Calculation of hyperfine field in WIEN2k.
Technical report

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I. INTRODUCTION

Calculation of the density (or 'population') matrix $\hat{n}$ was implemented to WIEN2k in connection with the LDA+U method. Corresponding program package is SRC_lapwdm. Later we realized that the density matrix may be simply used to calculate the expectation value of any 'on site' operator $\hat{X}$ by taking the trace $<X>=Tr(\hat{n}\hat{X}^+)$ (hermitean conjugate of $\hat{X}$ appears because of the definition of $\hat{n}$ used in WIEN2k - see section III A). This is especially useful for calculation of the on site contributions from the spin (B_{dip})and orbital (B_{orb}) moments to the hyperfine field. In 2006 the calculation of $<X>$ was reanalysed, reprogrammed and this report was written.

II. GENERAL SCHEME

Hyperfine field on a nuclei of an atom may be written as

$$B_{hf} = B_c + B_{dip} + B_{orb} + B_{lat}, \tag{1}$$

where B_c is the Fermi contact term, B_{dip} is the dipolar field from the on-site spin density, B_{orb} is the field associated with the on-site orbital moment. B_{lat} is classical dipolar field from all other atoms in the system that carry the magnetic moment and its calculation is described in detail in [1]. Calculation of B_c is performed in standard spin-polarized WIEN2k calculation. Below we describe the calculation of B_c, B_{dip} and B_{orb} as given by Blügel *et al.* [2] ($\mathcal{B}$).

The energy of the interaction of nuclear magnetic moment $\vec{\mu}_I$ with the orbital moment $\vec{l}$ of an electron localized on atom in question ($\vec{l}$ is in units of $\hbar$) is ($\mathcal{B}$, eq.24)

$$E_{orb} = -\frac{e}{mc}\vec{\mu}_I \langle\Phi|\frac{S(r)}{r^3}\vec{l}|\Phi\rangle \tag{2}$$

where Φ is the large component of the relativistic wave function, m is the electron mass, $S(r)$ is reciprocal of the relativistic mass enhancement ($\mathcal{B}$, eq.21):

$$S(r) = \left[1 + \frac{\epsilon - V(r)}{2mc^2}\right]^{-1}. \tag{3}$$

The energy of dipolar interaction of nuclear spin with the electron spin magnetic moment $\vec{\mu}$ is ($\mathcal{B}$, eq.26)

$$E_{dip} = \langle\Phi|\frac{S(r)}{r^3}[\vec{\mu}\,\vec{\mu}_I - 3(\vec{\mu}\,\vec{\hat{r}})(\vec{\mu}_I\,\vec{\hat{r}})]|\Phi\rangle; \quad \vec{\hat{r}} = \vec{r}/r. \tag{4}$$

We now use that $\mu_B = e\hbar/2mc$ and $\vec{\mu} = 2\mu_B\vec{s}$, $\vec{s}$ being the electron spin. Eqs. (5, 6) then reduce to:

$$E_{orb} = -2\vec{\mu}_I\,\mu_B\langle\Phi|\frac{S(r)}{r^3}\vec{l}|\Phi\rangle \tag{5}$$

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$$E_{dip} = 2\mu_B \, \vec{\mu}_I \langle\Phi| \frac{S(r)}{r^3} \left[\vec{s} - 3(\vec{s}\vec{r})\,\vec{\hat{r}} \right] |\Phi\rangle. \tag{6}$$

The contact interaction is ($\mathcal{B}$ 32)

$$E_c = -\frac{8\pi}{3}\mu_B \, \vec{\mu}_I \, \vec{m}_{av}; \; \vec{m}_{av} = \int dr' \delta_T(r')\vec{m}(r'), \tag{7}$$

where

$$\vec{m}(r') = \langle\Phi| \vec{\sigma}\, \delta(\vec{r} - \vec{r}') |\Phi\rangle \tag{8}$$

σ are the Pauli matrices,

$$\delta_T(r) = \frac{1}{4\pi r^2} \frac{r_T/2}{[(1 + \epsilon/2mc^2)r + r_T/2]} \tag{9}$$

$r_T = Ze^2/mc^2$ is the Thomas radius.

Energy of the nuclear magnetic moment in an external magnetic field $\vec{B}$ is

$$E_{ext} = -\vec{\mu}_I \vec{B}. \tag{10}$$

Comparing this equation with (5, 6, 7) we get the expressions for the hyperfine fields $\vec{B}_{orb}$, $\vec{B}_{dip}$ and $\vec{B}_c$:

$$\vec{B}_{orb} = 2\mu_B \langle\Phi| \frac{S(r)}{r^3}\vec{l} |\Phi\rangle \tag{11}$$

$$\vec{B}_{dip} = 2\mu_B \langle\Phi| \frac{S(r)}{r^3} \left[3(\vec{s}\vec{r})\,\vec{\hat{r}} - \vec{s} \right] |\Phi\rangle, \tag{12}$$

$$\vec{B}_c = \frac{8\pi}{3}\mu_B \, \vec{m}_{av} \tag{13}$$

In Blügel *et al.* formulation contact field is parallel to the spin density on nucleus.

Assuming that $\vec{B}_{dip}$ arises from electrons with orbital moment l, (12) may be recast by using the equivalent operators (Ref. [3], 17.43, 17.44).

$$3(\vec{s}\vec{r})\vec{\hat{r}} - \vec{s}, \longrightarrow \frac{2}{(2l+3)(2l-1)} \left[l(l+1)\vec{s} - \frac{3}{2}(\vec{l}\,\vec{s})\,\vec{l} - \frac{3}{2}\vec{l}\,(\vec{l}\,\vec{s}) \right] \tag{14}$$

which gives

$$\vec{B}_{dip} = \frac{4\mu_B}{(2l+3)(2l-1)} \langle\Phi| \frac{S(r)}{r^3} \left[l(l+1)\vec{s} - \frac{3}{2}(\vec{l}\,\vec{s})\,\vec{l} - \frac{3}{2}\vec{l}\,(\vec{l}\,\vec{s}) \right] |\Phi\rangle \tag{15}$$

The projections of $\vec{B}_{orb}$, $\vec{B}_{dip}$ on the quantization axis ζ are

$$B_\zeta^{orb} = 2\mu_B \langle\Phi| \frac{S(r)}{r^3} l_\zeta \,|\Phi\rangle, \tag{16}$$

$$B_\zeta^{dip} = \frac{4\mu_B}{(2l+3)(2l-1)} \langle\Phi| \frac{S(r)}{r^3} \left[l(l+1)s_\zeta - \frac{3}{2}(\vec{l}\,\vec{s})\,l_\zeta - \frac{3}{2}l_\zeta\,(\vec{s}\,\vec{l}) \right] |\Phi\rangle \tag{17}$$

A. B_c in WIEN code

B_c is calculated by program *mixer*. First, the subroutine *hyper* calculates the electron density *hyperf* at the spherical slab around the nucleus

$$hyperf = \frac{q_{el}}{\frac{4}{3}\pi(r_T^3 - r_1^3)} \,, \tag{18}$$

where q_{el} is number of electrons in the spherical slab

$$q_{el} = \int_{r_1}^{r_T} \rho_{00}(r)dr \tag{19}$$

ρ_{00} is the spherical component of the radial density, r_T is Thomas radius, r_1 is the first point in spherical mesh of given atom. *hyperf* has three indexes: $hyperf(atom, elc, spin)$, elc= 1,2,3,4 correspond to valence, semicore, core and total density, $spin$=1,2 corresponds to $\uparrow$, $\downarrow$. Afterwards in the main program *mixer* B_c is calculated in kG:

$$B_c(atom, elc) = 524.3 \left[hyperf(atom, elc, 1) - hyperf(atom, elc, 2) \right]. \tag{20}$$

III. B_{orb} AND B_{dip} IN WIEN2K

Equations (11, 15) require evaluation of the mean value of a single particle operator $\hat{X}(r, \vec{l}, \vec{s})$ that belongs to a specific atom. We'll neglect any contributions beyond corresponding atomic sphere. Total mean value is then the sum over orbital numbers (note that we neglect the contribution nondiagonal in l, i.e. the crossterms $\langle l, m|\hat{X}|l'm'\rangle$; $l \neq l'$). The population matrix that is calculated by the program *lapwdm* may then be used to evaluate the mean value.

A. Population matrix

In the atomic spheres the basis functions are atomic-like. As a consequence their angular part is described by spherical functions $Y_{lm}(\hat{r})$. The Kohn-Sham eigenfunctions φ_i are linear combinations of the basis functions, thus in a given atomic sphere they may be represented as linear combinations of $Y_{lm}(\hat{r})$ too. The population matrix $\hat{n}_l$ is defined as an operator in the $|l, m, \sigma>$ subspace ($m_l = -l, ...l$; $\sigma = \pm 1/2$) of the atomic-like states. The matrix elements of $\hat{n}_l$ as calculated in WIEN2k are (cf. SRC_lapwdm/*xsplt.f*):

$$\hat{n}_{l,m\sigma,m'\sigma'} = \sum_{\varepsilon_i \leq E_F} \langle l, m', \sigma'|\varphi_i\rangle\langle\varphi_i|l, m, \sigma\rangle. \tag{21}$$

Note that $\hat{n}_{WIEN2k}$ defined in this way is hermitean conjugate to the population matrix introduced by Shick *et al.* ([4] eq. 12). The population matrix is hermitean: $n_{l,m\sigma,m'\sigma'} = (n_{l,m'\sigma',m\sigma})^*$. The cross terms $\sigma = \mp1/2$, $\sigma' = \pm1/2$ are nonzero when the spin-orbit interaction is present or the system has noncollinear spins. In WIEN the population matrix is calculated in the local coordinate system of given site.

B. Mean value of on-site operators

The mean value of the single particle operator $\hat{X}$ may be expressed as

$$\langle\Phi|\hat{X}|\Phi\rangle = \sum_{\varepsilon_i \leq E_F} \langle\varphi_i|\hat{X}|\varphi_i\rangle. \tag{22}$$

Assuming that $\hat{X}$ is nonzero within the atomic sphere only and neglecting the terms nondiagonal in l, this may be rewriten as

$$\sum_{\varepsilon_i \leq E_F} \sum_{lmm'}^{\sigma\sigma'} \langle\varphi_i|lm\sigma\rangle\langle lm\sigma|\hat{X}|lm'\sigma'\rangle\langle lm'\sigma'|\varphi_i\rangle =$$

$$\sum_{lmm'}^{\sigma\sigma'} \hat{X}_{lm\sigma,lm'\sigma'} \sum_{\varepsilon_i \leq E_F} \langle\varphi_i|lm\sigma\rangle\langle lm'\sigma'|\varphi_i\rangle = \tag{23}$$

$$\sum_{lmm'}^{\sigma\sigma'} \hat{X}_{lm\sigma,lm'\sigma'}\hat{n}_{l,m'\sigma',m\sigma} = \sum_l \operatorname{Tr}(\hat{n}_l^+\hat{X}) = \sum_l \operatorname{Tr}(\hat{X}^+\hat{n}_l).$$

Again note that hermitean conjugate of $\hat{n}$ appears in above expression because of the definition (21). If more conventional definition of $\hat{n}$ as in [4] would be used $\langle\hat{X}\rangle = \sum_l \operatorname{Tr}(\hat{n}_l\hat{X})$.

IV. IMPLEMENTATION TO WIEN2K

A. Population matrix

Population matrices $\hat{n}_l$ are calculated by the package LAPWDM. The input data are described in Table I.

The principal subroutine is *l2main* that is shortened and modified version of *l2main* of the LAPW2 package. *l2main* calls subroutine *xsplt* that calculates the unsymmetrized population matrix denoted there as *xqtl*

$$xqtl(ly, lpy, mu, ii, nd) = \sum_{\varepsilon_i \leq E_F} \langle\varphi_i|lm'\sigma'\rangle\langle lm\sigma|\varphi_i\rangle \tag{24}$$

where mu is index of equivalent atom, $nd = 1..N_l$ is index, specifying the orbital number. Indices $ly = m+l+1$, $lpy = m' + l + 1$ numerates m, m', acquiring values $1 \leq ly, lpy, \leq 2l + 1$. ii is the spin index in the calculation with the spin-orbit:
$ii = 1, 2, 3$ corresponds to the sequence $\langle\uparrow \mid \uparrow\rangle, \langle\uparrow \mid \downarrow\rangle, \langle\downarrow \mid \downarrow\rangle$.

Note that because complete calculation of *xqtl* is done within the loop over the atom types, *xqtl* has no index specifying the type of the atom.

Matrix *xqtl* is then symmetrized by applying to it all symmetry operations as listed in struct file, summing the results and dividing it by the number of symmetry operations. The details of symmetrization depend on whether calculation is with or without the s-o coupling and whether it is spin-polarized or not.

The output of standard LAPWDM calculation are the matrices $x\hat{q}tl = \hat{n}_l$, the mean value of spin and orbital operator $\langle\vec{s}\rangle, \langle\vec{l}\rangle$ and their projection on the direction of magnetization (for s-o calculation) as specified in case.inso file. More information may be printed if $iprint = 0$ in lapwdm.f is changed to $iprint = n$ and the package is recompiled. For bigger n bigger output is obtained.

In WIEN2k the basis functions $|lm\sigma\rangle$ inside atomic spheres in general consist of four terms

$$|lm\sigma\rangle = \sum_{j=1}^4 d_{lm\sigma,j}^{(\vec{k})} u_{l,j}^{(\sigma)}(r) Y_{lm}(\hat{r}), \tag{25}$$

where $j=1$ corresponds to solution of atomic Dirac equation, $j=2$ is its energy derivative, $j=3,4$ corresppond to local orbitals. Coefficients of the expansion depend on the $\vec{k}$ point. When calculating matrix $\hat{n}$ the radial integrals

$$ri_{mat}(l, j, j', \sigma, \sigma') = \int_0^{R_{MT}} u_{l,j}^{(\sigma)}(r) u_{l,j'}^{(\sigma')}(r) r^2 dr \tag{26}$$

must be evaluated. This is done in subroutine *radint* outside the loop over $\vec{k}$ points and eigenvalues. When calculating $\langle\hat{X}\rangle$ again *radint* is used and instead of above integrals the integrals

$$ri_{mat}(l, j, j', \sigma, \sigma') = \int_0^{R_{MT}} u_{l,j}^{(\sigma)}(r) X(r) u_{l,j'}^{(\sigma')}(r) r^2 dr \tag{27}$$

are computed.

Calculation of $Tr(\hat{X}\hat{n}_l^+)$ is performed in subroutine *output* of LAPWDM package.

line	parameter	explanation
1	E_{min}	only eigenfunctions with $\varepsilon_i \geq E_{min}$ will be considered
2	N_{atoms}	number of atom types for which $\hat{n}$ will be calculated
$3..N_{atoms}$	$iatom$, N_l, $(l_1..l_{N_l})$	index of atom type, number of l's, orb. numbers.
$N_{atoms}+3$	k_{rad}, k_{ls}	r-index and (l, s)-index identifying operator $\hat{X}$.

TABLE I: Input data for LAPWDM. Last line is added *only* if calculation of $\hat{X}$ mean value is required. Third line is repeated N_{atom} times, E_{min} is important in case when it is desirable to obtain $\hat{n}$ separately for valence and semicore states (represented by local orbitals).

$krad$	$\hat{X}_r$	kls	$\hat{X}_{ls}$
1	$\hat{I}$	1	$\hat{I}$
2	$Sr^{-3}\hat{P}$	2	$\hat{s}_\zeta$
3	$Sr^{-3}\hat{P}$	3	$\hat{l}_\zeta$
4	$r^{-3}\hat{P}$	4	$(\hat{l}\,\hat{s})$
-11	$1/r$	5	l, s part of B_{dip} (cf. eq. 17)
-10-n	$1/r^n$	6	$\hat{l}_\zeta^2$
10+n	r^n	7	approximation to l, s part of B_{dip} (eq. 29)

TABLE II: Operators corresponding to input $krad$ (subroutine *radint*) and kls (*couplx*). $\hat{P}$ is projector on the large component of the relativistic wave function.

B. Mean value of operator $\hat{X}_l$

Operators in (16, 17) are products of $|\vec{r}|$ and $\vec{l}$, $\vec{s}$ dependent operators:

$$\hat{X} = \hat{X}_r\,\hat{X}_{ls}. \tag{28}$$

For l fixed, the radial parts of the basis functions $u_l(r)$, $\dot{u}_l(r)$, ev. local orbitals are m-independent. Providing that in the relativistic correction (3) the energy ϵ is approximated by the center of the band, the radial and angular parts of $\langle\hat{X}\rangle$ (cf. eqs. 11, 12) may be calculated outside the loop over the eigenvectors. This is performed in subroutines *radint* and *couplx*. The data $krad, kls$ specifying $\hat{X}_r$, $\hat{X}_{ls}$ are given in the last line of the input file. Possible values of $krad, kls$ and corresponding operators are listed in Table II. Any allowed $krad$ could be combined with any allowed kls so that e.g. $krad = 1$, $kls = 2$ corresponds to s_ζ operator.

In older than WIEN2k_06 releases to calculate B_ζ^{dip} it was assumed that $\vec{l} \parallel \vec{s} \parallel \zeta$ i.e. in the spin-orbit coupling the cross term $(l_+s_- + l_-s_+)/2$ was neglected. (17) then reduces to:

$$B_\zeta^{dip} = \frac{2}{(2l+3)(2l-1)}\langle\Phi|\frac{S(r)}{r^3}s_\zeta\left[l(l+1) - 3\,l_\zeta^2\right]|\Phi\rangle. \tag{29}$$

This approximation to B_{dip} corresponds to $krad = 4$, $kls = 7$ (cf. Table II), but as demonstrated in the Appendix B it might be too rough or even misleading..

To obtain the hyperfine fields from in T from the mean value of operators in Tab. 2, specified by $krad = 3$ or $krad = 4$ and $kls = 3, 5, 7$ this mean value is multiplied by factor 12.5169.

To get more detailed output for $\langle\hat{X}\rangle$ calculation the value of variable *iprx* should be nonzero (main program *lapwdm*).

C. Approximations

Below the approximation made are summarized

- Relativistic mass enhancement is only approximatelly accounted for (Appendix A). Note that relativistic mass enhancement was fully included in the AVERX program of WIEN97.

- Contributions from the interstitial and from $\langle l||l'\rangle$ terms were neglected.

krad	kls	B_{orb} T	included
3	3	21.25088	relativistic mass enhancement
4	3	21.27513	no relativistic mass enhancement

TABLE III: Contribution B_{orb} of orbital momentum to hyperfine field.

krad	kls	B_{dip} T	included
3	5	-0.23792	relativistic mass enhancement, nondiagonal terms
4	5	-0.23820	no relativistic mass enhancement, nondiagonal terms
3	7	0.64983	relativistic mass enhancement, diagonal terms only
4	7	0.65057	no relativistic mass enhancement, diagonal terms only

TABLE IV: Contribution B_{dip} of on-site spin momentum to hyperfine field.

APPENDIX A: APPROXIMATE INCLUSION OF RELATIVISTIC MASS ENHANCEMENT

According to eq. 3 the relativistically enhanced electron mass is

$$M(r) = m \left[1 + \frac{\epsilon - V(r)}{2mc^2} \right].$$ (A1)

The problem with implementation is that:

- Enhancement M is r dependent, consequently when calculating the matrix elements the integration from $r=0$ to $r = R_{MT}$ must be performed.

- M is energy dependent, as a consequence radial integration must be performed inside the loop over the eigenvectors.

This would make the inclusion of M computationally costly and also programming is then not straightforward. We thus neglected the energy dispersion and put

$$\epsilon_i(\vec{k}) \cong E_l$$ (A2)

where E_l is the energy used for the orbital number l and selected atom to obtain the radial part of the orbital. M is then no longer energy dependent and the radial integrals can be calculated outside the loop over eigenvectors, as until now. Only *atpar.f* and *radint.f* subroutines need to be slightly modified and increase of the CPU time is negligible. Note that in atomic units $m = 1$. An alternative (not implemented) would be to assume that the enhancement is small and use an approximation

$$S(r) = 1/M(r) = m \left[1 - \frac{\epsilon - V(r)}{2mc^2} \right].$$ (A3)

APPENDIX B: TEST EXAMPLE - HEXAGONAL GADOLINIUM

Standard WIEN calculation using GGA (option 13 in gd.in0) + spin-orbit coupling ($\vec{M}||[001]$) was performed. Unit cell parameters $a = 6.878606$ a.u., $c=10.923188$ a.u. Number of $\vec{k}$ points in Brillouin zone was 2000, $R_{MT}k_{max} =7$, R_{MT}(Gd)=3 a.u.
Converged magnetic moments were
μ_s(Gd)=7.208 μ_B, μ_s(4f)=6.7408 μ_B, μ_l(4f)=0.2301 μ_B.
Contact hyperfine field from the standard output B_c=29.224 T.
Values of B_{orb}, B_{dip} are given in Table III, IV. To compare older calculations (up to WIEN2k_06) with better ones (inclusion of relativistic mass enhancement, nondiagonal terms in B_{dip}) the calculation was performed for different $krad, kls$. We checked that the results obtained with (krad,kls)=(4,3) and (4,7) are identical to results in WIEN2k_06 with (krad,kls)=(3,3) and (3,5).
It is seen from Tables III, IV that while relativistic mass enhancement has little effect, the inclusion of the nondiagonal terms qualitatively changes B_{dip}. To analyze the reason for this difference we compare the calculation

for krad=3 and kls=5 with calculation for krad=3 and kls=7. All matrices involved in calculations are real. The matrix $\tilde{n}$ of the radial part is the same in both cases:

```
Radial matrix
                  up                                              dn
   -3     -2     -1      0      1      2      3      -3     -2     -1      0      1      2      3
 7.990  0.000  0.000  0.000  0.000  0.000  0.001   0.000 -0.347  0.000  0.000  0.000  0.000  0.000
 0.000  7.971  0.000  0.000  0.000  0.000  0.000   0.000  0.000 -0.464  0.000  0.000  0.000  0.000
 0.000  0.000  7.965  0.000  0.000  0.000  0.000   0.000  0.000  0.000 -0.516  0.000  0.000  0.000
 0.000  0.000  0.000  7.960  0.000  0.000  0.000   0.000  0.000  0.000  0.000 -0.550  0.000  0.000
 0.000  0.000  0.000  0.000  7.964  0.000  0.000   0.000  0.000  0.000  0.000  0.000 -0.497  0.000
 0.000  0.000  0.000  0.000  0.000  7.971  0.000  -0.005  0.000  0.000  0.000  0.000  0.000 -0.398
 0.001  0.000  0.000  0.000  0.000  0.000  7.997   0.000  0.003  0.000  0.000  0.000  0.000  0.000
 0.000  0.000  0.000  0.000  0.000 -0.005  0.000   0.039  0.000  0.000  0.000  0.000  0.000 -0.072
-0.347  0.000  0.000  0.000  0.000  0.000  0.003   0.000  0.100  0.000  0.000  0.000  0.000  0.000
 0.000 -0.464  0.000  0.000  0.000  0.000  0.000   0.000  0.000  0.077  0.000  0.000  0.000  0.000
 0.000  0.000 -0.516  0.000  0.000  0.000  0.000   0.000  0.000  0.000  0.144  0.000  0.000  0.000
 0.000  0.000  0.000 -0.550  0.000  0.000  0.000   0.000  0.000  0.000  0.000  0.147  0.000  0.000
 0.000  0.000  0.000  0.000 -0.497  0.000  0.000   0.000  0.000  0.000  0.000  0.000  0.320  0.000
 0.000  0.000  0.000  0.000  0.000 -0.398  0.000  -0.072  0.000  0.000  0.000  0.000  0.000  0.429
```

Matrix of the operator $\hat{X}_{ls} = \frac{2}{(2l+3)(2l-1)}\left[l(l+1)\vec{s} - \frac{3}{2}\vec{l}(\vec{l}\,\vec{s}) - \frac{3}{2}(\vec{l}\,\vec{s})\vec{l}\right]$, corresponding to kls=5 is

```
-0.333  0.000  0.000  0.000  0.000  0.000  0.000   0.000 -0.408  0.000  0.000  0.000  0.000  0.000
 0.000  0.000  0.000  0.000  0.000  0.000  0.000   0.000  0.000 -0.316  0.000  0.000  0.000  0.000
 0.000  0.000  0.200  0.000  0.000  0.000  0.000   0.000  0.000  0.000 -0.115  0.000  0.000  0.000
 0.000  0.000  0.000  0.267  0.000  0.000  0.000   0.000  0.000  0.000  0.000  0.115  0.000  0.000
 0.000  0.000  0.000  0.000  0.200  0.000  0.000   0.000  0.000  0.000  0.000  0.000  0.316  0.000
 0.000  0.000  0.000  0.000  0.000  0.000  0.000   0.000  0.000  0.000  0.000  0.000  0.000  0.408
 0.000  0.000  0.000  0.000  0.000  0.000 -0.333   0.000  0.000  0.000  0.000  0.000  0.000  0.000
 0.000  0.000  0.000  0.000  0.000  0.000  0.000   0.333  0.000  0.000  0.000  0.000  0.000  0.000
-0.408  0.000  0.000  0.000  0.000  0.000  0.000   0.000  0.000  0.000  0.000  0.000  0.000  0.000
 0.000 -0.316  0.000  0.000  0.000  0.000  0.000   0.000  0.000 -0.200  0.000  0.000  0.000  0.000
 0.000  0.000 -0.115  0.000  0.000  0.000  0.000   0.000  0.000  0.000 -0.267  0.000  0.000  0.000
 0.000  0.000  0.000  0.115  0.000  0.000  0.000   0.000  0.000  0.000  0.000 -0.200  0.000  0.000
 0.000  0.000  0.000  0.000  0.316  0.000  0.000   0.000  0.000  0.000  0.000  0.000  0.000  0.000
 0.000  0.000  0.000  0.000  0.000  0.408  0.000   0.000  0.000  0.000  0.000  0.000  0.000  0.333
```

Diagonal operator corresponding to kls=7 $\frac{2}{(2l+3)(2l-1)}s_\zeta\left[l(l+1) - 3\,l_\zeta^2\right]$ has the same diagonal elements, and zero nondiagonal elements. Because both operators $\tilde{n}$ and X_{ls} are real and have the same simple structure the diagonal elements of their product consist of two terms only:

$$(\tilde{n}^+ X)_{m_s m_l, m_s m_l} = \tilde{n}_{m_s m_l, m_s m_l}\, X_{m_s m_l, m_s m_l} + \tilde{n}_{m_s m_l, m_s-1 m_l+1}\, X_{m_s-1 m_l+1, m_s m_l}, \qquad (B1)$$

with the second term absent for kls=7.

Below the diagonal elements and their sums are compared for kls=5 and kls=7.

```
     kls=5
   <-3||-3><-2||-2><-1||-1> <0||0>  <1||1>   <2||2> <3||3>      sum       total
up -31.566   1.836  20.686  25.773  17.968  -2.035 -33.365     -0.702
dn   0.162   1.771   1.642   0.265  -1.163  -1.967  -0.245      0.464     -0.238


     kls=7
up -33.337   0.000  19.940  26.568  19.936   0.000 -33.365     -0.258
dn   0.162   0.000  -0.194  -0.481  -0.369   0.000   1.790      0.908      0.650
```

It is now clear that the big difference between calculation with the diagonal operator and the full operator originates from the cancelation of the large diagonal elements - the resulting sum is by two orders of magnitude smaller compared to the largest diagonal element.

Important conclusion is that the approximation for B_{dip} used in WIEN2k_06 and previous releases could be very bad at least in cases when the spin-orbit coupling is not much smaller comparing to the exchange splitting.

APPENDIX C: NEW AND MODIFIED PARTS

Comparing to WIEN2k_06 release the following subroutines were modified
atpar, couplx, l2main, lapwdm, modules, radint, xsplt.
There are three new subroutines:
printx, xoper, xqproduct.
Subroutine *couplx* now calculate matrices of all components of spin and orbital momentum in crystal coordinate system $(s_x, s_y, s_z, l_x, l_y, l_z)$ and in coordinate system where quantization axis is $\parallel$ to the magnetization $(s_\xi, s_\eta, s_\zeta, l_\xi, l_\eta, l_\zeta)$. *xoper* calculate desired matrices of $\hat{X}_{ls}$ operator by multiplying s_α and l_α matrices. Any new operator may be added there and also the new $\hat{X}(r)$ operator may be added by modifying subroutine *radint*.

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[1] P. Novák, *Classical dipolar field and anisotropy in magnetic systems.*
[2] S. Blügel, H. Akai, R. Zeller, and P.H. Dederichs, Phys. Rev. B**35**, 3271 (1987).
[3] A. Abragam, B. Bleaney, *Electron Paramagnetic Resonance of Transition Ions*, Clarendon Press, Oxford 1970.
[4] A.B. Shick, A.I. Liechtenstein, and W.E. Pickett, Phys. Rev. B**60**, 10763 (1999).