Orbital effects in magnetic dynamics of thulium iron garnets

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A microscopic approach has been developed for studying the magnetic dynamics of thulium iron garnet (TmIG) based on Mori's general method of dynamical relaxation functions. The transverse magnetic excitation spectrum is analyzed. Calculating the static correlations in the mean-field approximation, the temperature dependence of both the magnetic excitations and their effective g factors are obtained. The latter displays qualitative and even quantitative agreement with the results of ferromagnetic resonance in TmIG. The emphasis is put on the crystal-field effects which essentially influence the behavior of the effective g factor. The linewidth of the resonance mode is also calculated assuming that the broadening results from the interaction of Tm^{3+} with the crystal lattice.

I. INTRODUCTION

Thulium iron garnets (TmIG) display an unusually low value of the effective g factor at room temperature.[1-4] Pure TmIG has a g value of 1.63.[1] Thulium iron garnets doped with nonmagnetic ions have even lower g values. As an example, for $Tm_{2.4}Bi_{0.6}Fe_{3.7}Ga_{1.3}O_{12}$, g is equal to 1.0 which is the largest deviation from $g=2$ reported in the literature.[2] Moreover, the effective g factor was observed to be increasing with temperature rather than decreasing, i.e., contrary to the prediction of the isotropic spin-wave theory with no relaxation effects.[3]

As the aim of this work is the microscopic interpretation of the anomalous behavior of the effective g factor of thulium iron garnets observed in microwave resonance, it seems to be judicious initially to study a simple model of pure TmIG. Then the approach developed for TmIG will be considered as the limiting case in the investigation of the magneto-optical properties of thulium iron garnets doped with nonmagnetic ions, especially with bismuth. The latter have been studied extensively and widely in recent years because of their interesting applications. The crucial role in our model is played by the thulium sublattice. Tm^{3+} ions are rather strongly coupled with the lattice, and due to the crystal field acting on them, as well as due to their rather fast relaxation, they influence the g factor immensely.

In TmIG the octahedral $[a]$ and the tetrahedral (d) sites are occupied by Fe^{3+} ions whose ground state is the orbital S state, ${}^6S_{5/2}$. Tm^{3+} ions occupy the dodecahedral positions $\{c\}$. The ground state of Tm^{3+} is ${}^3H_6$ with large orbital contribution to the total angular momentum operator, J. Because of the spinlike nature of their ground states, the iron ions in the first approximation are not affected by crystal fields. The magnetic properties of thulium, however, are extremely sensitive to the influence of crystal fields produced by the environment. The influence of the crystal field on the magnetic properties of TmIG, especially its effective g factor, is studied. The major complication in a theoretical study of this problem arises from the complexity of the crystal structure of garnets. Moreover, until now there has been insufficient information available on the crystal-field parameters in TmIG. Absolute values of crystal-field parameters are very hard to estimate both experimentally and theoretically since there is a considerable uncertainty about the radial extent of the $4f$ orbits and the shielding effects of the closed $5s$ and $5p$ shells. The crystal field at the thulium site is considered to be cubic with a trigonal distortion and the orthorhombic distortion is neglected. The unique axis is chosen to be the $\langle 111 \rangle$. Assuming the most probable crystal-field energy structure for Tm^{3+} permitted by the symmetry of its environment, the model is fitted to experimental data such as the temperature dependence of magnetization. Unfortunately there is an uncertainty concerning the occurrence of a compensation point in TmIG. Pauthenet[5] reports the compensation point between 4 and 15 K, whereas later results[6,7] do not confirm its occurrence. All authors, however, claim that thulium has a strong influence on the magnetization of TmIG, especially at

lower temperatures.

Both the iron-iron and thulium-iron exchange interactions are assumed to be isotropic with negative values of the exchange parameters between nearest neighbors.

The effective g factor is defined as a coefficient of the uniform spin-wave spectrum according to the microscopic mechanism of the resonance in which it is observed.

In order to analyze the spin-wave spectrum of TmIG, Mori's formalism of a continued-fraction representation of a dynamical relaxation function is used.[8,9] The detailed analysis of the spectrum is limited to the center of the zone ($\mathbf{q}=\mathbf{0}$). However, the formalism can be applied immediately to the other points in the zone. The frequency and linewidth of the modes are obtained and the effective g factors are calculated as the derivatives of the frequencies of the spectrum with respect to the magnetic field.

The relaxation mechanism postulated for the thulium sublattice is inferred from a comparison of the linewidths of ferromagnetic resonance in YIG with those of TmIG. In our approach the influence of the relaxation parameter on the effective g factor of the resonance mode is investigated as well as on the linewidth of the latter.

The static correlations, which appear within the framework of Mori's formalism, are treated in the mean-field approximation. It is not easy to establish the extent of the errors introduced by the latter approximation. However, it appears to work quite well for many rare-earth systems. Even here in the case of an antiferrimagnet, the approximation does not seem to be bad for our purpose since our calculations are carried out far from the Néel point.

The analysis of the spin-wave spectrum of rare-earth iron garnets was carried out earlier by Harris,[10] who uses the isotropic spin-wave approach. In the case of rare-earth garnets the application of such an approach is justified only for the iron garnets with gadolinium in its c sublattice, i.e., when crystal-field effects can be totally neglected.

Specifically the anomalous behavior of the effective g factor in rare-earth systems has been studied by Huber.[11] His approach will be discussed later in more detail since it has some points in common with the approach presented here. Yang, Cooper, Huang, and Sugawara[12] have investigated the influence of the exchange interaction on the effective g factor in paramagnetic cerium monopnictides also by using the Mori memory-function approach. The same memory-function technique has also been applied by Becker, Fulde, and Keller[13] to study the linewidth of the crystal-field excitations in metallic rare-earth systems.

The organization of this paper is as follows. In Sec. II our version of the general Mori's memory-function theory specified for TmIG is presented. The mean-field approximation and its results are discussed in Sec. III. In Sec. IV the excitation spectrum and its effective g factors in the mean-field random-phase approximation (RPA) are analyzed, including the extrinsic relaxation effects as well. Section V contains a brief summary of our conclusions.

II. THE THEORY

TmIG can be described by the following simplified Hamiltonian:

$$\mathcal{H}=-g_s\mu_B H^z\left[\sum_{i\in a}S^z_{ai}+\sum_{k\in d}S^z_{dk}\right]-g_J\mu_B H^z\sum_{f\in c}J^z_f+\sum_{f\in c}V_{\mathrm{CF}}(f)-\tfrac{1}{2}\sum_{i\neq j}J^{aa}_{ij}\mathbf{S}_{ai}\cdot\mathbf{S}_{aj}-\sum_{i,k}J^{ad}_{ik}\mathbf{S}_{ai}\cdot\mathbf{S}_{dk}-\tfrac{1}{2}\sum_{k\neq l}J^{dd}_{kl}\mathbf{S}_{dk}\cdot\mathbf{S}_{dl}-\sum_{k,f}J^{dc}_{kf}\mathbf{S}_{dk}\cdot\mathbf{J}_f\ ,$$

$$g_s=2\quad\text{and}\quad g_J=\tfrac{7}{6}\ , \tag{1}$$

where the external field H^z is applied along the direction of the resultant magnetization of TmIG, i.e., along the $\langle 111\rangle$ crystal axis. V_{CF} is the crystal-field Hamiltonian of the c sublattice. All the exchange Hamiltonians are postulated to be isotropic Heisenberg-like. In our further calculation the exchange interactions are limited to nearest neighbors, only. Moreover, from the start the c-c and c-a interactions are assumed to be negligible in comparison to the d-c ones. As already mentioned, the crystal field at the rare-earth site is treated as cubic with a trigonal distortion. The ortho-rhombic distortion is neglected with respect to the exchange field of the d sublattice. The crystal-field axis is along the threefold $\langle 111\rangle$ axis. The cubic crystal field splits the ground state of Tm^{3+} into three triplets, a doublet, and two singlets. The trigonal distortion removes the degeneracy of the triplets. In order to obtain all possible energy-level configurations of a trigonal field, six crystal-field parameters are needed. For lack of information about these parameters, the most probable configuration of the trigonal field energy levels of Tm^{3+} that accommodates the best fit to the temperature dependence of magnetization of TmIG (Refs. 6 and 7) is assumed. Thus the ground state of Tm^{3+} is the doublet Γ_3^T and the next excited state is the singlet Γ_1^T separated by the energy, Δ. It is also assumed that the remaining energy levels of Tm^{3+} are well separated from the two lowest-lying levels and, consequently, they have no significant effect on the magnetic properties of TmIG.

The operator J^z, which determines the order parameter of the c sublattice, takes the following form in the basis of the Tm^{3+} crystal-field states:

$$J^z=\eta(L_{11}-L_{22})\ , \tag{2}$$

where η is a fitting parameter in this model. In the mean-field approximation, the exchange field of the d sublattice splits the ground-state doublet Γ_3^T, producing two singlets with their respective magnetic moments equal to η and $-\eta$ (in Bohr magnetons). To fit η to the experimental values of the magnetization of the thulium sublattice at $T=0$ K, the value of $\eta=-1.085$ is required[7] and from now on η is treated as a fixed parameter.[7,14] $L^f_{\alpha\beta}(\alpha,\beta=1,2,3)$ are the Haley-Erdös standard-basis operators introduced for the Tm^{3+} single-ion energy structure.[15] These operators obey the following commutation relation:

$$[L^f_{\alpha\alpha'},L^{f'}_{\beta\beta'}]=\delta_{ff'}(\delta_{\alpha'\beta}L^f_{\alpha\beta'}-\delta_{\alpha\beta'}L^f_{\beta\alpha'})\ . \tag{3}$$

Any operator can be written as a linear combination of the $L_{\alpha\beta}$'s since they form a complete set.

In order to analyze the magnetic excitation spectrum of TmIG, Mori's memory-function formalism is applied.[8,9] The essential step in developing the formalism is to identify dynamical variables which are associated with the excitations that are to be studied. The aim of the interpretation of the ferromagnetic resonance is the appropriate choice for the transverse dynamical variables $S_{aq}^{\dagger}$, $S_{dq}^{\dagger}$, and $J_q^{\dagger}$, where

$$S_{aq}^{\dagger}=\sum_{\mathbf{R}+\gamma_a} e^{i\mathbf{q}\cdot(\mathbf{R}+\gamma_a)}S_{\mathbf{R}+\gamma_a}^{+} , \tag{4a}$$

$$S_{dq}^{\dagger}=\sum_{\mathbf{R}+\gamma_d} e^{i\mathbf{q}\cdot(\mathbf{R}+\gamma_d)}S_{\mathbf{R}+\gamma_d}^{+} , \tag{4b}$$

$$J_q^{+}=\sum_{\mathbf{R}+\gamma_c} e^{i\mathbf{q}\cdot(\mathbf{R}+\gamma_c)}J_{\mathbf{R}+\gamma_c}^{+} , \tag{4c}$$

and γ_a, γ_d, and γ_c denote the positions of respective ions in the unit cell and $\mathbf{R}$ is the translational vector of the lattice. As mentioned before, the actual calculations are performed for $\mathbf{q}=0$, i.e., for the center of the zone. Moreover, taking the Fourier transforms in the form given by Eq. (4) means that all the a sites are treated as equivalent and the same holds for the d and c sites, respectively. Consequently, the dimension of our problem is reduced to 3 instead of the original 64. At the a and d sites the spin operators $S_{ai}^{\dagger}$ and $S_{dk}^{\dagger}$, respectively, give rise to transitions between the six equally distant molecular-field energy levels. Each transition is accompanied by the change of the magnetic moment of a given ion. At the c site, in the molecular-field basis, the operators J_f^{+} and J_f^{-} take the following form:

$$J_f^{+}=\gamma(L_{31}^{f}-L_{23}^{f}) \tag{5a}$$

and

$$J_f^{-}=\gamma(L_{13}^{f}-L_{32}^{f}) , \tag{5b}$$

where γ is the crystal-field "transition" parameter which will be fitted to the experimental results. J_f^{+} and J_f^{-} can be interpreted as transition operators moving a Tm^{3+} ion between its mean-field energy levels originated from the Γ_3^T doublet and the nonmagnetic Γ_1^T singlet.

The three dynamical variables are then arranged in the form of a one-column matrix:

$$A_0=\begin{bmatrix} S_a^{+} \\ S_d^{+} \\ J^{+} \end{bmatrix} , \tag{6a}$$

and, consequently,

$$A_0^{\dagger}=[S_a^{-}S_d^{-}J^{-}] , \tag{6b}$$

where S_a^{+}, S_d^{+}, and J^{+} are the respective Fourier transforms with $\mathbf{q}=0$. The dynamical transversal relaxation function $(A_0(t),A_0^{\dagger})$ takes the form of a three-dimensional matrix whose elements are relaxation functions defined as follows:[8,9]

$$(A(t),B^{\dagger})=\int_0^{\beta}d\lambda\langle e^{\lambda H}e^{iHt}Ae^{-iHt}e^{-\lambda H}B^{\dagger}\rangle -\beta\langle A\rangle\langle B^{\dagger}\rangle , \tag{7}$$

where $\beta=(kT)^{-1}$ and the angular brackets denote thermal average.

The static ($t=0$) relaxation function, $(A^{\dagger},B^{\dagger})$, is the isothermal susceptibility, χ_{AB}.

The Laplace transform of the initial relaxation function matrix $(A_0(t),A_0^{\dagger})$ can be written in the following exact form,[8,9]

$$(A_0(i\omega),A_0^{\dagger})=[i\omega-i\omega_0+\Sigma(i\omega)]^{-1}(A_0,A_0^{\dagger}) , \tag{8}$$

where

$$(A_0(i\omega),A_0^{\dagger})=\int_0^{\infty}e^{-i\omega t}dt(A_0(t),A_0^{\dagger}) . \tag{8a}$$

$(A_0,A_0^{\dagger})$ is the static (isothermal) susceptibility matrix and $i\omega_0$ is the first-moment (frequency) matrix defined by

$$i\omega_0=(\dot{A}_0,A_0^{\dagger})(A_0,A_0^{\dagger})^{-1} , \tag{8b}$$

with $\dot{A}_0=dA_0/dt$.

$\Sigma(i\omega)$ is the Laplace transform of the memory-function matrix $(A_1(t),A_1^{\dagger})(A_0,A_0^{\dagger})^{-1}$ where

$$A_1=\dot{A}_0-i\omega_0A_0 \tag{9a}$$

and

$$A_1^{\dagger}=\dot{A}_0^{\dagger}-A_0^{\dagger}(i\omega_0)^{+} . \tag{9b}$$

The dynamical variables A_1 display the time dependence different from that of A_0:

$$A_1(t)=e^{it(1-P)\mathcal{H}}A_1e^{-it(1-P)\mathcal{H}} , \tag{10}$$

where the projection operator P is defined by

$$PX=(X,A_0^{\dagger})(A_0,A_0^{\dagger})^{-1}A_0 . \tag{10a}$$

In the mean-field approximation the inverse susceptibility matrix $(A_0,A_0^{\dagger})^{-1}$ takes the form

$$(A_0,A_0^{\dagger})_{\alpha\beta}^{-1}=(\chi_{0\alpha}^{\pm})^{-1}\delta_{\alpha\beta}-J_{\alpha\beta}(q=0)$$

where $\alpha,\beta=a,d,c$. (11)

$\chi_{0\alpha}^{\pm}$ is the single-ion isothermal transversal susceptibility which characterizes the sublattice and is expressed as follows:

$$\chi_{0\alpha}^{\pm}=\sum_{E_m\neq E_n}\frac{\langle m\mid S_{\alpha}^{+}\mid n\rangle\langle n\mid S_{\alpha}^{-}\mid m\rangle}{E_m-E_n}(P_n-P_m) , \tag{12}$$

where $\mid m\rangle$ are the single-ion mean-field eigenstates, E_m are the single-ion energy levels, and P_m are their thermal populations. S_{α}^{+} and S_{α}^{-} denote either the $S=\frac{5}{2}$ spin operators ($\alpha=a,d$) or the operators J^{+} and J^{-} ($\alpha=c$) defined by (5). $J_{\alpha\beta}$ ($\mathbf{q}=0$) is the Fourier transform of

the exchange integral.

Making use of the identity

$$(A^{\dagger},B^{\dagger})=-i\langle[A^{\dagger},B^{\dagger}]\rangle\ , \tag{13}$$

along with the commutation relations for the spin operators $S_a^{\pm},S_d^{\pm}$ and that for the operators

$$[J^{+},J^{-}]=(\gamma^2/\eta)J^z\ , \tag{14}$$

the frequency matrix is obtained in the following form:

$$(\omega_0)_{\alpha\beta}=\langle A_\alpha\rangle[(\chi_{0\alpha}^{\pm})^{-1}\delta_{\alpha\beta}-J_{\alpha\beta}(q=0)]\ , \tag{15}$$

where $A_\alpha=-S_a^z$, $-S_d^z$, and $(\gamma^2/\eta)J^z$ for $\alpha=a,d,c$, respectively.

The magnetic excitation frequencies are given by the eigenvalues of the matrix (15).

The damping is usually a complicated problem. Here a decoupling procedure is proposed followed by an approach that is similar to that of Huber.[11] First the memory-function matrix $(A_1(t),A_1^{\dagger})$ is rewritten in a more explicit form:

$$\begin{aligned}(A_1(t),A_1^{\dagger})=&(\dot{A}_0(t),\dot{A}_0^{\dagger})-(\dot{A}_0(t),A_0^{\dagger})(i\omega_0)^{+}\\&+(i\omega_0)(\dot{A}_0(t),A_0^{\dagger})\\&+(i\omega_0)(A_0(t),A_0^{\dagger})(i\omega_0)^{+}\ ,\end{aligned} \tag{16}$$

where the identity[8,9]

$$(A(t),\dot{B}^{\dagger})=-(\dot{A}(t),B^{\dagger}) \tag{17}$$

is used.

The equations of motion for the initial dynamical variables are

$$i\dot{S}_a^{+}=g_s\mu_B H^z S_a^{+}+\sum_{i,k}J_{ik}^{ad}(S_{ai}^{+}S_{dk}^z-S_{ai}^z S_{dk}^{+})\ , \tag{18a}$$

$$\begin{aligned}i\dot{S}_d^{+}=&g_s\mu_B H^z S_d^{+}+\sum_{i,k}J_{ik}^{ad}(S_{dk}^{+}S_{ai}^z-S_{dk}^z S_{ai}^{+})\\&+\sum_{k,f}J_{kf}^{dc}(S_{dk}^{+}J_f^z-S_{dk}^z J_f^{+})\ ,\end{aligned} \tag{18b}$$

$$\begin{aligned}i\dot{J}^{+}=&-g_J\mu_B H^z\eta J^{+}+\Delta\gamma(L_{31}+L_{23})\\&+\sum_{k,f}J_{kf}^{dc}(-\eta S_{dk}^z J_f^{+}+\gamma^2/\eta J_f^z S_{dk}^{+})\ .\end{aligned} \tag{18c}$$

Hence, the matrix elements of $(\dot{A}_0(t),A_0^{\dagger})$ contain three-operator relaxation functions which are decoupled in the following manner:

$$(A(t)S_\alpha^z(t),B^{\dagger})\rightarrow\langle S_\alpha^z\rangle(A(t),B^{\dagger})\ , \tag{19a}$$

$$(S_\alpha^z(t)A(t),B^{\dagger})\rightarrow\langle S_\alpha^z\rangle(A(t),B^{\dagger})\ , \tag{19b}$$

where $S_\alpha^z=S_a^z$, S_d^z, and J^z for $\alpha=a,d,c$, respectively. Thus the products of the order parameters and the two-operator transversal relaxation functions are obtained. They have a time dependence that is different from that of the analogous initial relaxation functions.

To the matrix elements of $(\dot{A}_0(t),\dot{A}_0^{\dagger})$, the same decoupling procedure is applied twice: first to the time-derivatives on the left-hand side of the relaxation functions, and, then, using the identity (17) the procedure of (19) is applied once again. The decoupling procedure used here is equivalent to that of the mean-field approximation so it is consistent with the rest of our calculations.

Aside from the decoupling, the approximation that is postulated is similar to that of Huber.[11] It is assumed that the possible damping is associated with the thulium sublattice and is only indirectly influenced by the iron sublattices. This assumption is realized by setting $J_{dc}=0$. It is a well-known fact that the resonance lines in TmIG are much broader than those in YIG, which are exceptionally narrow. It indicates the importance of thulium in the damping mechanism and supports these assumptions. With $J_{dc}=0$ the normalized memory-function matrix is obtained with all its elements equal to zero save the last one, which takes the following form:

$$\begin{aligned}&[(A_1(t),A_1^{\dagger})(A_0,A_0^{\dagger})^{-1}]_{cc}\\&\qquad=-\Delta^2(J^{+}(t),J^{-})_0(\chi_{0c}^{\pm})^{-1}\ ,\end{aligned} \tag{20}$$

where $(J^{+}(t),J^{-})_0$ is the single-ion relaxation function. Next, the following time dependence is introduced into the latter:

$$(J^{+}(t),J^{-})_0=(\chi_{0c}^{\pm})^{-1}e^{-t/\tau_c}\ , \tag{21}$$

where τ_c is the Tm^{3+} relaxation parameter related to the ion's transitions between the crystal-field doublet and singlet states. At this point of the discussion Huber's idea of intrinsic and extrinsic damping is utilized.[11] Extrinsic damping is not associated with Hamiltonians of the type given by Eq. (1). In this model it results from the direct interactions between the total-angular-momentum fluctuations of Tm^{3+} and the heat reservoir. The decay rate of the fluctuations is determined by τ_c. The intrinsic damping arises from interactions between the total- (or spin-) angular-momentum fluctuations. The decoupling in the form of Eq. (19), however, excludes the higher-order effects, so this discussion is restricted to the extrinsic damping. Although Huber[11] suggests that in systems having superexchange interactions, an intrinsic relaxation can be of great importance. This may not be true for TmIG, for due to the large orbital contribution to the total angular momentum of Tm^{3+}, extrinsic damping, coming from the direct interactions between magnetic degrees of freedom and the heat reservoir, is the most important relaxation mechanism.[16] The problem of intrinsic damping is still open to further discussion. A similar approach to extrinsic damping has been suggested by Lehmann-Szweykowska[17] for rare-earth systems with the Kramers degeneracy using the density-matrix formalism. Later in the numerical calculations different values of τ_c are used to estimate its influence on the effective g factor.

Of course, the Laplace transform of the normalized memory-function matrix also has one nonzero element:

$$[(A_1(i\omega),A_1^{\dagger})(A_0,A_0^{\dagger})^{-1}]_{cc}=i\frac{\Delta^2\omega}{\omega^2+\Gamma_c^2}-\frac{\Delta^2\Gamma_c}{\omega^2+\Gamma_c^2}\ , \tag{22}$$

where $\Gamma_c=\hbar/\tau_c$ ($\hbar$ is Planck's constant).

The last step of our procedure involves the transformation of the normalized memory-function matrix in order to express it in the same basis for which the frequency matrix takes its diagonal form. After some very simple algebra the normalized memory-function matrix with all nonzero matrix elements is obtained. The diagonal matrix elements consist of imaginary and real parts which determine the shift in the frequency caused by the damping effects and the linewidth, respectively. The influence of the off-diagonal matrix elements has been omitted in this analysis.[11]

The poles of the Laplace transform of the initial transverse relaxation function in Eq. (8) can finally be expressed as follows:

$$\omega_i'=\omega_i+\frac{V_i}{U}\frac{\Delta^2\omega_i}{\omega_i^2+\Gamma_c^2}+i\frac{V_i}{U}\frac{\Delta^2\Gamma_c}{\omega_i^2+\Gamma_c^2}\ , \tag{23}$$

where

$$V_i=\frac{\omega_k-\omega_l}{(\omega_i-\omega_{cc})(\omega_k-\omega_{aa})(\omega_l-\omega_{aa})}\ , \tag{23a}$$

and

$$U=\sum_{i=1}^{3}\frac{\omega_k-\omega_l}{(\omega_i-\omega_{aa})(\omega_k-\omega_{cc})(\omega_l-\omega_{cc})}\ , \tag{23b}$$

where $i=1,2,3$, $k=2,3,1$, and $l=3,1,2$; ω_i $(i=1,2,3)$ are the eigenvalues of the frequency matrix ω_0 and ω_{aa} and ω_{cc} are two of its diagonal matrix elements as seen in Eq. (15).

From Eq. (22), both the shift in frequency and the linewidth are originally frequency dependent. In Eq. (23), using $\omega=\omega_i$ $(i=1,2,3)$ in the expressions for the respective ω_i means that the corrected values of frequency are not expected to differ significantly from the mean-field results.

Due to the transformation of the memory-function matrix, the damping effects are obtained which at the beginning are associated with the c sublattice only. Now they are influenced indirectly by both iron sublattices.

For every branch of the magnetic transversal excitation spectrum, a value of the effective g factor can be calculated remembering that the magnetic moment of a state is given by the derivative with respect to the magnetic field of the energy of that state. Hence

$$g_{\text{eff}}=\frac{1}{\mu_B}\frac{\partial\omega_i}{\partial H^z}\ , \tag{24}$$

where i labels the modes.

It is obvious that the formula for g_{eff} is very complicated and the results can only be discussed numerically. The g_{eff} is expressed as a function of the order parameters for the three sublattices, i.e., $\langle S_a^z\rangle$, $\langle S_d^z\rangle$, and $\langle J^z\rangle$, the longitudinal isothermal susceptibilities χ_a^{zz}, χ_d^{zz}, and χ_c^{zz}, and the transverse single-ion susceptibility for the c sublattice, $\chi_{0c}^{\pm}$. These static correlations will be calculated in the mean-field approximation.

III. MEAN-FIELD RESULTS

The order parameters of the three sublattices are obtained as solutions of the following self-consistent equations:

$$\langle S_a^z\rangle=\frac{5\sinh\left[\frac{5h_a}{2kT}\right]+3\sinh\left[\frac{3h_a}{2kT}\right]+\sinh\left[\frac{h_a}{2kT}\right]}{2\left[\cosh\left[\frac{5h_a}{2kT}\right]+\cosh\left[\frac{3h_a}{2kT}\right]+\cosh\left[\frac{h_a}{2kT}\right]\right]}\ , \tag{25a}$$

$$\langle S_d^z\rangle=\frac{5\sinh\left[\frac{5h_d}{2kT}\right]+3\sinh\left[\frac{3h_d}{2kT}\right]+\sinh\left[\frac{h_d}{2kT}\right]}{2\left[\cosh\left[\frac{5h_d}{2kT}\right]+\cosh\left[\frac{3h_d}{2kT}\right]+\cosh\left[\frac{h_d}{2kT}\right]\right]}\ , \tag{25b}$$

$$\langle J^z\rangle=\eta(P_+-P_-)\ , \tag{25c}$$

where

$$h_a=g_s\mu_B H^z+J_{aa}\gamma_{aa}\langle S_a^z\rangle+J_{ad}\gamma_{ad}\langle S_d^z\rangle\ , \tag{26a}$$

$$h_d=g_s\mu_B H^z+J_{dd}\gamma_{dd}\langle S_d^z\rangle+J_{ad}\gamma_{da}\langle S_a^z\rangle+J_{dc}\gamma_{dc}\langle J^z\rangle\ , \tag{26b}$$

$$P_{\pm}=e^{\pm\eta h_c/kT}/Q_c\ ,$$

$$Q_c=e^{\eta h_c/kT}+e^{-\eta h_c/kT}+e^{-\Delta/kT}\ , \tag{26c}$$

$$h_c=g_J\mu_B H^z+J_{dc}\gamma_{cd}\langle S_d^z\rangle\ .$$

J_{aa}, J_{ad}, J_{dd}, and J_{dc} are values of the exchange integrals between nearest neighbors of the respective sublattices,

$$\gamma_{ij}(\mathbf{q})=\sum_{j'}e^{i\mathbf{q}\cdot(\mathbf{r}_i-\mathbf{r}_{j'})}$$

where the sum is over nearest-neighboring ions in the jth sublattice.

TmIG is a ferrimagnet. In order to solve Eqs. (25), the following ground states are assumed as a starting point for the three sublattices at $T=0$ K:

$$S_a^z \left| -\tfrac{5}{2} \right\rangle = -\tfrac{5}{2} \left| -\tfrac{5}{2} \right\rangle , \tag{27a}$$

$$S_d^z \left| \tfrac{5}{2} \right\rangle = \tfrac{5}{2} \left| \tfrac{5}{2} \right\rangle , \tag{27b}$$

$$J^z \left| \eta \right\rangle = \eta \left| \eta \right\rangle . \tag{27c}$$

As mentioned before, η is the fixed parameter ($\eta = -1.085$).[6,7] It is assumed that the values of the exchange integrals J_{aa}, J_{ad}, and J_{dd} are the same as those for YIG (Ref. 18) and that they are all negative. The values of Δ and J_{dc} have been chosen to fit the experimental magnetization data[6,7] of TmIG, with M, defined by

$$M = 3g_s \langle S_d^z \rangle + 2g_s \langle S_a^z \rangle + 3g_J \langle J^z \rangle . \tag{28}$$

The results are shown in Fig. 1 and the best agreement between the experimental and calculated temperature variation of M has been achieved for $\Delta = 40$ K and $J_{dc} = -16$ K.

The longitudinal magnetic susceptibilities, defined as

$$\chi_i^{zz} = \frac{\partial \langle A_i \rangle}{\partial H^z} , \tag{29}$$

where $A_i = S_a^z$, S_d^z, and J^z for $i = a,d,c$, respectively, are solutions to the system of the three mean-field linear equations:

$$\chi_a^{zz}(1 - J_{aa}\gamma_{aa}\chi_a^0) - \chi_d^{zz} J_{ad}\gamma_{ad}\chi_a^0 = g_s \chi_a^0 , \tag{30a}$$

$$-\chi_a^{zz} J_{ad}\gamma_{da}\chi_d^0 + \chi_d^{zz}[1 - J_{dd}\gamma_{dd}\chi_d^0] - \chi_c^{zz} J_{dc}\gamma_{cd}\chi_d^0 = g_s \chi_d^0 , \tag{30b}$$

$$-\chi_d^{zz} J_{dc}\gamma_{cd}\chi_c^0 + \chi_c^{zz} = g_J \chi_c^0 , \tag{30c}$$

where the single-ion longitudinal susceptibilities are of the Curie-Langevin type:

$$\chi_a^0 = \beta[\langle (S_a^z)^2 \rangle - \langle S_a^z \rangle^2] , \tag{31a}$$

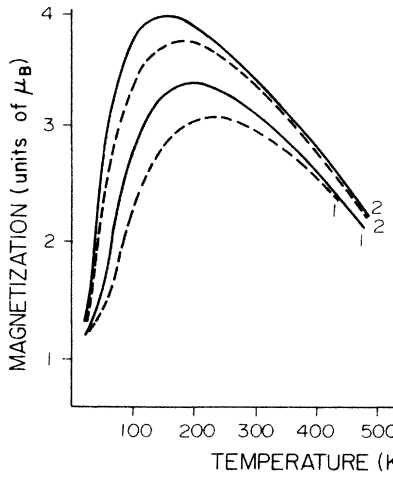

FIG. 1. The magnetization of TmIG vs temperature for different crystal-field and exchange parameters of Tm^{3+}. Solid line: 1, $\Delta = 40$ K, $J_{dc} = -16$ K; 2, $\Delta = 40$ K, $J_{dc} = -8$ K. Dotted line: 1, $\Delta = 400$ K, $J_{dc} = -16$ K; 2, $\Delta = 400$ K, $J_{dc} = -8$ K.

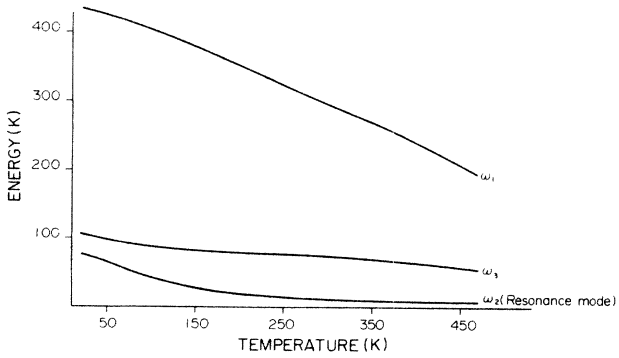

FIG. 2. Energies of the transverse magnetic modes of TmIG vs temperature. The resonance mode is indicated. $\Delta = 40$ K, $J_{dc} = -16$ K, $\gamma = 1$.

$$\chi_d^0 = \beta[\langle (S_d^z)^2 \rangle - \langle S_d^z \rangle^2] , \tag{31b}$$

$$\chi_c^0 = \beta[\langle (J^z)^2 \rangle - \langle J^z \rangle^2] \quad \text{and} \quad \beta = (kT)^{-1} . \tag{31c}$$

IV. DYNAMICAL PROPERTIES OF TmIG

The spectrum of transverse magnetic excitations consists of three branches as might be expected. Because of the rare-earth ions on the c sublattice and the importance of crystal-field effects on that ion, the nature of magnetic excitations in TmIG is not pure spin-wave-like. Magnetic excitations in TmIG are combinations of spin waves and magnetic excitons.

For all sets of the parameters Δ, J_{dc}, and γ, the temperature variation of the spectrum has the same pattern as that shown in Fig. 2. Two spin-wave-like modes are obtained: the acoustical mode (ω_2), i.e., the resonance mode, and the optical high-lying mode (ω_1). Both display a rather pronounced temperature dependence. The frequency of the third mode (ω_3) is on the order of the crystal-field energy gap, Δ, and is less temperature dependent than the remaining branches. So it is concluded that the third mode represents a rather excitonic

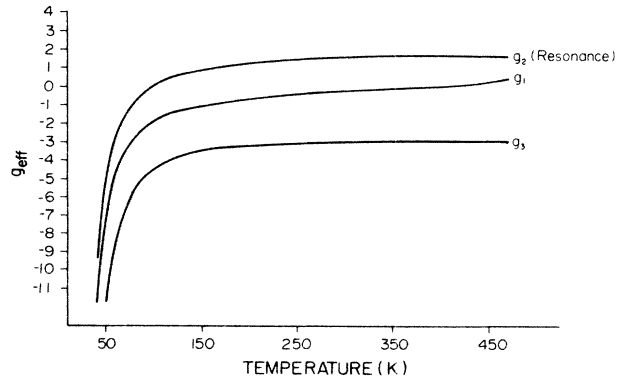

FIG. 3. Effective g factors for the three transversal magnetic modes vs temperature; g_{eff} of the resonance mode is indicated. $\Delta = 40$ K, $J_{dc} = -16$ K, $\gamma = 1$.

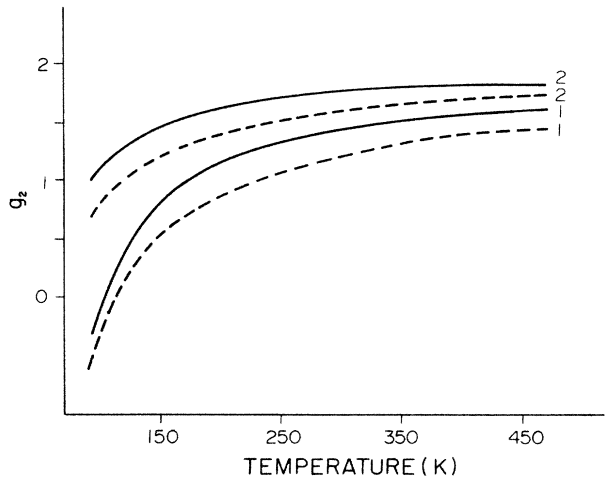

FIG. 4. Effective g factors of the resonance mode vs temperature for different crystal-field and exchange parameters of Tm^{3+}. Solid line: 1, $\Delta=40$ K, $J_{dc}=-16$ K, $\gamma=1$; 2, $\Delta=40$ K, $J_{dc}=-8$ K, $\gamma=1$. Dotted line: 1, $\Delta=400$ K, $J_{dc}=-16$ K, $\gamma=1$; 2, $\Delta=400$ K, $J_{dc}=-8$ K, $\gamma=1$.

type of resonance. The temperature dependence of the modes is stronger at lower temperatures where a singularity in the temperature variation of the magnetization is also observed. Unfortunately the analysis does not include the problem of dispersion. These results apply only to the center of the zone, i.e., the wave vector $\mathbf{q}=\mathbf{0}$.

The effective g factors for different sets of values of Δ and J_{dc} are shown in Figs. 3 and 4. Even omitting any damping, the temperature variation of the effective g factor of the resonance mode remains in good agreement, both qualitatively and quantitatively, with the experimental data.[1,2,4]

As seen in Fig. 3, all three effective g factors are increasing with temperature which contrasts strongly with the prediction of the isotropic spin-wave theory. Or, going even deeper, it is the large orbital contribution to the total angular momentum of Tm^{3+} that is most influential. For such ions it is necessary to include crystal-field effects. Another consequence is the strong coupling to the lattice which is an important relaxation mechanism. It is tacitly assumed that this relaxation mechanism is dominating in these systems and it is included in the form of extrinsic damping of independent thulium ions modified in accordance with Mori's formalism by the fractional susceptibilities of all magnetic degrees of freedom. Intrinsic damping is excluded as already mentioned. As seen in Figs. 5 and 6, this restricted damping does not influence significantly the former result, i.e., with no damping. In Fig. 7 the linewidth of the resonance mode is shown where the numerical results have been obtained assuming τ_c is on the order of 10^{-12}–10^{-13} s.

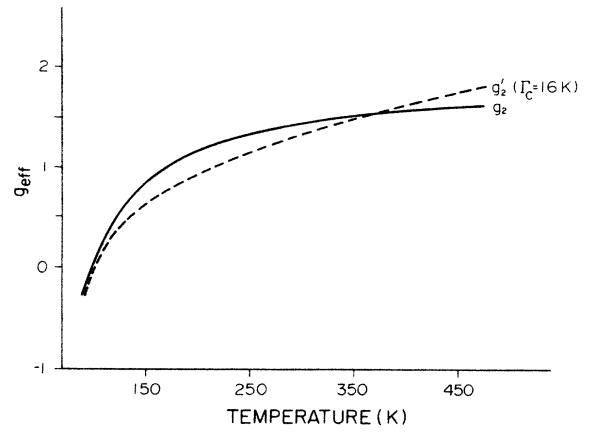

FIG. 6. g_{eff} of the resonance mode vs temperature with (dotted line) and without (solid line) damping. $\Delta=40$ K, $J_{dc}=-16$ K, $\gamma=1$.

V. SUMMARY

In summary, a microscopic approach has been developed for the transversal magnetic excitations in

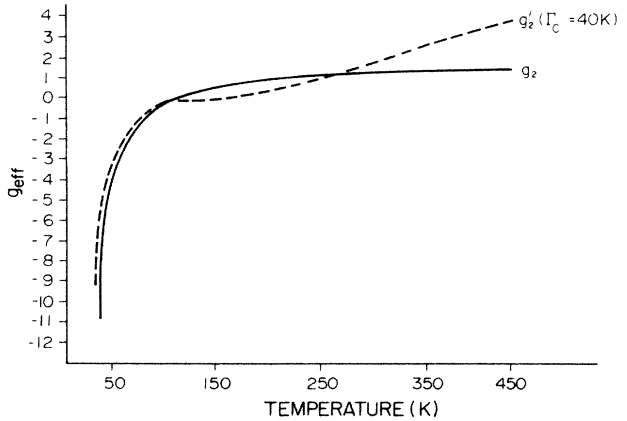

FIG. 5. g_{eff} of the resonance mode vs temperature with (dotted line) and without (solid line) damping. $\Delta=400$ K, $J_{dc}=-16$ K, $\gamma=1$.

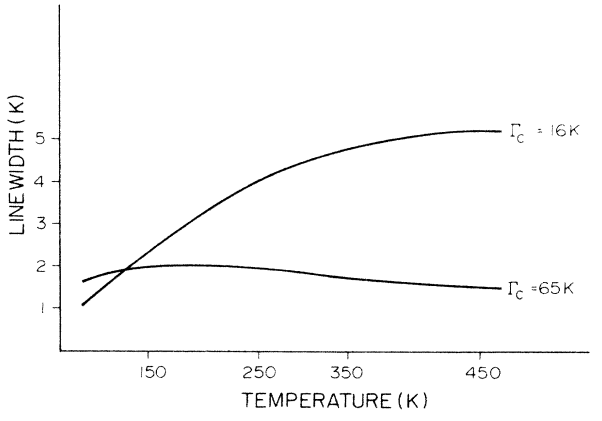

FIG. 7. Linewidth of the resonance mode vs temperature for two different values of $\tau_c=\hbar/\Gamma_c$. $\Delta=40$ K, $J_{dc}=-16$ K, $\gamma=1$.

iron garnets with thulium in the c sublattice. From a comparison with experimental data it is concluded that our approach, which considers the large orbital contribution to the total angular momentum of Tm^{3+}, is successful in interpreting the qualitative features of the results and to a certain extent, even the quantitative ones. The modified Mori's theory allows the determination of the temperature variation both of the eigenfrequencies and the linewidth of the transverse magnetic excitations as well as the effective g factor as functions of the appropriate fractional static correlations. The latter are obtained in the mean-field approximation. The analysis of damping is for extrinsic damping but it could be equally well evaluated for an intrinsic damping.

Further studies are needed. It would be important to go beyond the mean-field approximation in obtaining the static correlation functions. Moreover, further analysis of the damping mechanism is called for. The crystal-field energy structure has been assumed in this model and experimental measurements of the crystal field in TmIG are desirable.

Nevertheless our preliminary microscopic analysis predicts the origin of the anomalous behavior of the effective g factor and its low value at room temperature. It has been shown that the large orbital contribution to the total angular momentum of Tm^{3+} accounts for the anomalous temperature variation of g_{eff}.

From this analysis, as well as that from other papers,[1] it can be concluded that the same behavior of g_{eff} can be predicted by at least three different models: a crystal field, a strong anisotropy field, or very fast relaxation of the thulium ion. However, all can be considered as orbital effects. The conclusion is that the orbital contribution of the thulium ions lays the foundation for their interesting and unusual magnetic properties.

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