Available online at www.sciencedirect.com
ScienceDirect
Physica B 403 (2008) 1–4
PHYSICA B
www.elsevier.com/locate/physb

Nonlinear field dependence of the Faraday effect in neodymium gallium garnet under high magnetic field

Ming Gu[a,*], Wei Wang[b], Gongqiang Liu[a,c]

[a] Department of Physics, Shanghai Jiao Tong University, Shanghai 200240, PR China
[b] Department of Physics and Electronic, School of Science, Beijing University of Chemical Technology, Beijing 100029, PR China
[c] CCAST (World Laboratory), PO Box 8730, Beijing 100080, PR China

Received 11 December 2006; received in revised form 13 March 2007; accepted 14 March 2007

Abstract

In this paper, we present a theoretical investigation on the Faraday effect in paramagnetic neodymium gallium garnet ($Nd_3Ga_5O_{12}$) by taking account of the SO and CF interactions, the superexchange interaction and the external magnetic field. It is demonstrated that under high magnetic field, the Faraday rotation (θ) is strongly nonlinear with the external magnetic field (H_e) while the coefficients of H_e^i deeply dependent on the frequency of the incident light and temperature, and the Verdet constant $V(\theta/H_e)$ is also a function of H_e. Furthermore, theoretical calculations show that the reciprocity of the Faraday effect cannot be neglected under high magnetic field. The theoretical results are in good agreement with the experimental data.

PACS: 7820.L; 4660.D; 7520; 7530.E

Keywords: Nonlinear; Faraday effect; High magnetic field; Paramagnetic media

1. Introduction

The magneto-optical (MO) effects under high magnetic field have been paid much attention to over the last decade [1–3]. As it is known, current semi-classical and quantum MO theory indicates that θ is in proportion to $H_e(\theta = VH_eL)$ in paramagnetic media [4–6]. However, recently, Guillot et al. have reported the nonlinear property of the Faraday rotation in paramagnetic rare-earth gallium garnets under high magnetic field [7,8]. In Ref. [9], Guillot et al. have reported the nonlinear MO effect in paramagnetic NdF_3, meanwhile, utilizing the formula $\theta = VH_e + BH_e^2$, the experimental results were fitted. But they did not give a detailed theoretical explanation on this formula. To our knowledge, up to now, no theoretical studies have successfully interpreted these experimental phenomena in paramagnetic rare-earth gallium garnets.

In this paper, we present detailed quantum theoretical calculations on the field dependence of the Faraday rotation in paramagnetic media under high magnetic field range. A complicated relationship between θ and H_e is available when the external magnetic field is much higher. It has been proved that the MO effects originate mainly from intraionic electric dipole transition between the different levels, which are split by the spin-orbit (SO) and crystal field (CF) interactions, the superexchange interaction and the external magnetic field [10–14]. According to the above conclusions, a three-level model was established by Liu et al. [6]. In this paper, developing this model, the Faraday rotation in paramagnetic media under high magnetic field is analyzed by quantum theory. Finally, the curves of the Faraday rotation in paramagnetic $Nd_3Ga_5O_{12}$ versus the external magnetic field are fitted.

2. Theory

The MO property can be expressed in terms of the complex permittivity tensor ε [15]. With a neglect of the

*Corresponding author. Tel.: +862154743165; fax: +862154741040.
E-mail address: ftgm@sjtu.edu.cn (M. Gu).

0921-4526/$ - see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.physb.2007.03.017

local field acting on the electric dipole, the dielectric constant tensor elements can be written as

$$\varepsilon_{ij} = \varepsilon_0(\delta_{ij} + N\alpha_{ij}), \tag{1}$$

where α_{ij} is the ij component of the polarizability α, δ_{ij} is the Dirac function, and N is the atomic or ionic number in per unit volume.

Suppose the ground and the excited levels are labeled as a and b, respectively, ω is the frequency of the incident light, $\hbar\omega_{ab}$ is the energy-level separation between a and b, and Γ_{ab} is the line width. The components of the polarizability were given by Condon and Shortley as [16]

$$\alpha_{ij} = \sum_a \frac{\beta_a}{\hbar}\sum_b\left[\frac{P^i_{ab}P^j_{ba}}{\omega_{ab}+\omega-i\Gamma_{ab}} + \frac{P^j_{ab}P^i_{ba}}{\omega_{ab}-\omega+i\Gamma_{ab}}\right], \tag{2}$$

where $i,j = x,y,z$, β_a is the probability of an electron lying in the energy level α, and $\beta_{\max} = 1, P^x_{ab} = \langle\psi_a|ex|\psi_b\rangle$, is the element of the electric dipole matrix:

$$\varepsilon_{xy} = \frac{\omega_P^2}{2\mathrm{i}}\sum_{ab}\frac{\beta_a(\omega - i\Gamma_{ab})(f^+_{ab} - f^-_{ab})}{\omega_{ab}(\omega^2_{ab} - \omega^2 + \Gamma^2_{ab} + 2\mathrm{i}\omega\Gamma_{ab})}, \tag{3}$$

where $\omega_p^2 = \varepsilon_0 Ne^2/m$, and

$$f^{\pm}_{ab} = \frac{m\omega_{ab}}{\hbar e^2}\left|P^x_{ab} \pm iP^y_{ab}\right|^2$$

are the transition probabilities of electrons excited by right- or left-handed circularly polarized light from the ground level a to the excited level b, i.e., oscillator strengths.

For crystal systems where the symmetry is higher than that of the orthorhombic crystal system, inserting the quantum-mechanical expressions into the dielectric constant, the specific Faraday rotation θ caused by the electric dipole transitions can be given by

$$\theta = \frac{\omega_p^2\omega^2}{4\bar{n}c}\sum_{a,b}\frac{\beta_a}{\omega_{ab}}\cdot\frac{(\omega^2_{ab} - \omega^2 - \Gamma^2_{ab})(f^+_{ab} - f^-_{ab})}{(\omega^2_{ab} - \omega^2 + \Gamma^2_{ab})^2 + 4\omega^2\Gamma^2_{ab}}, \tag{4}$$

where c is the velocity of light in vacuum, and $\bar{n}$ represents the average refractive index.

The magnetic susceptibility χ of most paramagnetic media follows a Curie–Weiss law, and most of them are transformed from ferromagnetic, antiferromagnetic or ferrimagnetic media when the temperatures are above their magnetic phase transition points, $T > T_c(T_N)$. As it is known, the superexchange interaction between the nearest neighboring ions exists in paramagnetic rare-earth garnets [17]. That is, the SO–CF split energy levels will generally be split again by the superexchange interaction and the external magnetic field. In Fig. 1, the lowest SO–CF ground level labeled as a will be split into two levels (a_1, a_2) by combined actions of the effective exchange field H_v and the external magnetic field H_e [18]

$$\Delta E = 2\hbar\Delta = mg\mu_B\mu_0 H_i, \tag{5}$$

where m is the magnetic quantum number, g the Landé factor, μ_B the Bohr magnetron, and the effective field H_i is equal to $(H_i + vM)$. Here, v is the coefficient related to the molecular field coefficient, but not the molecular field coefficient itself.

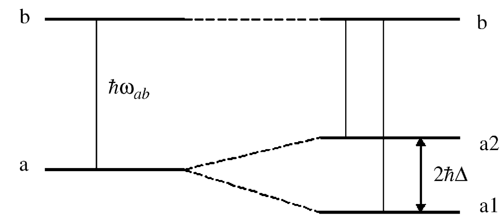

Fig. 1. The double electronic transition with ground state splitting.

The electrons occupy both of the split ground levels and follow a Boltzmann distribution.

$$\beta_{a1} = \frac{e^{-\beta E_1}}{\sum_l e^{-\beta E_l}},$$
$$\beta_{a2} = \frac{e^{-\beta E_2}}{\sum_l e^{-\beta E_l}} = \beta_{a1}e^{-2\beta\hbar\Delta}, \tag{6}$$

where $E_2 = E_1 + 2\hbar\Delta$, $\beta = 1/k_BT$,and k_B is the Boltzmann constant. Suppose that

$$\Gamma_{11} = \Gamma_{21} = \Gamma, \quad \begin{matrix} f^+_{11} = 0 & f^+_{21} = f^+, \\ f^-_{11} = f^- & f^-_{21} = 0. \end{matrix} \tag{7}$$

According to Eq. (4), the Faraday rotation is obtained:

$$\theta = \frac{\omega_p^2\omega^2 L}{4nc}\left[-\frac{\beta_{a1}}{\omega_{11}}\frac{(\omega^2_{11} - \omega^2)f^-}{(\omega^2_{11} - \omega^2)^2 + 4\omega^2\Gamma^2} + \frac{\beta_{a2}}{\omega_{21}}\frac{(\omega^2_{21} - \omega^2)f^+}{(\omega^2_{21} - \omega^2)^2 + 4\omega^2\Gamma^2}\right]. \tag{8}$$

Define Δ as the frequency shift caused by the ground level splitting, thus

$$\omega_{11} = \omega_0 + \Delta l, \quad \omega_{21} = \omega_0 - \Delta, \tag{9}$$

where $\hbar\omega_0$ is the energy-level separation between the ground state and the excited state split by the SO–CF interactions. Inserting Eq. (9) into Eq. (8), taking into account $\Delta \ll \Gamma \ll \omega_0$, and omitting the term of Δ^2 in the denominator, thus

$$\theta = \frac{\omega_p^2\omega^2 L}{4n\omega_0^2 c[(\omega_0^2 - \omega^2)^2 + 4\omega^2\Gamma^2]} \times \left\{\begin{matrix} -\beta_{a1}(\omega_0^2 + 2\omega_0\Delta - \omega^2)(\omega_0 - \Delta)f^- \\ +\beta_{a2}(\omega_0^2 - 2\omega_0\Delta - \omega^2)(\omega_0 + \Delta)f^+ \end{matrix}\right\}. \tag{10}$$

Here we expand β_{a2} as

$$\beta_{a2} = \beta_{a1}\left[1 - 2\beta\hbar\Delta + \frac{1}{2!}(2\beta\hbar\Delta)^2 - \frac{1}{3!}(2\beta\hbar\Delta)^3 + \frac{1}{4!}(2\beta\hbar\Delta)^4 + \cdots\right]. \tag{11}$$