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REPLY

Reply to the comment by L Helseth on 'Bismuth-induced increase of the magneto-optical effects in iron garnets'

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Abstract

The discussion concerns the origin of the giant Faraday rotation in bismuth- and lead-doped iron garnets. It is convincingly shown that this effect is due to the covalent admixture of Bi(Pb)6p-wavefunctions to oxygen 2p-orbitals in octahedral and tetrahedral Fe–O clusters of iron garnets. The crucial role of the quantum-chemical computation of electronic structure of such clusters is emphasized.

We show in the following that Helseth's claims are ungrounded.

(1) First, the article [1]—a basis for the analysis [2], reputed there to be 'a self-consistent molecular-orbital analysis'—is a rather trivial quantum mechanics exercise with a two-level system. Its main defect is its disregard for the fact that the garnet magneto-optics in the visible and UV range is mainly caused not by separate Fe^{3+} ions, but by octahedral $(FeO_6)^{9-}$ and tetrahedral $(FeO_4)^{5-}$ complexes. Starting from Bi–Fe molecular orbital *one cannot* explain the lead-induced increase of the Faraday effect in iron garnets which is analogous to the bismuth case—contrary to the assertion in the end of section 6 in [1]. Thus, both Helseth's papers [2, 3] stand on shaky ground. The question about the microscopic nature of transitions 'introduced' in [2] to obtain 'excellent agreement' with experiment is not even raised in [2].

(2) Back to Helseth's claims. Concerning his first claim: the bismuth distribution over the lattice is a matter of crystal growth and sample preparation and not of magneto-optical theory.

(3) The 'paramagnetic' and 'diamagnetic' line shape is rather an old empirical classification. Really, taking into account the perturbation theory corrections both to wavefunctions and energy levels results in *combined* contributions to the permittivity components having a mixture of the 'paramagnetic' and 'diamagnetic' spectral dependences [4].

(4) Variation of transition energies, amplitudes and line widths, taking into account the 'paramagnetic' and 'diamagnetic' components of each spectral peculiarity allow *any* magneto-optical spectrum to be modelled. The spectral range in which Helseth has measured the Faraday rotation is too narrow to allow a reliable fitting. Again, processing experimental curves without clearing up (or at least discussing) the *origin* of transitions whose energies are obtained seems to us a mere least-squares exercise. Moreover, it is based on an incorrect crucial formula (7) where, to mention but one error, the Lorentz–Lorenz local field correction factor is not taken into account.
We insist on the essentiality of modern quantum-chemistry computations of energy spectra and electronic structure of $(FeO_6)^{9-}$-type clusters [5]; otherwise, any fitting of magneto-optical spectra seems senseless.

(5) An explanation of the linear dependence of the Faraday rotation on the bismuth content has already been given in our article [6]. The cause of 'deviation between theory and experiment below 2.3 eV' can be found on page 6963 (footnote) [6].

(6) We processed (and noted in [6]) the spectra of $Y_{3-x}Bi_xFe_5O_{12}$ [7] at $x = 0.25$, 0.8, 1.0; thus, Helseth's assertion that we confined our analysis to a small bismuth content is groundless.

(7) The self-citation [3] is inappropriate, since this paper appeared when our article [6] was already published. The conceptual level of [3] is the same as that of [2], though. We find again 'excellent agreement' under 'introduction' of only two transitions.

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