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# On the origin of the magneto-optical effects in Li, Mg, Ni, and Co ferrite

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A reexamination of the magneto-optical (MO) spectra of  $\text{Li}_{0.5}\text{Fe}_{2.5}\text{O}_4$ ,  $\text{MgFe}_2\text{O}_4$ ,  $\text{NiFe}_2\text{O}_4$ , and  $\text{CoFe}_2\text{O}_4$  is presented. Thus far the MO spectra of these compounds have been explained by either orbital promotion processes, oxygen to iron charge transfer transitions or crystal field transitions and, in the case of  $\text{CoFe}_2\text{O}_4$ , a single intervalence charge transfer (IVCT) transition. For  $\text{Li}_{0.5}\text{Fe}_{2.5}\text{O}_4$  and  $\text{MgFe}_2\text{O}_4$ , the major transitions in the MO–Kerr spectra between 0.5 and 5.0 eV are assigned to intersublattice charge transfer (ISCT) transitions. The MO–Kerr spectrum of  $\text{NiFe}_2\text{O}_4$  is more complex as in this case both  $\text{Fe}^{3+}$  and  $\text{Ni}^{2+}$  contribute to the spectrum. In  $\text{NiFe}_2\text{O}_4$  an additional IVCT transition is observed at 2.9 eV. The complex MO spectrum of  $\text{CoFe}_2\text{O}_4$  could be resolved using the dielectric tensors of  $\text{Co}^{2+}\text{Al}_x^{3+}\text{Fe}_{2-x}^{3+}\text{O}_4$  ( $x=0, 0.1, 0.6, \text{ and } 1$ ) and rigorously fitting both the diagonal and the off-diagonal elements of the dielectric tensor with one set of transitions which was consistent for all degrees of  $\text{Al}^{3+}$  substitution. Thus, in these ferrites a consistent assignment of all the major transitions in the 2.0 to 5.0 eV energy range to either IVCT or ISCT transitions is made. © 1998 American Institute of Physics. [S0021-8979(98)22011-1]

## I. INTRODUCTION

The identification and assignment of magneto-optical (MO) active transitions in spinel ferrites have been a subject of debate for many years. The identification of transitions is complex as in these compounds many bands overlap. In addition, the electronic structure of transition metal oxides such as ferrites is complex<sup>1,2</sup> which has resulted in a variety of interpretations of the observed transitions. Recently, we have unraveled the MO–Kerr spectrum of  $\text{Fe}_3\text{O}_4$  between 0.7 and 4.0 eV by rigorous fitting of all four elements of the dielectric tensor of  $\text{Fe}_3\text{O}_4$  simultaneously, using the equations of the elements of the dielectric tensor which describe the basic line shapes of transitions in the microscopic theory (Lorentz-type lineshape). In addition, the complete dielectric tensors of  $\text{Fe}_3\text{O}_4$  and of partially  $\text{Mg}^{2+}$  or  $\text{Al}^{3+}$  substituted  $\text{Fe}_3\text{O}_4$  were fitted with only one set of parameters for all samples. The influence on the intensity of transitions of the gradual substitution of  $\text{Fe}^{2+}$  and/or  $\text{Fe}^{3+}$  provided evidence that the major transitions in  $\text{Fe}_3\text{O}_4$  can be assigned to intervalence charge transfer and intersublattice charge transfer transitions (IVCT and ISCT, respectively).<sup>3</sup> IVCT transitions are transitions in which an electron, through optical excitation, is transferred to a neighboring cation. IVCT transitions between  $\text{Fe}^{3+}$  ions on different crystallographic sites are traditionally called ISCT transitions.<sup>4</sup> For reasons of clarity we will continue to refer to these transitions as ISCT transitions. (In our discussions parenthesis denote tetrahedral A sites and square brackets octahedral B sites.)

Thus far multiple interpretations have been reported for the ferrites  $\text{Li}_{0.5}\text{Fe}_{2.5}\text{O}_4$ ,  $\text{MgFe}_2\text{O}_4$ ,  $\text{NiFe}_2\text{O}_4$ , and  $\text{CoFe}_2\text{O}_4$ . We will discuss them in chronological order. Krinchik *et al.* reported the off-diagonal element of the dielectric tensor of  $\text{Li}_{0.5}\text{Fe}_{2.5}\text{O}_4$  between 2 and 5 eV.<sup>5</sup> These authors reported

also the off-diagonal elements of  $\text{NiFe}_2\text{O}_4$ ,  $\text{CoFe}_2\text{O}_4$ ,  $\text{CoCrFeO}_4$  between 1 and 5.4 eV and the equatorial Kerr effect of  $(\text{Fe})[\text{NiCr}]\text{O}_4$ .<sup>5,6</sup> In addition to the transitions already identified in  $\text{Li}_{0.5}\text{Fe}_{2.5}\text{O}_4$  and  $\text{MgFe}_2\text{O}_4$  an additional broad band near 3 eV was identified in  $\text{NiFe}_2\text{O}_4$ . This band was assigned to the CF transition  ${}^3A_{2g}(\text{F}) \rightarrow {}^3T_{1g}(\text{P})$  of octahedrally coordinated  $\text{Ni}^{2+}$ . However, in this interpretation the disappearance of this transition in  $(\text{Fe})[\text{NiCr}]\text{O}_4$  cannot be accounted for. From the observed influence of  $\text{Al}^{3+}$  and  $\text{Cr}^{3+}$  substitution on the equatorial Kerr spectra of  $\text{NiFe}_2\text{O}_4$  and  $\text{CoFe}_2\text{O}_4$ , Krinchik *et al.* conclude that the major transitions in  $\text{Li}_{0.5}\text{Fe}_{2.5}\text{O}_4$  and  $\text{MgFe}_2\text{O}_4$  are ISCT transitions.

Subsequently, Višnovsky *et al.* reported the diagonal and off-diagonal elements of the dielectric tensor of  $\text{MgFe}_2\text{O}_4$  and reflectivity and polar Kerr spectra of  $\text{Li}_{0.5}\text{Fe}_{2.5}\text{O}_4$  between 2 and 5.5 eV. These authors assigned the transitions identified to oxygen ( $2p$ ) to iron ( $3d$ ) charge transfer transitions.<sup>7</sup> From the work of Bocquet *et al.*, however, it follows that this is an unlikely interpretation as the oxygen to iron charge transfer transitions are centered around 6 eV (Ref. 2) in agreement with previous reports.<sup>4,8</sup> In 1983 Zhang *et al.* reported the diagonal (0.5–12 eV) and off-diagonal (0.5–5 eV) elements of the dielectric tensor of  $\text{Li}_{0.5}\text{Fe}_{2.5}\text{O}_4$  and  $\text{MgFe}_2\text{O}_4$ . The identified bands were assigned to  $3d^n \rightarrow 3d^{n-1}4s$  orbital promotion transitions.<sup>8</sup> However, these transitions are parity and partly spin forbidden and can, therefore, not account for the relatively large oscillator strength ( $10^{-3}$ ) of the transitions observed in these ferrites. Finally, the MO–Kerr spectrum of  $\text{CoFe}_2\text{O}_4$  has been extensively studied by Martens and co-workers.<sup>9,10</sup> Two narrow diamagnetic transitions at 0.8 and 2.0 eV were identified and assigned to CF transitions [ ${}^4A_2 \rightarrow {}^4T_1(\text{F})$  and  ${}^4A_2 \rightarrow {}^4T_1(\text{P})$ ] of tetrahedrally coordinated  $\text{Co}^{2+}$ . A broad paramagnetic band also at 2.0 eV was assigned to a  $[\text{Co}^{2+}] \rightarrow [\text{Fe}^{3+}]$  IVCT transition. The broad diamagnetic band in the region above 3 eV remained unresolved.

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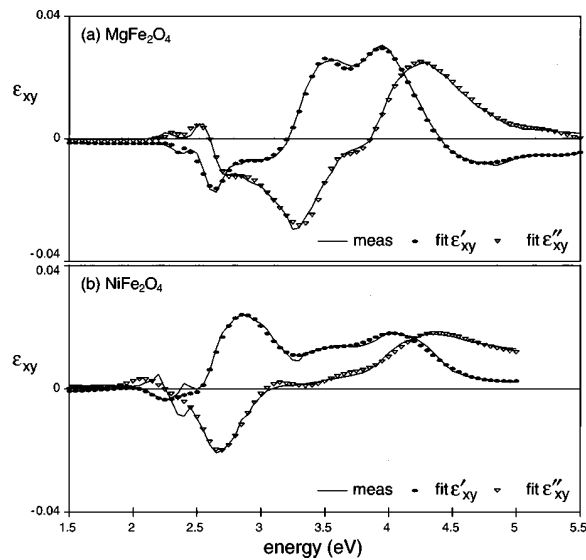


FIG. 1. (a) The measurement Ref. 7 and the fits of the real and imaginary part of the off-diagonal element of the dielectric tensor,  $\epsilon'_{xy}$  and  $\epsilon''_{xy}$ , of  $\text{MgFe}_2\text{O}_4$ . (b) The measurement Ref. 5 and the fits of the real and imaginary part of the off-diagonal element of the dielectric tensor,  $\epsilon'_{xy}$  and  $\epsilon''_{xy}$ , of  $\text{NiFe}_2\text{O}_4$ .

Here we report a study of the MO spectra of  $\text{Li}_{0.5}\text{Fe}_{2.5}\text{O}_4$ ,  $\text{MgFe}_2\text{O}_4$ ,  $\text{NiFe}_2\text{O}_4$ , and  $\text{CoFe}_2\text{O}_4$  and identify the major transitions. For this purpose, we have taken the elements of the dielectric tensor of these compounds from literature and applied the same method used to resolve the MO spectrum of  $\text{Fe}_3\text{O}_4$ .<sup>3</sup>

## II. RESULTS AND DISCUSSION

First, we examine  $\text{MgFe}_2\text{O}_4$ . We applied the method used to resolve the spectrum of  $\text{Fe}_3\text{O}_4$  to the diagonal and off-diagonal element of the dielectric tensor of  $\text{MgFe}_2\text{O}_4$  obtained from Refs. 8 and 7, respectively. The off-diagonal element and the fits are presented in Fig. 1(a). The set of transitions identified in  $\text{MgFe}_2\text{O}_4$  is listed in Table I. To

assign these transitions, we compare them to the transitions found in  $\text{Fe}_3\text{O}_4$ .<sup>3</sup> The MO spectrum of  $\text{Fe}_3\text{O}_4$  consists of two sets of transitions: IVCT transitions involving  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  and the ISCT involving two  $\text{Fe}^{3+}$  ions located on different crystallographic sites. Since the  $\text{Fe}^{2+}$  ions present in  $\text{Fe}_3\text{O}_4$  are replaced by the nonmagnetic  $\text{Mg}^{2+}$ , the ISCT transitions should be the only remaining major transitions. Substitution of one ion with another may distort the lattice resulting in a shift in energy of the ISCT transitions. However, replacing  $\text{Fe}^{2+}$  with  $\text{Mg}^{2+}$  only has a small effect on the lattice since the lattice parameter of  $\text{MgFe}_2\text{O}_4$  matches closely the lattice parameter of  $\text{Fe}_3\text{O}_4$ . We conclude from our fit [Fig. 1(a)] that the MO-Kerr spectra of  $\text{MgFe}_2\text{O}_4$  can be explained in a manner consistent with our interpretation of  $\text{Fe}_3\text{O}_4$ , based on ISCT transitions, as suggested previously.<sup>11</sup>

For  $\text{Li}_{0.5}\text{Fe}_{2.5}\text{O}_4$  the elements of the dielectric tensor were obtained from Refs. 8 and 5. The set of transitions identified from our fits is listed in Table I. The same three major bands as in  $\text{MgFe}_2\text{O}_4$  could be identified. The fact that the intensities in  $\text{Li}_{0.5}\text{Fe}_{2.5}\text{O}_4$  are substantially higher than in  $\text{MgFe}_2\text{O}_4$  is to be expected as this compound contains a larger amount of  $[\text{Fe}^{3+}]$ , which is one of the participating ions in the transition.<sup>5</sup> Thus also the MO-Kerr spectra of  $\text{Li}_{0.5}\text{Fe}_{2.5}\text{O}_4$  can be explained in a manner consistent with  $\text{Fe}_3\text{O}_4$  based on ISCT transitions.

Only the off-diagonal element of the dielectric tensor of  $\text{NiFe}_2\text{O}_4$  was available.<sup>5</sup> The off-diagonal element and fit are presented in Fig. 1(b). The results of the fit are given in Table I. Again we observe the same set of ISCT transitions as in the previous samples. In addition, we observe an additional strong, relatively broad transition at 2.9 eV. This transition cannot be assigned to a CF transition. First of all, CF transitions of ions on octahedral sites are parity forbidden for any ion. Second, transitions of  $\text{Ni}^{2+}$  on tetrahedral sites cannot occur, as the  $\text{NiFe}_2\text{O}_4$  sample used, is reported to be completely inverse, as expected for  $\text{NiFe}_2\text{O}_4$ .<sup>12</sup> Third, CF transitions of  $\text{Fe}^{3+}$  on tetrahedral sites do not occur, as the term of a free  $\text{Fe}^{3+}$  ion is nondegenerate ( $L=0$ ). Further-

TABLE I. The main MO-active transitions in  $\text{MgFe}_2\text{O}_4$ ,  $\text{Li}_{0.5}\text{Fe}_{2.5}\text{O}_4$ ,  $\text{NiFe}_2\text{O}_4$ , and  $\text{CoFe}_2\text{O}_4$ , between 0.5 and 5.0 eV. Listed are transition energy,  $\omega$  (eV), linewidth,  $\Gamma$  (eV), intensity,  $(\epsilon_{xy})_{\text{max}}$ , and assignment. Typical error bars for  $\omega$ ,  $\Gamma$ , and  $(\epsilon_{xy})_{\text{max}}$  are  $\pm 0.02$  eV;  $\pm 0.03$  eV, and  $\pm 0.002$ , respectively. (Note that the parenthesis denote tetrahedral coordination and square brackets octahedral coordination.)

Sample		CF ( $\text{Co}^{2+}$ ) ${}^4A_2 \rightarrow {}^4T_1(\text{F})$	CF ( $\text{Co}^{2+}$ ) ${}^4A_2 \rightarrow {}^4T_1(\text{P})$	IVCT $[\text{M}^{2+}]$ $\rightarrow [\text{Fe}^{2+}]t_{2g}$	ISCT ( $\text{Fe}^{3+}$ ) $t_2$ $\rightarrow [\text{Fe}^{2+}]t_{2g}$	ISCT $[\text{Fe}^{3+}]e_g$ $\rightarrow (\text{Fe}^{2+})t_2$	ISCT ( $\text{Fe}^{3+}$ ) $t_2$ $\rightarrow [\text{Fe}^{2+}]e_g$
$\text{MgFe}_2\text{O}_4$	$\omega$				2.64	3.47	3.95
	$\Gamma$				0.30	0.31	0.56
	$(\epsilon_{xy})_{\text{max}}$				-0.009	0.022	0.032
	shape				dia	dia	dia
$\text{Li}_{0.5}\text{Fe}_{2.5}\text{O}_4$	$\omega$				2.63	3.46	4.09
	$\Gamma$				0.23	0.39	0.64
	$(\epsilon_{xy})_{\text{max}}$				-0.026	0.036	0.050
	shape				dia	dia	dia
$\text{NiFe}_2\text{O}_4$	$\omega$			2.86	2.57	3.46	4.03
	$\Gamma$			0.53	0.22	0.54	0.67
	$(\epsilon_{xy})_{\text{max}}$			0.023	-0.007	0.011	0.017
	shape			dia	dia	dia	dia
$\text{CoFe}_2\text{O}_4$	$\omega$	0.83	1.82	2.21	$\pm 2.60$	3.55	4.00
	$\Gamma$	0.10	0.18	0.25		0.29	0.50
	$(\epsilon_{xy})_{\text{max}}$	-0.056	-0.076	-0.054		0.013	0.030
	shape	dia	para	para	dia	dia	dia

more, this additional transition cannot be due to an oxygen  $2p$  to metal  $3d$  charge transfer transition. Such a transition should occur at an energy of 6 eV (Ref. 1) and it would be both broader and more intense than the observed band. Therefore, we assign this transition to an IVCT transition between  $[\text{Ni}^{2+}]$  and  $[\text{Fe}^{3+}]$ .<sup>13</sup> This is consistent with the strength and width of similar transitions in  $\text{Fe}_3\text{O}_4$ .<sup>3</sup> Furthermore, the energy of an IVCT transition between two different cations is expected to be substantially higher than that between two identical cations which in  $\text{Fe}_3\text{O}_4$  occurs at 0.55 eV.<sup>3</sup>

Evidence that the transition at 2.9 eV is indeed due to an IVCT transition between  $[\text{Ni}^{2+}]$  and  $[\text{Fe}^{3+}]$  comes from the equatorial Kerr spectrum of  $(\text{Fe})[\text{NiCr}]\text{O}_4$  between 1 and 5.4 eV.<sup>6</sup> No transitions whatsoever are found in this spectrum, ruling out that the transition at 2.9 eV in  $\text{NiFe}_2\text{O}_4$  is due to a CF transition of  $\text{Ni}^{2+}$ . The absence of this transition is, however, in line with an IVCT involving  $[\text{Fe}^{3+}]$ . This also supports our assignment of the remaining transitions in  $\text{NiFe}_2\text{O}_4$  to ISCT transitions. Another indication that all transitions in  $\text{NiFe}_2\text{O}_4$  are similar transitions involving  $[\text{Fe}^{3+}]$  is the influence of  $\text{Al}^{3+}$  substitution on the MO spectrum of  $\text{NiAl}_x\text{Fe}_{2-x}\text{O}_4$  ( $0 \leq x \leq 1.2$ ) which remains completely inverse up to  $x = 1$ .<sup>14</sup> The entire spectrum remains unchanged up to  $x = 0.3$ , then the intensity of the entire spectrum decreases homogeneously.

The complete dielectric tensors of  $\text{CoFe}_2\text{O}_4$  and  $\text{CoAl}_x\text{Fe}_{2-x}\text{O}_4$  ( $x = 0.1, 0.6, 1$ ) were taken from Martens *et al.*<sup>9,10</sup> All four elements of the dielectric tensors of  $\text{CoFe}_2\text{O}_4$  and  $\text{CoAl}_x\text{Fe}_{2-x}\text{O}_4$  were fitted simultaneously with only one set of parameters for all spectra, thus allowing trends in intensity upon  $\text{Al}^{3+}$  substitution to be observed. The results of the fits for  $\text{CoFe}_2\text{O}_4$  are shown in Table I. Our assignment of the first three transitions is identical to the assignment made previously.<sup>9</sup> The first two are assigned to the CF transitions  ${}^4A_2 \rightarrow {}^4T_1(\text{F})$  and  ${}^4A_2 \rightarrow {}^4T_1(\text{P})$  ( $\text{Co}^{2+}$ ) on A sites. These are the only two CF transitions which are allowed from the singlet  ${}^4A_2$  ground state of ( $\text{Co}^{2+}$ ) (note that CF transitions from octahedrally coordinated  $[\text{Co}^{2+}]$  are parity forbidden). The third is assigned to a  $[\text{Co}^{2+}] \rightarrow [\text{Fe}^{3+}]$  IVCT transition. The remaining two transitions are two of the three ISCT transitions observed in each of the three previous investigated compounds. The third ISCT near 2.6 eV, present in all other spinel ferrites discussed so far, is obscured in  $\text{CoFe}_2\text{O}_4$  due to the intense band at 2.2 eV. To the evidence presented for the assignment of the CF transitions proposed by Martens *et al.*,<sup>9</sup> one can add the observed changes in intensity as reported for  $\text{CoAl}_x\text{Fe}_{2-x}\text{O}_4$ .<sup>14</sup>  $\text{CoAl}_x\text{Fe}_{2-x}\text{O}_4$  changes from inverse to normal spinel between  $x = 0$  to  $x = 1.2$ , i.e.,  $\text{Co}^{2+}$  migrates to the tetrahedral site. The intensity of the transition at 1.9 eV increases for  $x = 0.0$  to 0.8 then decreases for  $x > 0.8$ . The intensity of the transition at 2.2 eV slightly increases for  $x = 0.3$  then decreases. This enhancement effect on IVCT transitions of a small amount of  $\text{Al}^{3+}$ , resulting from perturbation of the octahedral symmetry, was observed also in  $\text{Fe}_3\text{O}_4$  containing a small amount of  $\text{Al}^{3+}$ .<sup>3</sup> The bands between 3 and 5 eV

decrease steadily for  $0 \leq x \leq 1.2$ . The dependencies of the intensities of the IVCT transition (2.2 eV) and the ISCT transitions on the degree of  $\text{Al}^{3+}$  substitution tend, as expected, towards zero for  $\text{CoAlFeO}_4$  in our fits. As discussed by Martens *et al.* the gradual decrease in intensity for the CF transitions of ( $\text{Co}^{2+}$ ), however, is not straightforward. Their claim is that the sharp drop in Curie temperature for the series causes this. The fact that the energy of the IVCT transition is considerably larger than that of the first IVCT in  $\text{Fe}_3\text{O}_4$  can be attributed to the fact that the IVCT is in this case between cations with different nuclei. The fact that the energy of this IVCT is smaller compared to the energy of the observed IVCT in  $\text{NiFe}_2\text{O}_4$  is also not unexpected, if we take into account the relative energy position of the  $3d$  bands.<sup>1</sup>

### III. CONCLUSIONS

The common feature of all spinel ferrites of the general type  $\text{M}_x\text{Fe}_{3-x}\text{O}_4$  with  $\text{Fe}^{3+}$  ions on both tetrahedral and octahedral sites is a set of ISCT transitions at 2.6, 3.5, and 4.0 eV. This is evident from Table I for those spinel ferrites, where M is the nonmagnetic  $\text{Mg}^{2+}$  and  $\text{Li}^+$  as well as for those where M is the magnetic  $\text{Ni}^{2+}$  and  $\text{Co}^{2+}$ . (This even applies to  $\gamma\text{-Fe}_2\text{O}_3$ , where M is in fact a vacancy.<sup>14</sup>) Whether these transitions dominate the spectrum depends on the presence of other contributing transitions. For  $\text{M} = \text{Mg}$ ,  $\text{Li}$ , or other nonmagnetic ion (or vacancies), the set of ISCT transitions is the main feature. In  $\text{Fe}_3\text{O}_4$  several IVCT transitions between  $[\text{Fe}^{2+}]$  and  $\text{Fe}^{3+}$  are main contributors.<sup>3</sup> In  $\text{NiFe}_2\text{O}_4$  an IVCT between  $[\text{Ni}^{2+}]$  and  $[\text{Fe}^{3+}]$  at 2.9 eV is a main contributor. In  $\text{CoFe}_2\text{O}_4$  CF transitions of tetrahedrally coordinated  $\text{Co}^{2+}$  and an IVCT transition between  $\text{Co}^{2+}$  and  $\text{Fe}^{3+}$  contribute significantly below 2.5 eV. These assignments are consistent with our interpretation of the MO spectra of  $\text{Fe}_3\text{O}_4$ .

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