ELECTRONIC AND MAGNETIC STRUCTURES OF FERRITES

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Ferrites ($ZnFe_2O_4$, $Li_{0.5}Fe_{2.5}O_4$, $Y_3Fe_5O_{12}$, etc.) must be included among the most interesting magnetic oxides of transition metals. In spite of enormous volume of experimental results on the magnetic and electrical properties of ferrites, their electronic structure have so far been described only qualitatively. For the most ferrites with 56 atoms (spinel) or 160 atoms (garnet) in the unit cell it is very difficult to calculate local densities of states (LDOS) by the traditional methods of band theory. Therefore we have proposed [1] the method for LDOS calculations in such complex crystal structures using the cluster recursion method in the tight-binding approximation. The method was earlier employed in studies of semiconducting heterojunctions and superlattices [2]. The results of LDOS calculations have been used for estimations of the equilibrium magnetic ordering, energies of "spin-flip" excitations and Heisenberg parameters for the mixed spinel $Li_{0.5}Fe_{2.5}O_4$ and the yttrium-iron garnet (YIG) $Y_3Fe_5O_{12}$ [3]. The LDOS can be expressed as

$$n_{\alpha\ell\sigma,\alpha\ell\sigma}(E) = -\pi^{-1} Im G_{\alpha\ell\sigma,\alpha\ell}(E) ,$$

where ℓ labels the sites, α is the type of the atomic functions and σ is the spin index. In the recursion method the Green function G(E) can be represented as a continued fraction with coefficients $\{a_n, b_n\}$ [4]. The preliminary stage of the actual recursion calculations was the choice of the Hamiltonian of the system and determination of its matrix elements in the basis of linear combinations of atomic orbitals (LCAO). In general $H = H_0 + H_1$, where H_0 is the tight-binding Hamiltonian and H_1 is the Hamiltonian of interelectron interactions. The diagonal elements of H_0 were chosen to be the Hartree-Fock (HF) values E_0 (of the atomic energy levels of system considered). The off-diagonal were expressed in terms of the usual Koster-Slater parameters and we have selected their numerical values according to [5]. The Hamiltonian H_1 leads in the HF approximation to a shift of the self-energies $E\alpha$ relative to those corresponding to the paramagnetic state and to the appearance of the Madelung field Vm. The final condition for the self-consistent determination of the position and splitting of the d-level has the form

$$E_{d\sigma} = E^0_{d\sigma} + V_m + V_{dd}(N_d - N^0_d) + V_{sd}(N_s - N^0_s) + \Delta_\sigma/2$$

$$\Delta_\sigma = (U - J)m_{d\nu\sigma} + J m_{d\bar{\sigma}}, m_{d\sigma} = \sum m_{d\nu\sigma}, N_{d\nu\sigma} = \int n_{d\nu\sigma}(E)dE, N_d = \sum N_{d\nu\sigma}$$

Here ν labels d-orbitals, U is the intraatomic Coulomb integral between orbitals of the same symmetry, U'-between d-orbitals of different symmetries and J-intraatomic exchange integral (see [1]). The Madelung potential was calculated in the model of point ions by the Evald method. All Coulomb integrals were screened by dielectric constant ϵ. The value Δ_σ in the proposed procedure changes in each iteration step until the required convergence of results is reached. We have carried out recursion calculations for a central and surface regions of a cluster consisting of 1500 - 3000 atoms with garnet and spinel crystal structure where magnetic iron atoms occupy two inequivalent positions of the octahedral and tetrahedral types. In the atomic bases we have included $2s$ and $3p$ oxygen orbitals, $3d$ and $4s$ iron orbitals, $4d$ and $5s$ yttrium orbitals. In the modeling of the magnetic structure we assumed

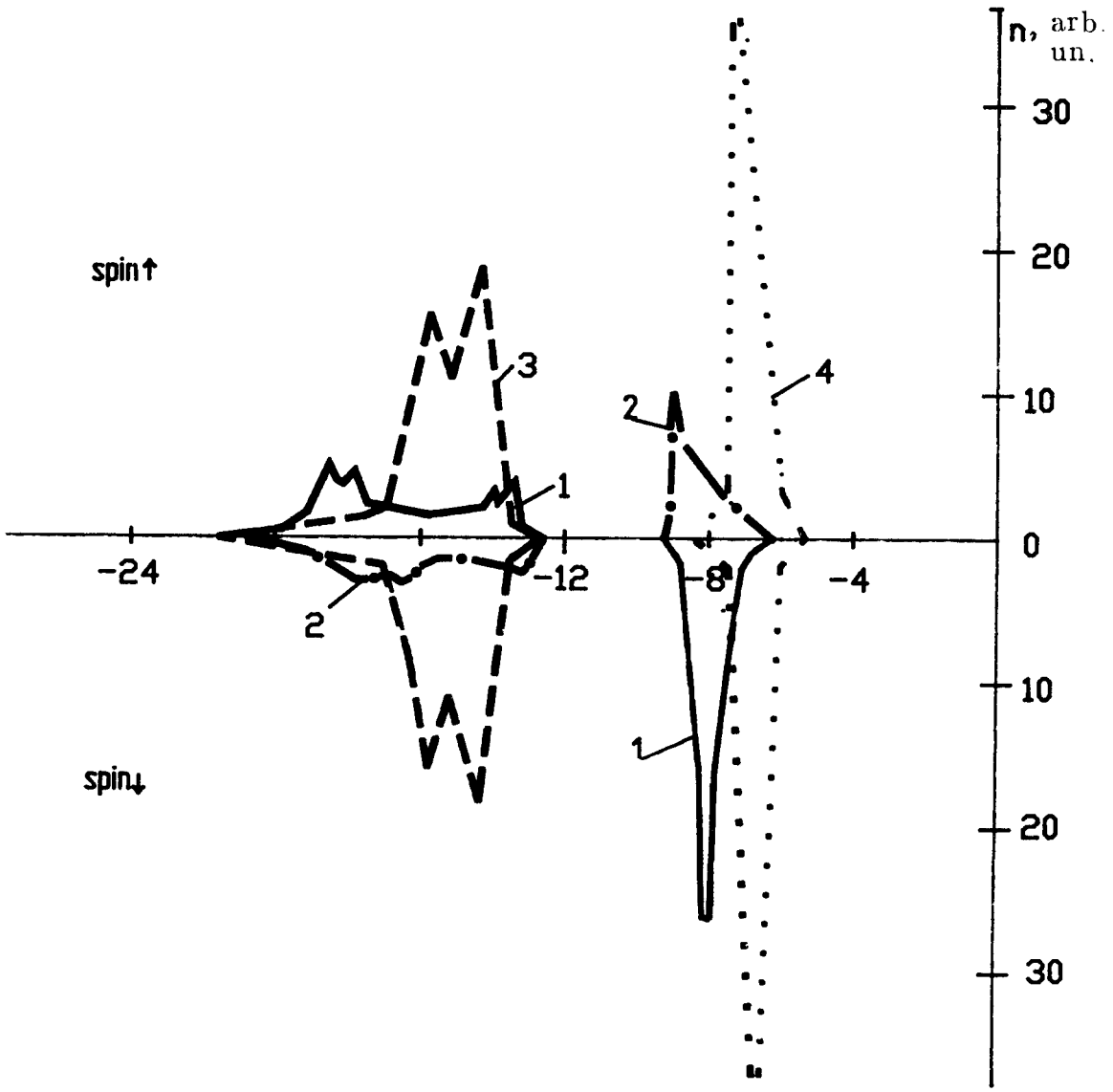

Fig. LDOS for the AF ordering between iron magnetic sublattices in $Y_3Fe_5O_{12}$: *1* – $n_d(E)$, iron (oct.); *2* – $n_d(E)$, iron (tetr.); *3* – $n_p(e)$, oxygen; *4* – $n_d(E)$, yttrium

that atomic magnetic moments m_i are collinear. The magnitudes of the m_i were obtained after a complete cycle of self-consistent calculations for antiferromagnetic (AF) and ferro magnetic (FM) mutual ordering of the magnetic sublattices in mixed spinel $Li_{0.5}Fe_{2.5}O_4$ and garnet $Y_3Fe_5O_{12}$ (YIG). It was found that the AF state corresponds to the minimum of a full energy. The difference ΔE_{sf} between energies of AF and FM configurations for both systems is negative. From the Heisenberg model for two sublattice ferrimagnet we can estimate the Heisenberg exchange parameter $J_{AB} = 4J_{AB}m_Am_Bk$ and Neel temperature $T_N = \Delta E_{sf}/6$, where $k = 7.2$ for the lithium ferrite and $k = 4.8$ for YIG. Fig. gives the LDOS for the YIG in AF configuration averaged over a single unit cell in the central region of a cluster. The YIG band gap $E_g = 3$ eV agrees well with experimental value 2.85 eV. The calculated values of mi, J_{AB} and T_N are listed in Table. For the YIG the agreement with experiment is rather good.

Table

	ΔE_{sf}, eV	J_{AB}	T_N, K	m, μ_B
$Li_{0.5}Fe_{2.5}O_4$	0.92	60	$1800(950^{exp})$	4.75(oct.)-4.39(tetr.)
$Y_3Fe_5O_{12}$	0.324	31	$600(545^{exp})$	4.43(oct.) - 4.5(tetr.)

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