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Hyperfine fields and local lattice relaxation at $4d$ and $5sp$ impurities in bcc iron

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The magnetic hyperfine fields B_{hf} for the $4d$ and $5sp$ elements Rb through Xe at the substitutional site in ferromagnetic iron were derived from *ab initio* calculations of the electronic structure. The full-potential linearized augmented plane-wave technique for $\mathrm{Fe}_{15}X$ supercells was used for a density functional with generalized-gradient corrections. The influence of lattice relaxation on the position of the nearest neighbor to the oversized impurities was calculated self-consistently. The values obtained for B_{hf} are in most cases in excellent agreement with the experimental results, except for Rb and Sr. For the series Ag through Xe systematic calculations with different choices for the density functional and also a larger supercell ($\mathrm{Fe}_{31}X$), where the position of the second neighbors could be relaxed as well, gave only slightly different results. The calculated impurity-induced volume changes and magnetic moments account well for available experimental results from lattice constant data and magnetization measurements, respectively.

I. MOTIVATION

Hyperfine interaction techniques, based on the observation of the coupling between nuclear moments and internal fields, have been widely used to study condensed matter on an atomic scale. The magnetic hyperfine fields at impurities in the simple ferromagnetic metals Fe, Co, Ni, and Gd, in particular, can be measured with a wide variety of experimental methods. A very abundant amount of data material is therefore available in the literature.[1] In this paper we demonstrate that the experimental trends can now be quantitatively reproduced with *ab initio* full-potential band-structure calculations even for rather heavy elements.

The early attempts towards a qualitative understanding of hyperfine fields at impurities in metals,[2] particularly in iron, were stimulated by the rather complete series of experimental data available for the $4d$ and $5sp$ impurities treated in this work. Numerous parametrized treatments have been reported in the literature.[3,4] All of them are able to reproduce the strong Z dependence qualitatively by adjusting parameters. The experimental trend could also be qualitatively reproduced by *ab initio* calculations using a cluster model.[5,6] A breakthrough in the theoretical understanding was achieved, for lighter impurity elements in particular, when the parameter-free density-functional method in the Green's-function KKR version was applied to the problem of hyperfine fields in ferromagnets.[7–9] A comprehensive review is given in Ref. 10. For the heavier impurities of the $4d$ and $5sp$ series these pioneering calculations have, however, not reproduced the experimental data. As possible reasons for this failure neglect of the lattice relaxation about the impurity or lack of a fully self-consistent treatment of the potentials at the neighboring atoms have been suggested.

In order to apply an independent computational procedure to this problem we have started to calculate the magnetic hyperfine fields at impurities in Fe using a supercell method. The full-potential linearized augmented plane-waves (FLAPW) technique[11,12] based on density-functional theory, in particular, has been highly successful in calculating various properties of solids, including the hyperfine interaction at nuclei.[13,14] In recent years the computing power available as well as the improvement of the code efficiency have rendered the application of supercell calculations to impurity problems practically feasible. In this contribution we have studied the $4d$ and $5sp$ elements. These impurities, where strong discrepancies between experiments and the earlier calculations had been observed, are still light enough such that a fully relativistic treatment is not necessary. The scalar-relativistic approach used in this work should be sufficient.

This paper is organized as follows: We first present a short description of the theoretical methods used, followed by the numerical results. Next we compare with experimental data and earlier calculations. We close with a summary of our conclusions and a short outlook.

II. CALCULATIONS

The theoretical calculations of the electronic structure and hyperfine fields were performed using the full-potential linearized augmented plane-waves (FLAPW) method, based on density-functional theory. We have chosen the computer code WIEN97 (Ref. 15) that has been shown to yield reliable results for the band structure of various solids. It also allows the calculation of hyperfine fields.[14]

The FLAPW method is well suited to calculate the electron distribution in the solid state for periodic structures. To apply it to the impurity problem one artificially constructs a large enough unit cell with one or a few impurity atoms to effectively eliminate their interaction. For precision calculations the required number of atoms is quickly becoming too large for the available computing resources. Particularly for studies of local properties like hyperfine fields in ferromagnetic metals, however, one could imagine using rather small supercells. In order to investigate the suitability of this ap-

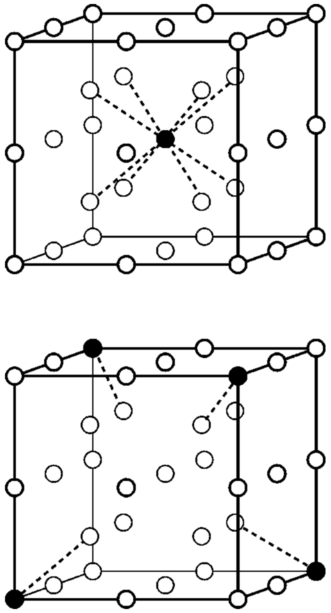

FIG. 1. $Fe_{15}X$ simple cubic unit cell and 1/8 of an fcc $Fe_{31}X$ supercell used in the calculations. Black symbols are impurities, the dashed lines connect impurities and their nearest neighbors.

proach, we have made systematic calculations using a unit cells with 15 matrix atoms per impurity. In order to check that the results are representative for the dilute impurity, a cell with 31 matrix atoms per impurity was also used for some cases. The simple cubic $Fe_{15}X$ cell and 1/8 of the fcc $Fe_{124}X_4$ (effectively $Fe_{31}X$) cell employed in this work are shown in Fig. 1.

The calculations were performed with the FLAPW code WIEN97 using the most advanced density functional with generalized-gradient corrections (GGA96) (Ref. 16) for the exchange-correlation (XC) energy. It has been demonstrated that in this way the properties of pure Fe can be reproduced quite well.[17] Some systematic checks with an earlier GGA version (GGA92) (Ref. 18) and the simple local-density approximation (LDA) (Ref. 19) were performed to investigate the influence of different choices.

In the FLAPW procedure wave functions, charge density, and potential are expanded in spherical harmonics within nonoverlapping atomic spheres of radius R_{MT} and in plane waves in the remaining space of the unit cell. R_{MT} values of 2.2 a.u. for Fe and 2.4 a.u. for the impurity were chosen throughout. The maximum l for the waves inside the atomic spheres was confined to $l_{max}=10$. The wave functions in the interstitial region were expanded in plane waves with a cutoff of $k_{max}=8/R_{MT}$, while the charge density was Fourier expanded up to $G_{max}=16$. A mesh of 35 special k points was taken in the irreducible wedge of the Brillouin zone for the $Fe_{15}X$ cell (10 for $Fe_{31}X$). The lowest valence states were either $3d$(Rb), $4s$(Sr-In), $4p$(Cd-Sb), or $4d$(Te-Xe). All these technical parameters were checked to yield converged results. For the calculations in the unrelaxed Fe matrix the experimental lattice constant of 5.4235 a.u. was taken.

The hyperfine fields acting at the nuclei are, in the absence of orbital magnetism, dominated by the Fermi contact interaction. For the scalar-relativistic wave functions used in the present calculations the effective spin density at the nucleus was computed following Ref. 20 by averaging over a small region near the nucleus with diameter $r_T = Ze^2/mc^2$.

III. RESULTS

For all systems a calculation for the unrelaxed $Fe_{15}X$ cell with the lattice constant of 10.847 a.u. was performed first. The obtained values for B_{hf} are included in Table I, and the calculated forces at the nearest-neighbor (NN) position are shown in Fig. 2(a). Positive forces are directed away from the impurity. The force resulting from this calculation gave a good guideline to the amount of lattice expansion and NN relaxation expected in the optimal structure. Around this extrapolated structure several geometries were then chosen, for which the total energy and the NN forces were determined. By interpolation the final lattice constant and NN position were found, at which the total energy was at a minimum and the NN force vanished. In Table I the lattice constants, NN distances and final B_{hf} values are summarized. In Fig. 2 the final geometrical parameters are compared to the forces obtained in the unrelaxed calculation. Clearly, a very similar trend is evident in all three curves. In Fig. 3 the final B_{hf} values are shown together with the experimental numbers of Ref. 1.

For the series Ag-Xe in the $Fe_{15}X$ geometry the B_{hf} values were also calculated with the two other density functionals mentioned above. As can be seen in Table II, the results for these are quite close to the ones with GGA96. The small changes relative to the results in Table I are due to the use of

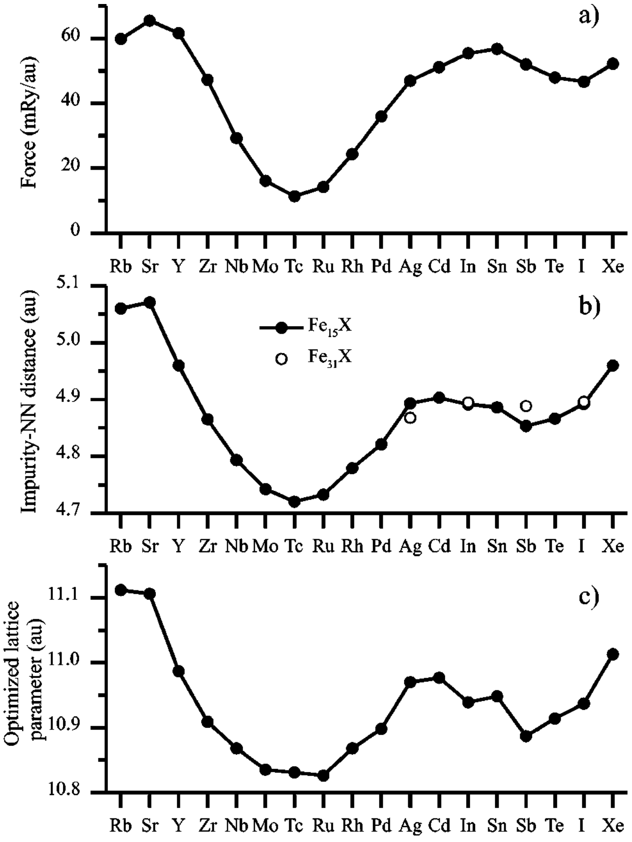

FIG. 2. (a) Forces (in mRy/au) on nearest-neighbor Fe (NN) in an unrelaxed lattice ($a_0=10.847$ a.u.), (b) distance between impurity and NN for optimized $Fe_{15}X$ and $Fe_{31}X$ supercells, (c) lattice parameter (a.u.) for optimized $Fe_{15}X$.

TABLE I. Calculated lattice parameters (a and d_{NN} in a.u.) of the $Fe_{15}X$ supercells and resultant hyperfine fields (B_{hf} in T) for the $4d$ and $5sp$ impurities as compared to results without lattice relaxation, experimental data, and earlier calculations.

	a	d_{NN}	B_{hf} unrel	B_{hf} relax	B_{hf} expt	Refs. 8 and 9	Ref. 10
Rb	11.112	5.060	-17.3	-26.1	5.4(10)	3.6	
Sr	11.106	5.071	-30.3	-26.1	-10.0(30)	-19.0	
Y	10.987	4.960	-25.0	-30.4	-30.7(4)	-25.4	-26.3
Zr	10.909	4.865	-26.9	-29.8	-27.4(4)	-29.3	-27.1
Nb	10.868	4.793	-24.9	-26.1	-26.57(2)	-23.8	-27.8
Mo	10.835	4.742	-25.7	-26.1	-25.60(1)	-22.8	-29.3
Tc	10.831	4.720	-30.9	-31.2	-31.7(5)	-26.7	-31.7
Ru	10.826	4.732	-47.1	-47.7	-50.5(1)	-31.6	-39.2
Rh	10.868	4.779	-56.7	-57.6	-56.0(1)	-34.9	-44.4
Pd	10.898	4.821	-55.4	-56.2	-59.4(12)	-34.2	-46.2
Ag	10.970	4.893	-47.9	-51.0	-44.72(2)	-31.4	-40.8
Cd	10.977	4.903	-41.4	-43.4	-39.2(4)	-30.9	
In	10.939	4.891	-40.4	-34.8	-28.68(3)	-27.6	
Sn	10.948	4.886	-23.8	-15.0	-8.60(2)	-22.2	
Sb	10.887	4.853	10.6	20.5	23.27(1)	-7.7	
Te	10.914	4.866	49.4	68.0	68.1(4)	11.6	
I	10.937	4.892	102.2	128.3	114.52(2)	34.8	
Xe	11.013	4.960	163.3	154.7	157.0(50)	60.9	

a slightly different geometry in these comparative studies.

For the same series (Ag-Xe) calculations with the $Fe_{31}X$ cell were performed. The geometry was estimated from the $Fe_{15}X$ result by taking the same impurity volume and NN distance. For the second neighbor (NNN) distance a starting value was chosen according to the displacement ratio expected between NN and NNN proportional to $1/d^2$. The forces at NNN obtained in this way, also included in Table II, show a rather systematic trend. The B_{hf} values from this calculation are practically not changed from the $Fe_{15}X$ values. For a few cases (Ag, In, Sb, and I) the three free atomic positions were then slightly readjusted to resultant negligible forces in two to three more cycles of calculation. The B_{hf} results were unchanged and the NN displacements obtained were almost identical to the $Fe_{15}X$ values, as can be seen in Fig. 2(b). No total energy minimizations could be performed to determine the $Fe_{31}X$ lattice constants from first principles because of the long computer running times. For the $Fe_{15}X$ calculations, however, the NN displacements closely parallel the independently calculated unit-cell expansion [Figs. 2(b) and (c)]. Our procedure used to estimate the $Fe_{31}X$ unit cell volume is then justified by the fact that the displacements change only very slightly in going from $Fe_{15}X$ to $Fe_{31}X$.

Since for the cases treated here the hyperfine fields at the nucleus are determined by the polarization of the s electrons, the density of states with s symmetry (s DOS) in the impurity sphere has been extracted from the calculations. As has been pointed out before,[8] this quantity has a rather complex energy dependence, and the position of the Fermi energy E_F in this structure is important. For the presently studied properties we are only interested in the sum of the contributions of all valence electrons with s character. The integral of the s DOS for both spin directions is therefore plotted for representative cases in Fig. 4. Their difference, directly related to

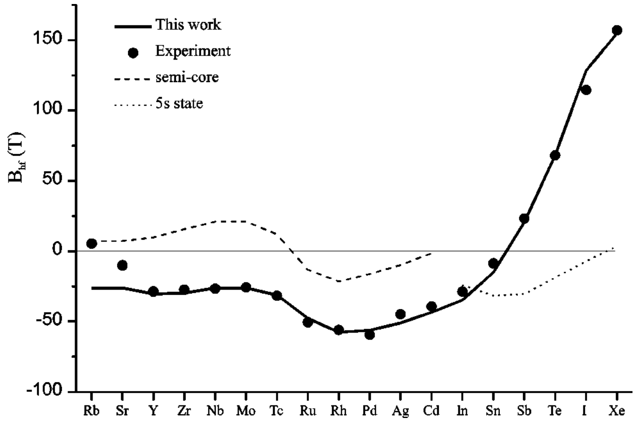

FIG. 3. Calculated hyperfine fields compared to experimental data. The semicore contributions ($4s$ for Rb-Cd) and the contribution from the split-off $5s$ state (for In-Xe) are shown separately.

TABLE II. Hyperfine fields (B_{hf} in T) for the $5sp$ elements calculated in the $Fe_{15}X$ supercell with various XC functionals as compared to $Fe_{31}X$ results and residual forces on NNN (in mRy/a.u.).

	LDA	GGA92	GGA96	unrel	$Fe_{31}X$	F_{NNN}
Ag	-40.5	-50.5	-51.0	-47.9	-52.9	-4.9
Cd	-35.1	-43.3	-43.5	-41.4	-45.5	-3.0
In	-29.5	-35.1	-34.7	-34.6	-36.6	-5.4
Sn	-12.5	-15.8	-14.9	-18.0	-17.3	-6.6
Sb	19.4	21.3	21.7	11.3	14.5	-4.6
Te	64.5	66.7	68.0	49.3	57.5	-0.6
I	123.3	128.2	128.7	102.5	128.0	5.4
Xe	143.5	161.6	161.8	163.2	165.1	10.1

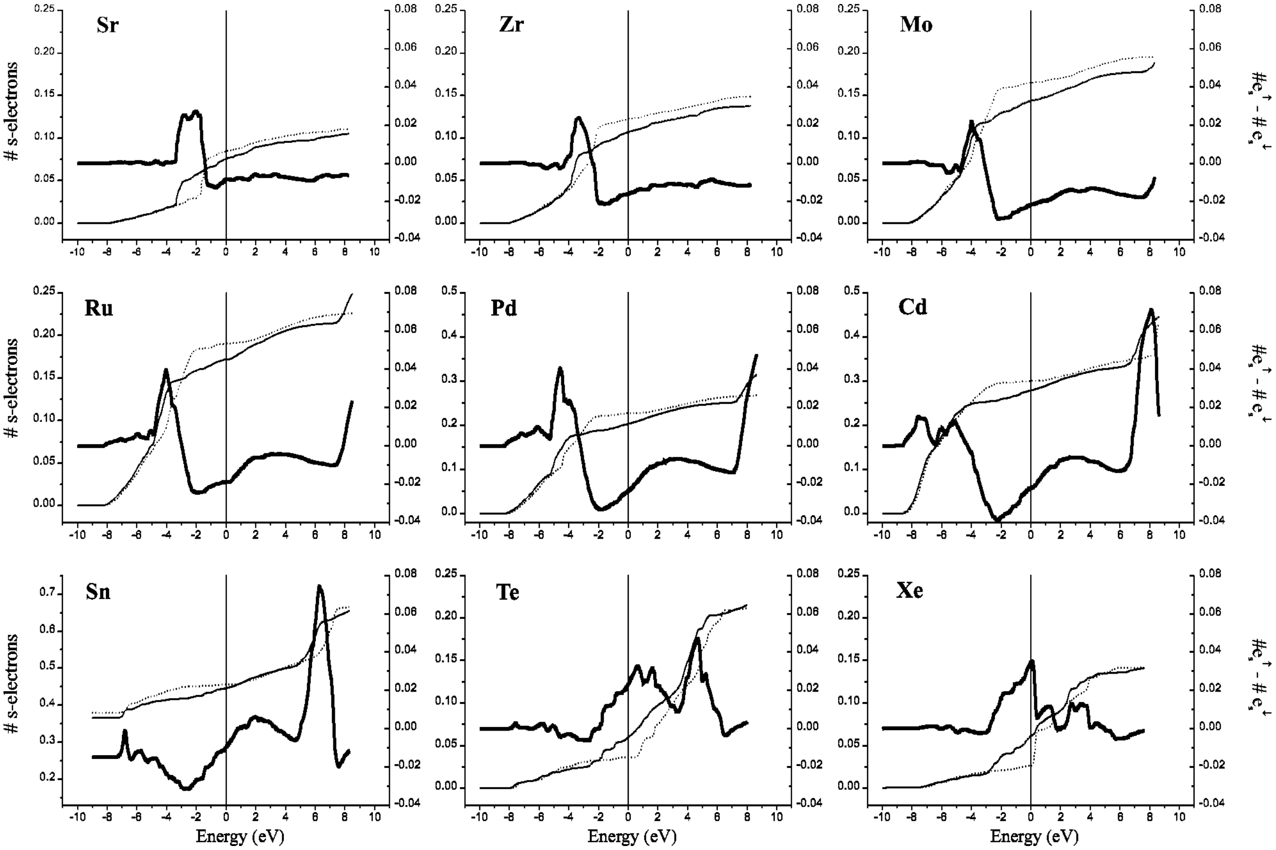

FIG. 4. Integral of partial spin (up/down) s DOS (electrons) within R_{MT} of the impurity (left scale) and their difference (right scale) for representative cases. Mind the change in scale for Pd, Cd, and Sn.

the B_{hf} contribution of the valence electrons, is the most important information in Fig. 4 and allows direct conclusions on the origin of B_{hf}. Note that the B_{hf} values reported in Tables I and II and in Fig. 3 were not calculated from this atomic s DOS difference, but directly from the s DOS difference at the nucleus.

Obviously also deeper levels, being polarized by the unpaired spin distribution of the valence electrons, will make a contribution to B_{hf}. In Fig. 3 this core polarization contribution is included separately for the series Rb-Cd. Since core polarization is rather insignificant for the heavier sp elements, here the contribution of the split-off valence state of essentially $5s$ character is shown.

IV. COMPARISON WITH EXPERIMENTAL DATA AND EARLIER CALCULATIONS

A. Hyperfine fields

As evident from Fig. 3 and Table I, the present theoretical values of B_{hf} for the $4d$ and $5sp$ impurities in Fe are very close to the experimental results, with the exceptions of Rb and Sr. From this quantitative agreement one may safely conclude that even for heavier impurities the electronic structure is well described by the FLAPW technique.

The reason for the substantial disagreement in the cases of Rb and Sr is not completely clear. For some impurities the integrated difference s DOS has a peak in the region of E_F, as to be seen for the examples Sr and Xe in Fig. 4. If the steep slope of such a peak is very close to E_F, any small error in the calculation could shift the relative position slightly and thus change the resulting B_{hf} substantially. For Rb and Sr the calculated separation of this peak from E_F is, however, so large, that it is difficult to conceive that it would in reality coincide with E_F. One should therefore also consider the possibility that a problem in the experimental measurements might be the reason for the discrepancy. Both measurements were performed with the low-temperature nuclear orientation (NO) technique on implanted samples.[21] This method allows no independent check of the fraction of nuclei occupying substitutional sites, a critical parameter in the experimental analysis. Actually the occupation of a unique substitutional site of Rb and Sr on implantation into iron appears quite unlikely for thermodynamic reasons.[22]

Also included in Table I are results of earlier *ab initio* calculations with the Korringa-Kohn-Rostoker (KKR) method in the atomic sphere approximation (ASA).[8,9] Since only graphs were published, numbers were read from these. A complete set of results is available in the literature only for a calculation where just the impurity potential was treated self-consistently. This approximation can account for the experimental data for $4sp$ impurities reasonably well. It apparently fails, however, for the $5sp$ elements. Furthermore, there are substantial disagreements in the $4d$ series, though the qualitative trends are reproduced. Fortunately for the $4d$ impurities more advanced calculations are available in the literature,[10] where apparently the potential for the neighboring Fe sites was calculated with the impurity present. The results come considerably closer to experiments, though still not as well as the present ones. In order to determine whether the choice of exchange potential was the cause for the difference between the theoretical numbers, a calculation using the LDA was performed for the representative case of Rh. The result agreed almost completely with the KKR calculation, thus demonstrating that our better agreement with experiment in these cases stems from the use of gradient correction.

The use of gradient corrections in density-functional calculations of hyperfine fields has been criticized[23] on the grounds that the gradient terms lead to unphysical divergences near the nucleus due to the strong Coulomb field there. The functional applied here does not suffer from this divergence with increasing normalized gradient, so its use is equivalent to the local-density approximation in the nuclear region. For the regions where the inner atomic orbitals interact with the outer ones to produce the electronic core polarization, however, the gradient terms will lead to a modification of the potential that considerably changes the calculated fields. Though there is no certainty that the chosen form for the gradient term, optimized to reproduce the valence charge distribution in simple molecules,[16,18] will improve the treatment, our much better agreement with experiment, particularly for Ru, Rh, and Pd, is strong evidence that it does so. Actually for the $4d$ impurities one expects an additional small contribution to the hyperfine field from the orbital moments, that would have to be incorporated in a complete treatment. Such a term has been included in a relativistic calculation of the hyperfine fields of the $5d$ impurities.[24] Even there it represents only a small correction. Since the spin-orbit interaction for the $4d$ elements is only about half that of the $5d$ series, the orbital contribution to B_{hf} is only expected to contribute values of the order of 3 T.

B. Lattice expansion

The calculated theoretical lattice constants of the $\mathrm{Fe}_{15}X$ supercells allow directly to determine the effective volume taken up by the impurity X. Since it is generally observed, however, that density-functional methods do not reproduce experimental lattice constants exactly, the calculated lattice parameters of the $\mathrm{Fe}_{15}X$ cells must not be compared with the experimental value for pure Fe, but with the corresponding theoretical one. We have therefore calculated the lattice constant for pure Fe in exactly the same way as for the impurity cells. The value obtained, $a_{Fe} = 5.38$ a.u., is less than 1% smaller than the experimental result at $T = 0$. The effective impurity volume obtained using this number was converted to a lattice expansion coefficient $d \ln a/dc$. The numbers thus obtained are shown in Fig. 5 together with the experimental information available for impurities in Fe.

It is obvious that the general trend of the volume effect calculated reproduces the experimental information quite well. Since the present calculation relies on the total energy of a rather large unit cell, numerical uncertainties could be significant. It is thus not clear whether the fine structure of the lattice expansion seen in Fig. 5, the drop from Sn to Sb, e.g., really has statistical significance.

Included in Fig. 5 is also the volume expansion expected from elastic continuum theory,[25] using the experimental atomic volumes and bulk moduli.[26] Apparently such a treatment is a very rough approximation to nature at best. It clearly does not take into account specific chemical bonding effects. Actually to obtain an even approximate agreement with the data using the continuum model, we have used for the bulk modulus of the impurity sphere not the value of the impurity B_{imp}, but the geometric mean $B_{eff} = \sqrt{B_{Fe} B_{imp}}$.

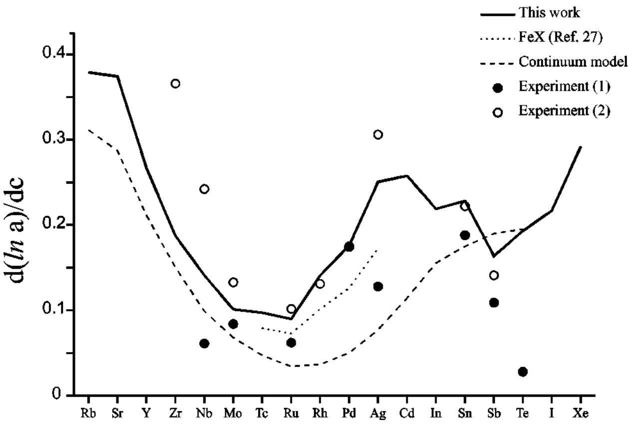

FIG. 5. Calculated lattice expansion $d \ln a/dc$ with a the lattice parameter and c the impurity concentration, evaluated for an alloy with $c = 6.25\%$ impurities, as compared to an early tabulation (experiment 1, Ref. 30, except Ref. 31 for Ag) and newer experiments (Experiment 2, Ref. 28, except Ref. 32 for Ag. Sn and Sb: recalculation). Expectations for a simple continuum model and theoretical calculations for ordered FeX alloys are also shown.

For some $4d$ elements earlier theoretical calculations for ordered FeX alloys with CsCl structure are available.[27] From the theoretical volumes calculated for these extremely concentrated supercells one can also estimate the effective impurity volume. It is quite reassuring that the numbers thus calculated for Tc-Ag come quite close to our present results.

Some comments are necessary as to the accuracy of the experimental lattice expansion information. For the soluble systems Zr, Nb, Mo, Ru, Rh, Pd, Sn, and Sb the data are quite old[28,29] and have apparently not been remeasured with modern techniques. We have in some cases reanalyzed the available data and obtained somewhat different coefficients from King.[30] For the insoluble system Ag in Fe more recent measurements on sputtered samples are available. Apparently the sample preparation technique has a substantial influence, however, as evident from the different results for Ag in Fe when produced by magnetron sputtering[31] or laser ablation.[32] For Te the point given in the old tabulation[30] is in complete disagreement with our result. It obviously comes from a wrong interpretation of the original data[33] that unfortunately do not really allow a determination of $d \ln a/dc$.

C. Magnetization

The systematic calculations performed in the present work also allow us to investigate the important question of the magnetization change of iron by foreign atoms. The $\mathrm{Fe}_{15}X$ supercell effectively represents an alloy with 6.25% atomic concentration of impurities. Since the spin polarization in all atomic spheres and in the interstitial region is directly calculated in the FLAPW procedure, the total (spin) magnetization is available. By subtracting the magnetization of 15 atoms in pure Fe, the effective impurity moment is obtained. Since such a subtraction process of large numbers is naturally prone to errors, some care must be taken in doing so, however. We have actually calculated the magnetization

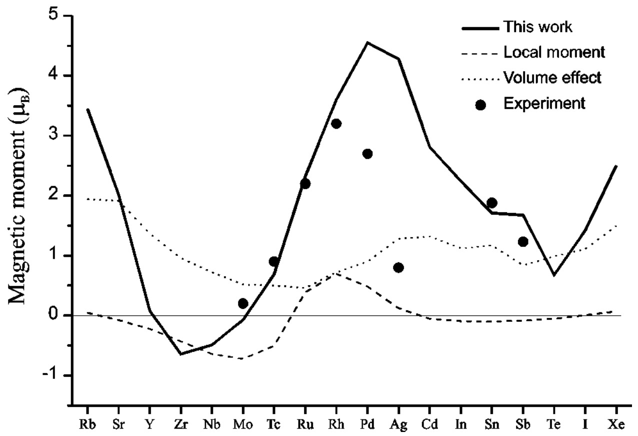

FIG. 6. Calculated impurity-induced moment as compared to results from magnetization measurements. The contribution of the local moment and the (hypothetical) effect of lattice expansion are shown separately.

of pure Fe with exactly the same parameters as in the impurity treatment, using the theoretical lattice constant as discussed above. The spin magnetization obtained in this way for Fe is $M_{Fe}=2.224\mu_B$. The resultant numbers for the impurities are shown in Fig. 6, together with some experimental data available for this property.

The rather imprecise experimental magnetization measurements are well reproduced by our calculation, with the exception of palladium[34] and particularly silver.[31] Silver, however, is exactly the case where also the experimental lattice expansion numbers are not consistent, so that the sample preparation should have a strong influence on the magnetization data. Obviously a measurement on samples produced by laser ablation would be highly interesting. This is even more the case, because for the impurity Ag the effective moment deviates strongly from the result of an earlier calculation with an unrelaxed lattice.[35]

Our calculations allow us to separate various contributions to the effective impurity moment: the local moment at the impurity, the moment change due to lattice expansion and the magnetization change at the NN (and further) Fe positions. The local moments obtained by us, also included in Fig. 6, are virtually identical to the previous treatment,[35] and also to the earlier less sophisticated ASA calculation,[7] for that matter. A comparison with experimental data shows that these difficult to measure quantities are apparently rather unreliable, as has also been found for the $5d$ impurity moments, where newer data[36] agree much better with the theoretical numbers.

The influence of lattice expansion on the effective impurity magnetization has been pointed out before.[37] In order to give a quantitative estimate of this effect, we have calculated the moment of Fe as function of volume. The calculated derivative $d\ln M/d\ln V=0.653$ can also be extracted from all earlier self-consistent calculations,[38,39,17] in reasonable agreement with the only experimental result available.[40] The volume effect on the effective impurity moment thus calculated is seen in Fig. 6 to make a significant contribution.

V. CONCLUSIONS

Taken together with earlier calculations using the Green's-function KKR method, the present calculations demonstrate that a rather quantitative account of the hyperfine fields acting at even heavy impurities in ferromagnets is possible with density-functional techniques. Within the FLAPW method even with rather small supercells values close to the experimental ones can be obtained. The inclusion of lattice expansion and lattice relaxation about the impurity actually was found to make only a small contribution even for the very oversized atoms treated in the present work. The discrepancies with experiment found in earlier Green's-function KKR treatments were traced to limiting the self-consistent treatment to the impurity site rather than to neglecting lattice relaxation. For the $5sp$ impurities (Ag-Xe) the results furthermore depend not much on the exchange-correlation potential chosen. For the d impurities, where core-polarization is important, the use of gradient corrections brings a substantial improvement of the B_{hf} values. In certain very isolated cases, when the change of spin polarization very close to the Fermi level is very abrupt, the presently applied approximations may not be sufficiently accurate, the alkali elements Cs (and possibly Fr) being particularly problematic.

The *ab initio* calculation of lattice expansion by impurities with the supercell FLAPW technique appears to reproduce experimental data astonishingly well. This will allow us to estimate with confidence also the corresponding parameters for systems where an experimental determination is not feasible.

The possibility to account for the magnetization changes by impurities in ferromagnets even with rather small supercells is an additional feature of the present calculations. Apparently the large extent of the magnetic influence of a single impurity is incorporated in this treatment implicitly by the cumulative effects at the solvent sites from several impurity atoms.

VI. OUTLOOK

Clearly the present investigation makes additional experimental and theoretical investigations highly desirable:

The magnetic fields acting on Rb and Sr in Fe should be remeasured, in order to clear up the discrepancy with the present calculation. This is unfortunately a quite difficult task, since these elements are completely insoluble in Fe and thus will probably not reach a substitutional site on implantation. The implantation of a radioactive precursor (Zr or Y), preferably at high energy, could perhaps eliminate this problem.

For Fr as impurity in Fe the hyperfine field has recently been measured.[41] Preliminary calculations for this case have shown that the strong lattice relaxation and the weakly bound valence electron will require a larger supercell than used in the present investigation. In addition the effects of spin-orbit interaction will have to be included.

Preliminary calculations of the type described here for the very light impurities (O, F, Ne) at a substitutional site have resulted in B_{hf} values incompatible with the experimental measurements. Obviously in these cases similar calculations for the interstitial sites could lead to further insight (as well as lattice location studies via channeling experiments).

The predictions for large lattice expansion due to Ag and In impurities in Fe could possibly be checked with modern precision lattice constant measurements. A remeasurement of the lattice expansion by the only weakly soluble impurities Zr and Nb, where the old data scatter widely, could be a further test of the present treatment.

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[1] G.N. Rao, Hyperfine Interact. **24/26**, 1119 (1985).

[2] D.A. Shirley and G.A. Westenbarger, Phys. Rev. **138**, A170 (1965).

[3] I. Sondhi, J. Chem. Phys. **62**, 1385 (1975).

[4] M.B. Stearns and J.M. Norbeck, Phys. Rev. B **20**, 3739 (1979).

[5] B. Lindgren and A. Rosen, Hyperfine Interact. **9**, 431 (1981).

[6] B. Lindgren, Hyperfine Interact. **49**, 357 (1989).

[7] H. Akai, M. Akai, S. Blügel, R. Zeller, and P.H. Dederichs, J. Magn. Magn. Mater. **45**, 291 (1984).

[8] M. Akai, H. Akai, and J. Kanamori, J. Phys. Soc. Jpn. **54**, 4246 (1985).

[9] H. Akai, M. Akai, and J. Kanamori, J. Phys. Soc. Jpn. **54**, 4257 (1985).

[10] H. Akai, M. Akai, S. Blügel, B. Drittler, H. Ebert, K. Terakura, R. Zeller, and P.H. Dederichs, Prog. Theor. Phys. Suppl. **101**, 11 (1990).

[11] D.D. Koelling and G.O. Arbman, J. Phys. F: Met. Phys. **5**, 2041 (1975).

[12] D.J. Singh, *Planewaves, Pseudopotentials and the LAPW Method* (Kluwer Academic, Boston, 1994).

[13] K. Schwarz and P. Blaha, Z. Naturforsch., A: Phys. Sci. **47**, 197 (1992).

[14] P. Dufek, P. Blaha, and K. Schwarz, Phys. Rev. Lett. **75**, 3545 (1995).

[15] P. Blaha, K. Schwarz, and J. Luitz, WIEN97, A Full Potential Linearized Augmented Plane Wave Package for Calculating Crystal Properties (Karlheinz Schwarz, Tech. Univ. Wien, Vienna, 1999). Updated version of P. Blaha, K. Schwarz, P. Sorantin, and S.B. Trickey, Comput. Phys. Commun. **59**, 399 (1990).

[16] J.P. Perdew, S. Burke, and M. Ernzerhof, Phys. Rev. Lett. **77**, 3865 (1996).

[17] M. Ekman, B. Sadigh, K. Einarsdotter, and P. Blaha, Phys. Rev. B **58**, 5296 (1998).

[18] J.P. Perdew, J.A. Chevary, S.H. Vosko, K.A. Jackson, M.R. Pederson, D.J. Singh, and C. Fiolhais, Phys. Rev. B **46**, 6671 (1992).

[19] J.P. Perdew and Y. Wang, Phys. Rev. B **45**, 13 244 (1992).

[20] S. Blügel, H. Akai, R. Zeller, and P.H. Dederichs, Phys. Rev. B **35**, 3271 (1987).

[21] A.L. Allsop, D.I. Bradley, V.R. Green, and N.J. Stone, Hyperfine Interact. **15/16**, 313 (1983).

[22] P. Herzog, Hyperfine Interact. **60**, 563 (1990).

[23] M. Battocletti, H. Ebert, and H. Akai, Phys. Rev. B **53**, 9776 (1996).

[24] H. Ebert, R. Zeller, B. Drittler, and P.H. Dederichs, J. Appl. Phys. **67**, 4576 (1990).

[25] J.D. Eshelby, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press, New York, 1955), Vol. 3.

[26] K.A. Gschneider, Jr., in *Solid State Physics* (Ref. 25), Vol. 16.

[27] V.L. Moruzzi, P.M. Marcus, and S.L. Qiu, Phys. Rev. B **52**, 3448 (1995).

[28] E.P. Abrahamson and S.L. Lopata, Trans. Metall. Soc. AIME **236**, 76 (1966).

[29] W.B. Pearson, *A Handbook of Lattice Spacings and Structures of Metals and Alloys* (Pergamon, Oxford, 1967), Vol. 2.

[30] H.W. King, J. Mater. Sci. **1**, 79 (1966).

[31] N. Kataoka, K. Sumiyama, and Y. Nakamura, J. Phys. F: Met. Phys. **15**, 1405 (1985).

[32] M. Störmer, S. Fähler, H.U. Krebs, A. Crespo, and P. Schaaf, Nucl. Instrum. Methods Phys. Res. B **122**, 503 (1997).

[33] S. Chiba, J. Phys. Soc. Jpn. **10**, 837 (1955).

[34] S.L. Zhang, K. Sumiyama, and Y. Nakamura, J. Magn. Magn. Mater. **73**, 58 (1988).

[35] B. Drittler, N. Stefanou, S. Blügel, R. Zeller, and P.H. Dederichs, Phys. Rev. B **40**, 8203 (1989).

[36] H. Ebert, R. Wienke, G. Schütz, and W.M. Temmerman, Physica B **172**, 71 (1991).

[37] B.W. Corb, R.C. O'Handley, and N.J. Grant, Phys. Rev. B **27**, 636 (1983).

[38] J.F. Janak, Phys. Rev. B **5**, 2206 (1979).

[39] V.L. Moruzzi, P.M. Marcus, K. Schwarz, and P. Mohn, Phys. Rev. B **34**, 1784 (1986).

[40] E.I. Kondorskii and V.L. Sedov, Zh. Éksp. Teor. Fiz. **38**, 773 (1960) [Sov. Phys. JETP **11**, 561 (1960)].

[41] B. Will, P. Herzog, R. Paulsen, J. Camps, P. DeMoor, P. Schuurmans, N. Severijns, A. VanGeert, L. Vanneste, I. Berkes, M. DeJesus, M. Lindroos, and P. VanDuppen, Phys. Rev. B **57**, 11 527 (1998).