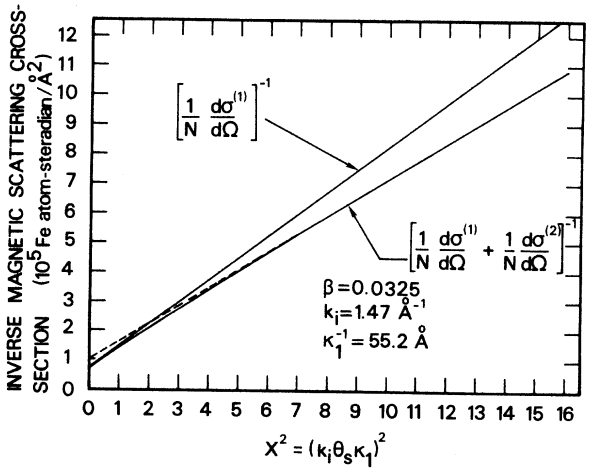

FIG. 3. Theoretical OZD plots of single-scattering and single-plus-double-scattering cross sections using relevant parameters from an experiment of Fe by Passell *et al.* Experimentally probed region corresponds to $4.5 \lesssim x^2 \lesssim 15.2$.

$$\frac{d\tilde{\sigma}}{d\Omega} = \frac{d\sigma^{(0)}}{d\Omega}\left[1 + \eta J(x)\right], \tag{4.50}$$

where

$$J(x) = 1 - x^{-1}\tan^{-1}x + \tfrac{1}{2}\ln(1+x^2) \tag{4.51}$$

and $d\sigma^{(1)}/d\Omega$ is given by Eq. (4.41). Now, equating Eq. (4.50) to the cross section containing both single and double scattering, Eq. (4.42), we obtain

$$\eta = K(x)\beta \tag{4.52}$$

where $K(x) \equiv H(x)/J(x)$, $H(x)$ given by Eq. (4.44). The function $K(x)$ is plotted in Fig. 4. As $T \to T_c$ for a fixed nonzero scattering angle, $x \to \infty$. In this limit, $\eta = 4\beta$. Therefore, for an experiment performed in this region, an apparent η, induced from double scattering, of 0.05 or greater would result for experimental conditions where $\beta \geq 0.0125$. The function $K(x)$ has a minimum of 2.5 at $x \cong 2$, therefore this would be the optimal region to probe experimentally in a search for a real η. However, even at $x \cong 2$, experiments for which $\beta \geq 0.02$ will produce an apparent $\eta \geq 0.05$.

Table I gives values of β corresponding to experimental conditions for several neutron scattering experiments from ferromagnetic crystals in which a value for η was sought. Also tabulated are the values for η experimentally measured.

V. DISCUSSION AND CONCLUSION

In this paper, we have presented a general formulation for multiple scattering of thermal neutrons and have applied it to double scattering. We have tried to assess the importance of double scattering on critical magnetic neutron scattering from ferromagnets slightly above their Curie temperatures. For typical experimental conditions, the double scattering produces small, but non-negligible deviations in Lorentzian line shape of magnetically scattered neutrons. In this regard, it com-

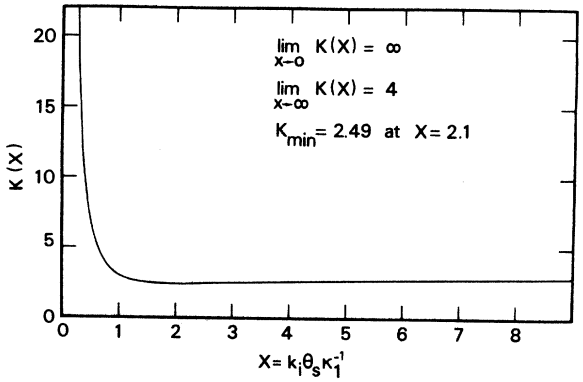

FIG. 4. Function $K(x)$ appearing in Eq. (4.51) vs x.

TABLE I. Experimental parameters and theoretical β values for several experiments on magnetic scattering of neutrons from ferromagnets.

Sample	k_i (Å^{-1})	r_1 (Å) [a]	Target size: Thickness (mm)	Experimental range of x	β	Experimental η
Fe [b]	1.47	1.01	12	$0.35 \lesssim x \lesssim 3.9$	0.032	c
			4.3		0.012	c
Fe [d]	5.03	1.01	2.5	$0.2 \lesssim x \lesssim 4.0$	0.000 58	0.07 ± 0.05
EuO [e]	4.30	1.48	<1	$0.1 \lesssim x \lesssim 5.4$	0.000 35	$-0.10 \lesssim \eta \lesssim 0.06$ [f]
EuS [e]	4.30	1.72	<1	$0.35 \lesssim x \lesssim 3.5$	0.000 17	$-0.07 \lesssim \eta \lesssim 0.11$ [f]

[a] Theoretically determined from Eq. (4.46).
[b] Reference 15.
[c] Curves with $\eta = 0.10$, 0.15, and 0.20 fitted experimental curve about as well as curve with $\eta = 0$.
[d] Reference 17.
[e] Reference 18.
[f] η determined by scaling relation: $\eta = 2 - \gamma/\nu$.

petes with similar line-shape deviations predicted by modifications in the spin correlations from their Ornstein-Zernike form used in a single-scattering theory.

Critical magnetic single scattering from antiferromagnets is pronounced in nonforward directions around magnetic Bragg peak positions. Double scattering, therefore, makes a negligible contribution.

Although our interests were primarily directed toward scattering near the critical region and in the quasielastic approximation, we have presented a formulation for double scattering which should be applicable to more general conditions of inelastic scattering.[19] To efficiently treat such cases when the target average $\langle B \rangle \neq 0$ (magnetic systems below the critical temperature, spin-wave scattering, or nuclear scattering from liquids) it would be desirable to write the probe-target interaction, $\phi = AB$, in the form $\phi = A\langle B \rangle + A(B - \langle B \rangle)$, so that target correlations involve fluctuations around their average value. Such a modification of the formulation given here presents no problems.[20]

The assumed interaction form $\phi = AB$, will also accomodate light scattering in the dipole approximation. The internal variables $\vec{a}$ and $\vec{b}_j$ described in Sec. III are then the photon polarization and target-atom dipole moment, respectively. However, for multiple light scattering, refraction, and extinction effects cannot be neglected[21] as they may be for neutrons. Also, the critical scattering of photons in the optical region is not restricted to the forward direction, therefore, determination of the target-size parameter $\langle l \rangle$ of Sec. IV is more complicated.[16]

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*Portions of this work are taken from a Ph.D. thesis submitted by W. A. Holm to the Georgia Institute of Technology, 1976.

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