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Effect of lattice relaxations on the hyperfine fields of heavy impurities in Fe

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Abstract

We present first-principles calculations of hyperfine fields of heavy impurities in BCC Fe. In particular, the effect of lattice relaxations on the calculated hyperfine fields are studied. The calculations are based on a full-potential Korringa–Kohn–Rostoker Green's function method for defects and employ the local spin–density approximation for the exchange and correlation effects. The non-spherical parts of the potential and the charge density are treated correctly, while the forces are calculated by an ionic version of the Hellmann–Feynman theorem. Lattice statics methods are used to describe the longer ranged relaxations. The calculated hyperfine fields of 5sp and 6sp elements are compared with the available experimental data and it is shown that the inclusion of lattice relaxations in the calculation improves the agreement with experiments. © 2001 Elsevier Science B.V. All rights reserved.

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The hyperfine fields of impurities in the ferromagnetic hosts Fe, Co, and Ni have been extensively studied and a huge amount of experimental data exists [1,2]. The present calculations are motivated by recent measurements [3,4] of the hyperfine fields of Cs and in particular of Fr in Fe. The measured hyperfine field of Fr is a factor of 5 smaller than the previous KKR calculations [5–9] predicted, which were, however, based on spherical potentials, i.e., using atomic sphere approximation (ASA), and neglected the impurity-induced lattice relaxations. These lattice relaxations are in fact long-ranged and lead to a volume change of the crystal. Many defect properties are influenced by these and a detailed understanding of these properties is still far from complete. In order to learn more about these effects in metals, in particular for very large impurity atoms, we present in this paper KKR calculations for impurities of the 5sp and 6sp series in BCC Fe. To these series belong atoms which have the largest atomic volumes of all elements in the periodic table. For these reasons also particularly large lattice relaxations are to be expected.

Our calculations are based on the density-functional theory with the exchange and correlation effects being treated in the local spin-density approximation (LSDA). The calculations are performed using a full-potential KKR Green's function method for defects. The method takes the advantage of a Green's function method to embed a cluster containing the impurity atom and a few disturbed neighboring host shells correctly in the ideal crystal, which is ideal for the case now studied, i.e., iron alloys in the dilute limit. Lattice statics methods are used to describe the longer ranged relaxations and to speed up the determination of the final positions of the atoms from the ab initio forces [10].

The theoretical LSDA lattice constant 5.204 a.u. and an angular momentum cutoff $\ell_{\max} = 4$ for the Green's function and for the radial functions are used. The potentials of five neighboring shells of Fe atoms are allowed to be perturbed, so that in total 59 perturbed potentials are recalculated self-consistently in the impurity iterations. The calculation of the hyperfine field is based on Breit's formula, which is the correct relativistic expression for hyperfine fields. We calculate the hyperfine fields by

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Table 1
Hyperfine fields (kG) of 5sp and 6sp impurities in Fe. The KKR–ASA values taken from Ref. [9] and the experimental ones from Refs. [1–4] are also shown

	Exper.	FP relax.	FP ideal	ASA
Cd	− 300	− 314	− 298	− 393
In	− 280	− 256	− 240	− 338
Sn	− 100	− 129	− 134	− 205
Sb	210	110	45	26
Te	700	475	330	355
I	1140	1223	917	780
Xe	1600	1345	1269	1276
Cs	270	796	1130	1603
Ba	− 100	− 104	− 62	38
La	− 460	− 347	− 405	− 426
Au	− 1300	− 978	− 914	− 1213
Hg	− 800	− 811	− 777	− 1010
Tl	− 200	− 528	− 558	− 740
Pb	280	− 7	− 204	− 229
Bi	1190	896	456	512
Po	2380	2182	1407	1462
At	2540	3648	2660	2605
Rn	1700	3940	3930	3858
Fr	949	990	2240	4771
Ra	− 130	− 807	− 773	− 284
Ac	− 300	− 1274	− 1452	− 1984

using the relativistic generalization [11] of the contact term and neglect the orbital contribution in line with the scalar relativistic approximation used in our calculations.

The impurity-induced lattice relaxations are found to be relatively small. The largest relative volume changes are about 90% of the elementary volume of Fe, which are really small compared to the huge atomic volumes of these impurities. This behavior is very different from the one found for 3d and 4d impurities in Cu, for which the volume changes scale reasonably well with the atomic volumes of the impurities introduced into the Cu host by substitution. In terms of elasticity language one could therefore classify the sp impurities as highly compressible.

The calculated hyperfine fields of the 5sp and 6sp substitutional impurities in bcc Fe are compared with experiments and earlier KKR–ASA calculations in Table 1. It is seen that both the full-potential and the ASA calculations give almost identical hyperfine fields at the beginning of the series up to the rare gas atoms. Contrary to this, at the beginning of each row in the periodic table the ASA trend differs considerably from the full-potential one. It can also be seen from Table 1 that the inclusion of lattice relaxations substantially improves the agreement with the experiment. This is particularly true for the sp-impurities in the first half of the series, where the hyperfine field is increased due to lattice relaxations. On the other hand, for the later sp-impurities the experimental hyperfine fields for the sp-impurities are not that well reproduced, especially in the 6sp series there are large discrepancies for At, Rn as well as for Ra and Ac.

In order to search for possible errors in the theoretical treatment it is important to realize that the LSDA is known to underestimate the lattice constant and thereby also the magnetic moment. On the other hand it is known that the generalized gradient approximations (GGA) give a much better description of these properties. Recently, Cottenier and Haas [12] have calculated hyperfine fields of 4d and 5sp impurities in BCC iron by using a GGA. Their values for hyperfine fields of the 5sp impurities are slightly larger than our results. We have also done LSDA calculations using the experimental lattice constant and thereby obtained a larger magnetic moment for the iron host [13]. These calculations showed a similar effect than the results by Cottenier and Haas, i.e., the absolute values of hyperfine fields increased only slightly. Thus, unfortunately we cannot pin down the origin for the large errors in the hyperfine fields of the late 6sp impurities.

In summary, we have performed first-principles full-potential calculations of the lattice relaxations and the hyperfine fields of heavy sp impurities in iron. The calculated hyperfine fields of 5sp and 6sp substitutional impurities reproduce the experimentally observed trend qualitatively well and also the quantitative agreement is good for the most of the impurities. The inclusion of lattice relaxations in the calculations was also seen to improve the overall agreement of the calculated results with experiments even though the relaxation effects are substantially smaller than we originally expected.

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