Molecular field theory analysis of magneto-optic sensitivity of gallium-substituted yttrium iron garnets

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The temperature dependence of the magneto-optic sensitivity of gallium-substituted yttrium iron garnets was measured at 1.3 μm and compared with a model based on molecular field theory. The model incorporates results of measurements of both the saturation magnetization and saturation Faraday rotation versus temperature. These measurements were analyzed in the context of molecular field theory to extract the fundamental molecular field coefficients and the magneto-optical coefficients as functions of gallium content. The model and direct sensitivity measurements both indicate that the magneto-optic sensitivity of garnet compositions with gallium substitution levels near 0.8 should exhibit a vanishing first-order temperature sensitivity. [S0003-6951(96)03831-4]

Magneto-optic magnetic field sensors based on iron garnets exhibit a variety of characteristics attractive for magnetic field sensing including small size, wideband frequency response, compatibility with fiber optics, and sensitivity now approaching 1 pT/$\sqrt{\text{Hz}}$.[1] Both the magneto-optic sensitivity and the temperature dependence of the sensitivity of these materials are highly composition dependent. Previous studies have demonstrated that temperature sensitivity in the iron garnet family $(Tb_xY_{1-x})_3Fe_5O_{12}$ is minimized at a specific value of x.[2] A different study indicated that gallium-substituted yttrium iron garnet (Ga:YIG) might also be a candidate garnet family for minimization of temperature sensitivity.[3]

For magneto-optic magnetic field sensing, the key material parameters that determine both the magneto-optic sensitivity and the temperature dependence of the sensitivity are the saturation magnetization M_{sat} and saturation Faraday rotation $\theta_{F,\text{sat}}$. Both parameters are temperature and composition dependent, whereas $\theta_{F,\text{sat}}$ is also wavelength dependent. For bulk iron garnets, the standard model for the magneto-optic sensitivity is described by the equation

$$S=\frac{\theta_{F,\text{sat}}}{N_D M_{\text{sat}}}, \tag{1}$$

where N_D is the geometrical demagnetization factor.[4]

Iron garnets are generally ferrimagnetic; that is, they are characterized by two (or even three) magnetic sublattices which, though physically superimposed, have distinct electronic, magnetic and optical characteristics. Gallium substitutes for iron in yttrium iron garnet in both the octahedral and tetrahedral crystal sublattices. The explicit composition of this family of garnets can, thus, be written $Y_3[Fe_{2-x}Ga_x](Fe_{3-y}Ga_y)O_{12}$, where the brackets indicate the constituents of the octahedral sublattice and the parentheses correspond to the tetrahedral sublattice. For Ga:YIG, the magnetizations of the two magnetic sublattices are effectively antiparallel for $y<2$ and $x<0.7$.[5] (Rare earth iron garnets, including $(Tb_xY_{1-x})_3Fe_5O_{12}$ are more complicated because of additional magnetic effects caused by the rare earth ions in the dodecahedral sublattice.) For such compositions, the temperature dependence of the saturation magnetization is well described by the Néel model of ferrimagnetism. The macroscopic saturation magnetization M_{sat} in this model is given simply by the difference between the magnetizations of the tetrahedral and octahedral sublattices,

$$M_{\text{sat}}(T)=M_d(T)-M_a(T), \tag{2}$$

where a and d refer to the octahedral and tetrahedral sublattices, respectively. In the Néel model, $M_d(T)$ and $M_a(T)$ are governed by the Brillouin function. The primary material-dependent parameters of these functions are known as the molecular field coefficients. For Ga:YIG, these coefficients depend linearly on x and y.[6] Thus, in the Néel model, specific values of x and y correspond to unique $M_{\text{sat}}(T)$ functions. Conversely, x and y can themselves be independently determined by fitting the Néel model (through Brillouin functions) to measurements of $M_{\text{sat}}(T)$.[5]

Magneto-optical effects in Ga:YIG originate from magnetic and electric dipole transitions in both the octahedral and tetrahedral sublattices. Expressing the saturation Faraday rotation as a first-order power series expansion of the sublattice magnetizations, yields

$$\theta_{F,\text{sat}}(T)=AM_a(T)+DM_d(T), \tag{3}$$

where A and D are the wavelength-dependent magneto-optical coefficients of the a and d sublattices. Similar equations can be used to describe the Faraday ellipticity. Higher-order product terms in the power series expansion of $\theta_{F,\text{sat}}$ produce somewhat better fits to experimental data at the expense of having to incorporate additional model parameters.[7]

Gallium-substituted YIG compositions generally exhibit smaller M_{sat} values than pure YIG. At the wavelength 1.3 μm, gallium substitution also generally reduces values of $\theta_{F,\text{sat}}$. In a comparative sense, however, gallium substitution reduces the magnetization more than it reduces the saturation Faraday rotation.[3] Equation (1), thus, predicts that gallium substitution should have a net effect of increasing magneto-optic sensitivity. This predicted effect has been confirmed by experiment.[4]

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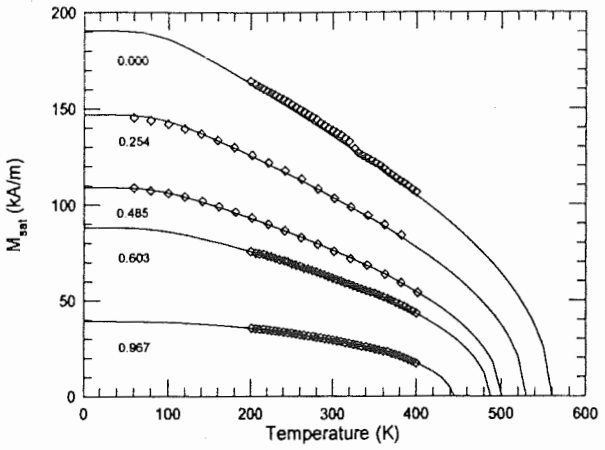

FIG. 1. Saturation magnetization vs temperature for YIG and four Ga:YIG compositions. Values printed underneath theoretical curves indicate best-fit gallium substitution z ($z=x+y$) for each sample.

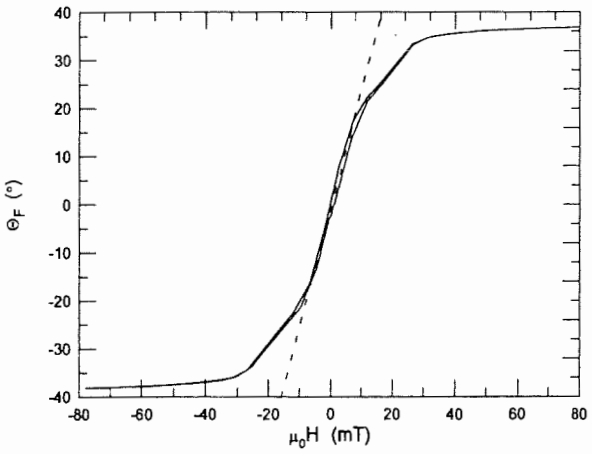

FIG. 2. Typical magneto-optic response function curve. S is calculated from the slope of the linear low-field portion of the curve indicated by the dashed line.

The molecular field coefficients of Ga:YIG, which allow calculation of both $M_a(T)$ and $M_d(T)$, are known linear functions of x and y. To predict the temperature dependence of either the saturation magnetization or the saturation Faraday rotation, x and y must, thus, be experimentally determined. Moreover, because the relative distribution of gallium ions substituted into the tetrahedral and octahedral sublattices depends on specific growth processes that cannot be directly controlled, x and y must be treated as independent material parameters.[5]

Determination of x and y was based on analysis of superconducting quantum interference device (SQUID) magnetometer measurements of $M_{sat}(T)$. A fitting algorithm based on the Néel model was applied to $M_{sat}(T)$ measurements from 60 to 400 K to determine x and y for four Ga:YIG compositions. Flux-grown single-crystal cylinders with diameters ranging from 2 to 5 mm and lengths ranging from 1 to 2.5 mm were characterized. The room-temperature saturation magnetizations of these compositions ranged from approximately 24 to 110 kA/m (300 to 1400 G). A comparison of the experimental $M_{sat}(T)$ SQUID measurements with theoretical curves based on the best-fit x and y values is shown in Fig. 1. In the figure legend, values of the gallium substitution z, formed simply by the sum of the best-fit x and y values used to generate the theoretical curves, are indicated underneath each corresponding theoretical curve. Measurements made on unsubstituted YIG are included in Fig. 1 for reference. The agreement between the best-fit theoretical curves and the experimental $M_{sat}(T)$ measurements supports the validity of the Néel ferrimagnetism model for these compositions. Table I lists the actual best-fit values of x, y, and z as well as the corresponding molecular field coefficients for the five compositions.

Magneto-optical measurements in this study were all made at 1.3 μm. This is a particularly popular wavelength for iron garnet-based magneto-optical sensors both because of the optical and magneto-optical properties exhibited by iron garnets at this wavelength and because of the availability of a wide variety of optical sources. Magneto-optic response functions $\theta_{F,sat}(H)$ were measured for each sample by sweeping the applied magnetic field H through one complete cycle and simultaneously recording the corresponding Faraday rotation with a polarization–modulation apparatus.[8] For each sample, response functions were measured at temperatures between 173 and 450 K.

Generally, the magneto-optic response functions generated by these measurements exhibit a low-field region characterized by low hysteresis and linear behavior bounded by nonlinear transitional behavior at larger fields, eventually exhibiting saturation. A typical response function is shown in Fig. 2. The two quantitative parameters extracted from these curves are the magneto-optic sensitivity S (defined by the slope of the low-field linear region) and the saturation Faraday rotation $\theta_{F,sat}$.

The magneto-optical coefficients A and D for each composition were determined by fitting $\theta_{F,sat}(T)$ measurements to Eq. (3). The resultant best-fit values, listed in Table I, were then used to generate theoretical $\theta_{F,sat}(T)$ curves that are compared with the original measurements in Fig. 3. The generally good agreement between the best-fit theoretical curves and the measurements indicates that, for these materials, inclusion of higher-order magneto-optical coefficients

TABLE I. Best-fit Ga:YIG stoichiometric values ($x+y=z$), molecular field coefficients (N_{aa}, N_{dd}, and $N_{ad}=N_{da}$), and magneto-optical coefficients (A and D) at $\lambda=1.3$ μm.

Sample	x	y	z	N_{aa}	N_{dd}	N_{ad}	A	D
YIG	0	0	0	−65.0	−30.4	97.0	55.2	−31.0
Ga:YIG No. 1	0	0.25	0.25	−58.1	−30.4	94.0	14.0	−2.1
Ga:YIG No. 2	0.02	0.46	0.48	−52.4	−30.1	91.1	15.4	−3.8
Ga:YIG No. 3	0.03	0.57	0.60	−49.4	−30.0	89.6	3.9	5.3
Ga:YIG No. 4	0.09	0.88	0.97	−41.0	−29.3	85.2	−784	720

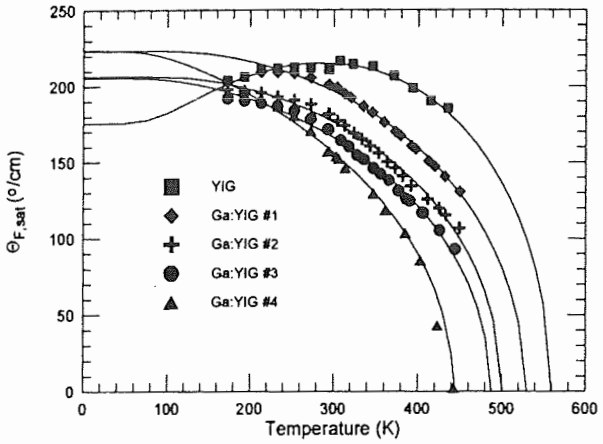

FIG. 3. Saturation Faraday rotation vs temperature for YIG and four Ga:YIG compositions at 1.3 μm. Solid lines correspond to the theoretical model based on best-fit molecular field coefficients and magneto-optical coefficients for each sample.

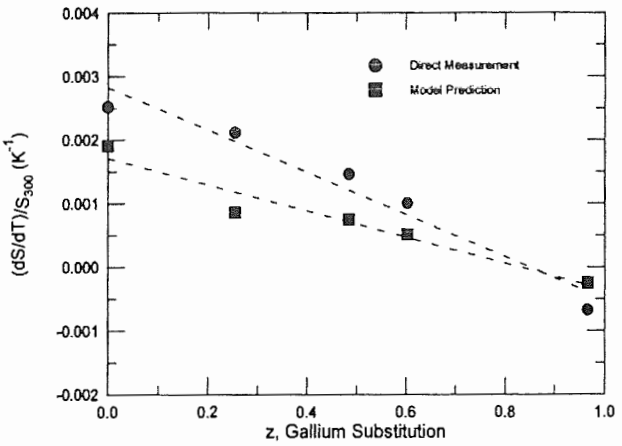

FIG. 5. Experimental and theoretical sensitivity temperature coefficients vs gallium substitution. Dashed lines indicate least-squares fits to each set of temperature coefficients.

than those that appear in Eq. (3) is unnecessary. Hansen and Witter arrived at the same conclusion when determining the magneto-optical coefficients of Ga:YIG compositions at 633 nm.[7] Also similar to that study is the divergent behavior of the magneto-optical coefficients for gallium substitution levels approaching the compensation composition ($z \approx 1.1$).

From the experimentally determined values of x, y, A, and D, $S(T)$ was calculated (assuming $N_D = 1$) for each composition according to Eqs. (1)–(3). Values of $S(T)$ normalized to the theoretical values of S at 300 K are plotted in Fig. 4. The curves indicate that the slope of $S(T)$ changes sign somewhere between gallium substitution levels of 0.60 and 0.97. This indicates that the sensitivity for some composition within this range should exhibit a vanishing first-order temperature dependence.

The normalized temperature coefficients of the sensitivity, $(dS/dT)/S(300\ \mathrm{K})$, were determined from the theoretical curves of $S(T)$ (shown in Fig. 4) by a least-squares fitting routine (over the temperature range between 170 and 450 K) and are plotted vs gallium substitution in Fig. 5. Figure 5 also shows experimentally determined normalized temperature coefficients for each composition. These values were

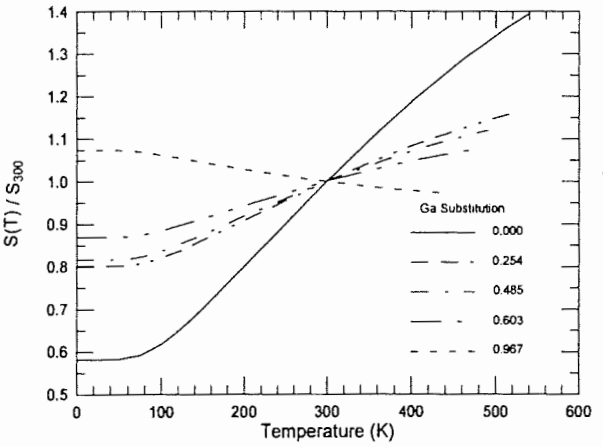

FIG. 4. Theoretical normalized sensitivity functions based on experimentally determined molecular field coefficients and magneto-optical coefficients.

determined from analyzing S values obtained directly from the experimental $\theta_F(H)$ curves. Specifically, the normalized temperature coefficients were computed from a least-squares fit performed on the experimental $S(T)$ data over approximately the same temperature range used for the theoretical least-squares fits.

Though the absolute values of the experimentally determined temperature coefficients are consistently smaller than the theoretical values, both types of coefficient exhibit the same trend with increasing gallium substitution. Moreover, a simple linear fit to either set of coefficients predicts that the magneto-optic sensitivity of a composition with a gallium substitution of approximately 0.8 should not exhibit a first-order temperature dependence. Such a composition would be approximately three times more sensitive than pure YIG.[4]

The temperature dependence of the magneto-optic sensitivity of iron garnets is governed by the garnet composition and can be tailored by appropriate levels of diamagnetic substitution. A model for the temperature dependence of the magneto-optic sensitivity of gallium-substituted yttrium iron garnets based on molecular field theory was developed and compared with direct sensitivity measurements at 1.3 μm. The model results agree qualitatively with the experimental results, indicating that the particular composition $Y_3Fe_{4.2}Ga_{0.8}O_{12}$ should be expected to exhibit a first-order temperature dependence approaching zero. Gallium substitution, thus, offers a technique for simultaneously minimizing temperature sensitivity and increasing magneto-optic sensitivity.

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