Relaxation mechanism for ordered magnetic materials

C. Vittoria and S. D. Yoon

Center for Microwave Magnetic Materials and Integrated Circuits, ECE Department, Northeastern University, Boston, Massachusetts 02115, USA

A. Widom

Physics Department, Northeastern University, Boston, Massachusetts 02115, USA (Received 20 November 2009; revised manuscript received 2 December 2009; published 15 January 2010)

We have formulated a relaxation mechanism for ferrites and ferromagnets (insulators and metals) whereby the coupling between the magnetic motion and lattice is based purely on continuum arguments concerning magnetostriction. This theoretical approach contrasts with previous mechanisms based on microscopic formulations of spin-phonon interactions employing a discrete lattice. Our model explains the scaling of the intrinsic ferromagnetic resonance linewidth with frequency, with temperature { $\propto 1/M_s(T)$ } and the anisotropic nature of magnetic relaxation in ordered magnetic materials. Here, $M_s(T)$ is the thermal saturation magnetization. Without introducing adjustable parameters, our model is in reasonable quantitative agreement with experimental measurements of the intrinsic magnetic resonance linewidths of important class of ordered magnetic materials including both insulators and metals.

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I. INTRODUCTION

Since the discovery of magnetic resonance, the physics community has been fascinated with possible mechanisms to explain the absorption linewidth or the relaxation time in magnetic materials. It was, and still is, a very challenging problem. Magnetic relaxation is important to understand because it affects a number of technologies, including computer, microwave, electronics, nanotechnology, and medical applications. Ultimately, the physical limitation of any technology which incorporates magnetic materials of any size, shape, and combinations thereof comes down to precise knowledge of the relaxation time of the magnetic material being utilized.

The background of various calculations or formulations of magnetic relaxation for the past 60 years or so can be summarized briefly as follows: (i) the relaxation times in paramagnetic materials¹ are characterized by two parameters, T_1 and T_2 , wherein T_2^{-1} describes the magnetic resonance linewidth and T_1 describes the time taken for the external magnetic field Zeeman energy density $-\mathbf{H}_{ext} \cdot \mathbf{M}$ to relax into thermal equilibrium. These times have been modeled in terms of various coupling schemes, i.e., spin-spin and/or spin-lattice interactions.² Since the coupling between spins is relatively weak, as it should be in a paramagnetic material, the coupling to the lattice involves discrete spin sites rather than a collective cluster of spins. As such, paramagnetic coupling is necessarily microscopic in nature. For example, a microscopic coupling scheme was formulated³ whereby a spin Hamiltonian was modulated by the lattice motion. Variants to this approach have been very successful in explaining relaxation in paramagnetic materials. (ii) The magnetic relaxation of ferrimagnetic or ferromagnetic resonance (FMR) linewidth is characterized by the Gilbert parameter⁴ α , or equivalently by the Landau-Lifshitz parameter⁵ λ_L . In such equations of motion, the magnitude of the magnetization M $=|\mathbf{M}|$ remains constant in time. Thus spins rotate collectively in the Gilbert equation of motion so that this may occur.

Much of the successful microscopic approaches or formulations utilized in paramagnetic materials were transferred over to models⁶ which attempted to explain the Gilbert equations. In some sense this presented a contradiction or paradox which was conveniently ignored. As it is well known that collective excitations in a ferrimagnetic or ferromagnetic state can be adequately described in terms classical continuum models, although microscopic descriptions remain perhaps more accurate.⁷ To our knowledge very few or perhaps any microscopic models have been successful in explaining the origin of the Gilbert equations. For example, much attention was given in 1970s to explain the FMR linewidth in yttrium iron garnet $(Y_3Fe_5O_{12})$ since its linewidth was the narrowest ever measured in a ferrimagnetic material.⁸ Clearly, there was less to explain, and perhaps spin-lattice interactions could be treated at discrete spin sites as in paramagnetic materials. These calculations⁸ contained many approximations and predicted an FMR linewidth about 1/10 to 1/100 of the measured linewidth. We believe that this is the best agreement between theory and experiment on relaxation in an ordered magnetic material. The purpose of this work is to improve upon the predictability of a theoretical model not only on a given material but in general for any ordered magnetic materials without restoring to any approximations and assumptions.

We have adopted a conventional continuum magnetomechanical description of the magnetic and elastic states of the ferrimagnetic or ferromagnetic crystal.^{9–11} The advantage of this description is that the microscopic spin-lattice coupling need not be formulated since it has already been included in the continuum model which has been proved to be experimentally correct. We introduce a thermodynamic argument stating that the heat exchange between the magnetic and elastic systems must treated as the same. As such, the Gilbert equations may be directly related to the elastic sound wave relaxation time and the coupling strength between the magnetic and elastic systems. Specifically, we will show that $\tilde{\alpha}$ is proportional to the square of the magnetostriction constants and inversely proportional to $\gamma M \tau$ wherein τ is the elastic relaxation time. In addition, the model predicts that $\tilde{\alpha}$ cannot be presumed to be a scalar as it often has been done in the past; i.e., $\tilde{\alpha}$ is predicted to be an anisotropic second rank tensor in a single-crystal material. Some other workers have previously noted the tensor nature of the Gilbert damping coefficients.^{12,13}

It is clear that one needs an interaction between phonons and electron spins to account for Gilbert damping parameter α . Suhl¹⁴ has considered such coupling schemes. The Gilbert damping parameter can be thought of as a transport coefficient in much the same way as conductivity and/or viscosity are transport coefficients. Such transport coefficients describe heating processes by which otherwise long-lived modes are damped. One can in fact relate the Gilbert damping parameter to conductivity and/or viscosity. For metallic ferromagnetic materials, conductivity as well as the equivalent electron viscosity produces a considerable amount of magnetic damping via eddy current heating. For magnetic insulators it is the viscosity which determines the magnetic damping. As it is well known, conductivity and viscosity can be nonzero even in zero-frequency limit. Hence, the implied Gilbert damping parameter is also nonzero at zero frequency. Remarkably our final expression for the Gilbert damping still hold true for metals wherein same electronic excitations dominate both the magnetic damping and the viscosity. Electronic viscosity is well known to dominate sound-wave attenuation in metals.¹⁵ In Suhl and Hickey and Moodera's¹⁶ papers they find, in the limit of zero frequency and zero wave number, that the real part of α is zero. This limiting case suggests that they have not included the zero-frequency transport coefficients consistently in their theory. In our derivation the expected result at zero frequency occur naturally in our formalism. In general, we believe the very nature of discreteness (as in paramagnetic materials) gives rise to relatively long magnetic relaxation times. However, the magnetic relaxation time of a coherent collection of spins (as in FMR) implies shorter relaxation times since it involves collective acoustic waves in the interaction scheme. Our present theoretical treatment takes this into account via the continuum magnetomechanics.

In Sec. II the Gilbert equations of motion are reviewed and the role of a dissipative magnetic intensity \mathbf{H}' is explained. In Sec. III, the thermodynamic notion of adiabatic magnetostriction coefficients are given as an adiabatic response in the strain to a magnetic moment change. Equivalently, there will be an adiabatic change in the magnetic field to an applied crystal stress. In Sec. IV, the heating rates due to Gilbert FMR damping and viscous sound-wave damping will be reviewed. A rigorous relationship is derived between the viscous heating rate for strain waves and the Gilbert heating rate for changing magnetic moments. This is a central result of this work. In Sec. V, a detailed comparison is made with previous experiments and with predictions of our theoretical works. Finally, in the concluding Sec. VI, we make a summary and a detailed comparison between our model and predictions of previous theoretical works.

II. GILBERT EQUATION

A distinguishing feature of the collective coherent magnetic moments in FMR is that the magnitude of the magnetization, $M = |\mathbf{M}|$ remains fixed which requires a magnetic resonance equation of the simple form

$$\frac{d\mathbf{M}}{dt} = \gamma \mathbf{M} \times \mathbf{H}_{tot},\tag{1}$$

wherein the gyromagnetic ratio $\gamma = ge/2mc$. The *total* magnetic intensity \mathbf{H}_{tot} has a thermodynamic part determined by the energy per unit volume u,

$$du = Tds + \mathbf{H} \cdot d\mathbf{M} + \boldsymbol{\sigma}: d\boldsymbol{e}, \qquad (2)$$

or the enthalpy per unit volume w,

$$dw = Tds + \mathbf{H} \cdot d\mathbf{M} - \boldsymbol{e}: d\boldsymbol{\sigma},\tag{3}$$

wherein σ is the crystal stress and e is the crystal strain. There is a dissipative part **H**' of the magnetic intensity determined by the Gilbert tensor $\tilde{\alpha}$,

$$\mathbf{H}' = \left(\frac{1}{\gamma M}\right) \widetilde{\boldsymbol{\alpha}} \cdot \frac{d\mathbf{M}}{dt}.$$
 (4)

All together

$$\mathbf{H}_{tot} = \mathbf{H} + \mathbf{H}',$$

$$\mathbf{H}_{tot} = \left(\frac{\partial w}{\partial \mathbf{M}}\right)_{s,e} + \left(\frac{1}{\gamma M}\right) \widetilde{\boldsymbol{\alpha}} \cdot \frac{d\mathbf{M}}{dt}.$$
 (5)

Eqs. (1) and (5) imply that all components of the magnetization must relax simultaneously in a way which conserves the magnitude of the magnetization.

III. MAGNETOSTRICTION

The adiabatic magnetostriction coefficients are defined in thermodynamics as

$$2\Lambda_{ijkl}M_k = M^2 \left(\frac{\partial e_{ij}}{\partial M_l}\right)_{s,\sigma} = -M^2 \left(\frac{\partial H_l}{\partial \sigma_{ij}}\right)_{s,\mathbf{M}},\tag{6}$$

wherein a thermodynamic Maxwell relation has been applied to Eq. (3). For adiabatic changes in time for the magnetization,

$$\dot{\mathbf{M}} = M\dot{\mathbf{N}}$$
 wherein $\mathbf{N} = \frac{\mathbf{M}}{M}$, (7)

Eq. (7) implies the time-varying magnetostrictive strain as given by

$$\dot{e}_{ij} = 2\Lambda_{ijkl} N_k \dot{N}_l. \tag{8}$$

Equation (8) describes magnetostriction as a time-varying strain e which results from a time-varying unit vector **N** in the direction of the magnetization.

IV. HEATING RATES

From Eq. (4), it is evident that the heating rate per unit volume due to the dissipative magnetic intensity \mathbf{H}' obeys

$$\dot{Q} = \frac{d\mathbf{M}}{dt} \cdot \mathbf{H}',$$

$$\dot{Q} = \left[\frac{1}{\gamma M}\right] \frac{d\mathbf{M}}{dt} \cdot \tilde{\alpha} \cdot \frac{d\mathbf{M}}{dt},$$

$$\dot{Q} = \frac{M}{\gamma} \dot{N}_i \alpha_{ij} \dot{N}_j,$$
(9)

and $\tilde{\alpha}$ is a second rank tensor

$$\widetilde{\alpha} = \begin{pmatrix} \alpha_{xx} & \alpha_{xy} & \alpha_{xz} \\ \alpha_{yx} & \alpha_{yy} & \alpha_{yz} \\ \alpha_{zx} & \alpha_{zy} & \alpha_{zz} \end{pmatrix}.$$
 (10)

In virtue of the magnetoelastic effect,¹⁷ a changing magnetization will produce a changing strain as in Eq. (8). Finally, the fourth rank crystal viscosity tensor, η_{ijkl} , determines the heating rate per unit volume due to the time-dependent strain

$$\dot{Q} = \dot{\boldsymbol{e}}_{ij} \eta_{ijkl} \dot{\boldsymbol{e}}_{kl}.$$
(11)

Employing Eqs. (8) and (11) and comparing the result to Eq. (9) yields the central result of our model. For any crystal symmetry the Gilbert damping tensor due to magnetostriction coupling is *rigorously* given by

$$\alpha_{ij} = \left[\frac{4\gamma}{M}\right] (\Lambda_{nmpi} N_p) \eta_{nmrl} (\Lambda_{rlqj} N_q).$$
(12)

The following properties of the Gilbert damping tensor in Eq. (12) are worthy of note: (i) the Gilbert damping tensor $\tilde{\alpha}$ is inversely proportional to the magnetization magnitude M. (ii) The Gilbert damping tensor $\tilde{\alpha}$ is proportional to the squares of the magnetostriction tensor elements. (iii) The tensor nature of $\tilde{\alpha}$ dictates that the magnetic relaxation is *anisotropic*. Finally, Eq. (12) can be derived on a microscopic fluctuation-dissipation theorem basis as shown in Appendix.

V. COMPARISON WITH EXPERIMENT

To a sufficient degree of accuracy, one may employ an average of the form

$$\alpha = \frac{1}{3} \operatorname{tr}\{\widetilde{\alpha}\} = \left[\frac{\alpha_{xx} + \alpha_{yy} + \alpha_{zz}}{3}\right]$$
(13)

defining a scalar function α . (iv) The crystal viscosity tensor η_{nmrl} may be employed to describe the acoustic-wave damping.¹⁸ For a mode label *a*, e.g., a longitudinal (a=L) or a transverse (a=T) mode, the acoustic absorption coefficient at frequency ω is given by¹⁸

$$\tau_a^{-1} = \frac{\omega^2 \eta_a}{2\rho v_a^2},\tag{14}$$

wherein v_a is the acoustic-mode velocity and ρ is the mass density. Finally, for a cubic crystal, there are only two *independent* magnetoelastic coefficients which may be defined

$$\Lambda_{xxxx} = \frac{3}{2} \lambda_{100} \equiv -\left(\frac{B_1}{c_{11} - c_{22}}\right),$$
$$\Lambda_{xyxy} = \frac{3}{2} \lambda_{111} \equiv -\left(\frac{B_2}{2c_{44}}\right),$$
(15)

wherein the Cauchy three index magnetostriction coefficients are λ_{ijk} and the Cauchy elastic moduli are c_{ij} .

The Gilbert damping factor α may be deduced from the measurement of the intrinsic FMR linewidth. However, the measurement of the intrinsic linewidth is, indeed, very difficult. The reason for this conclusion is that there are too many extrinsic effects that influence the measurement. For example, in ferromagnetic metals such as Ni, Co, and Fe the intrinsic linewidth contribution to the total linewidth measurement^{19,20} may be between 10% and 30%. The rest of the linewidth²¹ may be due to exchange-conductivity effects.

However, there may be other contributions, such as magnetostatic excitations, surface roughness, volume defects,²² crystal quality, interfaces,²³ size, excitation of high-order spin waves, etc.. Similar conclusions apply to ferrites except there are no exchange-conductivity effects.²¹ Thus, the reader should be mindful that when we quote or cite an experimental intrinsic value of the linewidth it may be an overestimation for there can be some hidden extrinsic contributions in an experiment. However, we have relied on data well established over the years. The criteria that we have adopted in choosing an ensemble of intrinsic linewidth measurements are the ones exhibiting the narrowest linewidths ever measured in the single-crystal materials. In addition, we required full knowledge of their elastic, magnetic, and electrical properties.^{19–21,24} The objective is not to introduce any adjustable parameters.

The experimental value of Gilbert damping parameter α_{exp} may be deduced from the FMR linewidth ΔH at frequency *f* as

$$\alpha_{\exp} = \frac{\sqrt{3}}{2} \left(\frac{\gamma \Delta H}{2 \pi f} \right). \tag{16}$$

The factor $\sqrt{3}/2$ assumes Lorentzian line shape of the resonance absorption curve. The theoretical Gilbert damping parameter α_{th} value is expressed in terms of known²⁰ parameters so that there are no adjustable parameters in our comparison to experiments, as shown in Table I. The theoretical prediction for the Gilbert damping parameter is that

$$\alpha_{\rm th} = \frac{36\rho\gamma}{M\tau} \left[\frac{\lambda_{100}^2}{q_L^2} + \frac{\lambda_{111}^2}{q_T^2} \right],\tag{17}$$

wherein ρ is the mass density, $q_T \approx v_T \frac{M}{2\gamma A}$ is the transverseacoustic propagation constant, q_L is the longitudinal-acoustic propagation constant, v_T is the transverse sound velocity, A is the exchange stiffness constant, λ_{100} and λ_{111} are magnetostriction constants for a cubic crystal magnetic material. The transverse-acoustic propagation constant was approximated on the basis that the relaxation process conserved energy and wave vector. Since the acoustic frequency is fixed in the process the longitudinal propagation constant may be also calculated to be $q_L = q_T(v_T/v_L)$ for magnetic materials,

Materials	$q_T (10^{-6} \text{ cm}^{-1})$	$\lambda_{100} \ (10^{-6})$	$\lambda_{111} (10^{-6})$	M (G/4 π)	$\begin{array}{c} A\\ (10^{-6} \text{ erg/cm}) \end{array}$	ΔH (Oe)	f (GHz)	$\tau (10^{-13} \text{ s})$	$\alpha_{\rm th}~(10^{-5})$	$\alpha_{\exp} (10^{-5})$
Y ₃ Fe ₅ O ₁₂ ^a	3.8	1.25	2.8	139	0.40	0.33	9.53	4.4	5.56	9.0
Y ₃ Fe ₄ GaO ₁₂ ^a	1.46	-1	-1	36	0.28	3.0	9.53	4.4	51	76
Li _{0.5} Fe _{2.5} O ₄ ^b	8.6	-8	+0	310	0.40	2.0	9.50	1.5	26	50
NiFe ₂ O ₄ ^b	7.49	-63	-26	270	0.40	35	24.0	1.5	710	350
MgFe ₂ O ₄ ^b	9.30	-10	-1	90	0.1	2.3	4.9	1.5	120	120
MnFe ₂ O ₄ ^b	6.6	-30	-5	220	0.4	238	9.2	1.5	930	1040
BaFe ₁₂ O ₁₉ ^c	9.6		15	350	0.4	6	55	1.5	18	26
Ni ^d	6.3	-46	25	484	0.75	102	9.53	1.8	770	2600
Fe ^d	8.75	20	-20	1690	1.9	9	9.53	1.8	30	220
Co ^d	5.1		80	1400	2.78	15	9.53	1.8	530	370

TABLE I. Calculated and measured Gilbert damping (α) parameters.

^aGarnets.

^bSpinels.

^cHexagonal ferrite.

^dFerromagnetic materials [note: longitudinal-acoustic wave constant is $q_L = (v_T/v_L)q_T$].

wherein v_L is the longitudinal sound wave velocity.

In Fig. 1, we plot the experimental and theoretical values Gilbert damping constants as given by Eqs. (16) and (17). We note that the agreement between theory and experiment is remarkable in view of the fact that any of the cited parameters could differ from the ones listed in Table I by as much as 20-30 %. For example, the linewidth reported in Table I may not be on the same sample where the elastic or magnetic parameters were cited. In a few cases we needed to extrapo-



FIG. 1. (Color online) Shown are the experimental and theoretical values of the Gilbert damping constants as given by Eqs. (16) and (17). The data was statistically analyzed employing a likelihood method with independent normal distributions of the slopes with unit variance. The likelihood function is shown in the insert as a function of the slope $s = \alpha_{ex} / \alpha_{th}$. We find that $s = 1.25 \pm 0.78$ with 95% confidence and within one standard deviation matches the theory. The uppermost and lowermost curves represent, respectively, the maximum and minimum error values of the theory. The central curve represents the maximum likelihood estimate.

late the value of A since there was no published value. For example, magnetostatic mode excitations have a deleterious effect on the dependence of the FMR linewidth on size. Most, if not all, previous FMR linewidth measurements have been performed on slabs, whiskers, etc., which can indeed support magnetostatic mode excitations. Additional complications arise as a result of exchange-conductivity excitations in magnetic metals. In magnetic metals after separating pure eddy current and/or exchange-conductivity losses the remainder of intrinsic linewidth is described by the electron viscosity. Thus our theory still holds true for electronic excitations in ferromagnetic metals. The spin-wave scattering contributions cannot be resolved within the statistical accuracy of the experimental data. Nevertheless, the agreement between theory and experiment is quite satisfactory.

VI. CONCLUSIONS AND DISCUSSIONS

Qualitative and quantitatively our model is in agreement with experimental observations of the intrinsic FMR linewidth reported over the years. Specifically, experimentally the most important characteristics of the intrinsic FMR linewidth, ΔH , measured on ordered magnetic materials (metal or insulator) for the past 50 years are that ΔH scales with frequency and $\frac{1}{M}$.^{19,25,26} Indeed, these are the predictions of our theory. In addition, ΔH scales with the magnetostriction constant squared, see Fig. 1. Figure 1 was plotted in a logarithmic scale only to be able to include all of the data in Table I.

Another prediction of our theoretical work is that the Gilbert damping parameter $\tilde{\alpha}$ is not simply a scalar parameter but a tensor quantity. This implies that the FMR linewidth is intrinsically anisotropic in single crystals of ferrimagnetic-ferromagnetic materials. There was much controversy in 1970s about whether or not the intrinsic linewidth should be anisotropic or not. Poor quality of samples seemed to have incited the controversy. Improved or more accurate angular linewidth data²⁵ supports the notion of an anisotropic line-

Relationships	$\alpha(\omega=0)$	$\alpha \sim \omega^n$	$\alpha \sim (1/M)^n$	$(B_1)^2$	$(B_2)^2$	\tilde{lpha}
Our model ^a	≠0	n = 0	n = 1	Yes	Yes	Tensor
Ref. 27 ^b	$\neq 0$	n = 0				Tensor
Ref. 28 ^c	$\neq 0$	n = 0	n = 1	Yes		Tensor
Ref. 16 ^d	$\neq 0$	n = 1		Yes		Tensor
Ref. 29 ^e	$\neq 0$	n = 1	n = 1			Scalar
Ref. 30 ^f	$\neq 0$	n = 0				Scalar
Ref. 31 ^g	$\neq 0$	n = 0				Scalar
Ref. 32 ^h	$\neq 0$	n = 0				Scalar
Ref. 14 ⁱ	=0	n=0				Scalar
Ref. 33 ^j	=0	n = 0	n = -1	Yes	Yes	Scalar
Experiment	$\neq 0$	n = 0	n = 1	Yes	Yes	Tensor

TABLE II. Functional dependence of Gilbert damping (α) parameters.

^aTheory predictions for both ferromagnets and ferrimagnets.

^bTheory predictions for Ref. 27.

^cTheory predictions for ferrimagnets.

^dTheory predictions for Ref. 16.

^eTheory predictions for ferromagnets.

^fTheory predictions for ferromagnets.

^gTheory predictions for both ferromagnets and ferrimagnets.

^hTheory predictions for ferromagnets.

ⁱTheory predictions for both ferromagnets and ferrimagnets.

^jTheory predictions for both ferromagnets and ferrimagnets.

width in ordered magnetic materials in agreement with our model.

In Table II we have listed the theoretical predictions of other models as well as ours and compared with experiments, both insulators and metals. In Eq. (42) of Ref. 28, the final expression for Gilbert damping does not contain the viscosity while our Eq. (12) contains viscosity as a central feature of our model. The reason for the difference is that there were no microscopic terms leading to sound-wave attenuation in the model of Ref. 28. In Table II, the (-) mark indicates the case where no predictions were made by a given model. Also, experiments do not include data taken at very low temperatures where the anomalous skin effects are important in metals, for example. In summary, we believe that the comparison between theory and experiment is very encouraging in terms of continuing this continuum approach to explain intrinsic linewidths in ordered magnetic materials.

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APPENDIX: KUBO FORMULAS

The Kubo transport expressions for the viscosity tensor reads as follows: for large volume V, let the mean fluctuation in the stress read

$$\bar{\sigma}_{ij}(t) = \frac{1}{V} \int_{V} \Delta \sigma_{ij}(\mathbf{r}, t) d^{3}\mathbf{r}, \qquad (A1)$$

and consider the correlation function

$$G_{ijkl}(t-t') = V \langle \bar{\sigma}_{kl}(t') \bar{\sigma}_{ij}(t) \rangle.$$
(A2)

With

$$\beta = \frac{\hbar}{k_B T},\tag{A3}$$

the Kubo formula for viscosity is

$$\eta_{ijkl} = \frac{1}{\hbar} \int_0^\infty \int_0^\beta \lim_{V \to \infty} G_{ijkl}(t+i\lambda) d\lambda dt.$$
 (A4)

Similarly let the mean fluctuation in the magnetic intensity read

$$\bar{h}_{ij}(t) = \frac{1}{V} \int_{V} \Delta H_i(\mathbf{r}, t) d^3 \mathbf{r}, \qquad (A5)$$

and consider the correlation function

$$F_{ij}(t-t') = V\langle \overline{h}_j(t')\overline{h}_i(t)\rangle.$$
(A6)

The Kubo formula for the Gilbert damping tensor reads

$$\frac{\alpha_{ij}}{\gamma M} = \frac{1}{\hbar} \int_0^\infty \int_0^\beta \lim_{V \to \infty} F_{ij}(t+i\lambda) d\lambda dt.$$
(A7)

Since Eq. (6) implies

$$\bar{h}_l = -\frac{2}{M} \Lambda_{ijkl} N_k \bar{\sigma}_{ij}, \qquad (A8)$$

it follows that

$$F_{ij}(t) = \frac{4}{M^2} (\Lambda_{nmpi} N_p) G_{nmrl}(t) (\Lambda_{rlqj} N_q), \qquad (A9)$$

putting our central result

$$\alpha_{ij} = \left[\frac{4\gamma}{M}\right] (\Lambda_{nmpi} N_p) \eta_{nmrl} (\Lambda_{rlqj} N_q), \qquad (A10)$$

on a firm microscopic transport coefficient basis.

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