PHYSICAL REVIEW B VOLUME 25, NUMBER 5 1 MARCH 1982

Band theory for the magnetic moment in the bcc Fe-Co alloy

Karlheinz Schwarz
Institut für Technische Elektrochemie, Technische Universität Wien, A-1060 Vienna, Austria

Dennis R. Salahub
Département de Chimie, Université de Montréal, Montréal, Québec H3C 3V1, Canada

(Received 16 December 1981)

Magnetic moments and equilibrium lattice constants for the bcc Fe-Co alloy are obtained by means of band-structure calculations using the augmented-spherical-wave method and the local-spin-density scheme. The calculated magnetic moment in Fe-Co does not vary linearly with composition, in agreement with experiment. A rigid-band picture is found to be inadequate; i.e., chemical effects are very important.

Some iron-based alloys have unusual magnetic properties, as for example the fcc Fe-Ni (Invar) alloy[1] or the bcc Fe-Co alloy,[2] where in both cases the average magnetic moment as a function of composition possesses a maximum and deviates from the Slater-Pauling curve. The anomalous properties have often been given phenomenological interpretations by either local or itinerant-electron (band) models; however, until recently no first-principles calculations had been carried out on these alloys. At the present time two approaches are being tried in an effort to obtain a first-principles explanation of the anomalies. The first of these[3] involves molecular-orbital cluster calculations and the second,[4] *ab initio* energy-band calculations using spin-density-functional theory.

These first attempts have dealt with the classical Invar alloy, fcc Fe-Ni, which is unfortunately complicated somewhat by a structural change from bcc to fcc for a composition which is quite close to the important region near the maximum in the magnetic moment. No such complications arise for the interesting range of composition of Fe-Co, which from pure Fe to about 70 at.% Co is a substitutional alloy based on the bcc structure. If one neglects the effects of disorder, then band theory can be applied for selected compositions to study the magnetism of these alloys.

In the present Communication we present band-theory results for FeCo, which, besides being interesting in their own right, may ultimately have some bearing on the more general Invar problem. Results are reported for bcc Fe, the hypothetical bcc Co, and FeCo, the 50% alloy, for which we assume the ordered CsCl structure. The augmented-spherical-wave (ASW) method of Williams *et al.*[5] is employed in connection with the local-spin-density-functional approximation.[6,7] Similar calculations have previously succeeded in reproducing the observed magnetic moments and atomic volumes for pure metals such as bcc Fe.[8-10] Self-consistent ASW calculations are performed for several lattice constants and the equilibrium volume is determined by either minimizing the total energy or equivalently by finding the volume for which the electronic pressure vanishes.

Figure 1 shows the experimental data for the Fe-

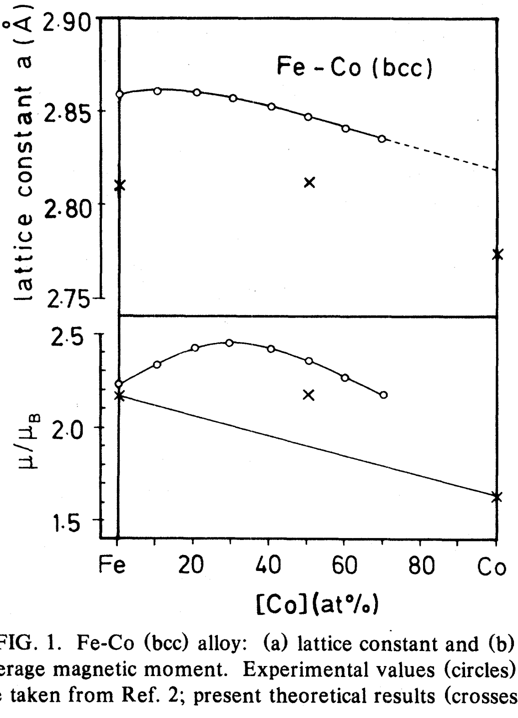

FIG. 1. Fe-Co (bcc) alloy: (a) lattice constant and (b) average magnetic moment. Experimental values (circles) are taken from Ref. 2; present theoretical results (crosses).

Co alloy[1] in comparison with the theoretical results of the present work: (i) The variation of the lattice constant with composition as given by experiment[1] is well represented in trend by the three theoretical values, but the latter are systematically smaller than experiment (by about 2%); (ii) the average magnetic moment μ obtained at the theoretical equilibrium volume does not follow a linear variation with composition; for the 50% alloy both theory and experiment yield a value of μ which is comparable to that of pure Fe and is significantly above a straight line connecting μ_{Fe} with μ_{Co}.

In the ASW method the atomic-sphere approximation is used, which allows the definition of local magnetic moments as the difference of the integrated spin densities inside each atomic sphere. In the present calculation for FeCo, where we have used equal sphere sizes for Fe and Co, we find the local magnetic moment remains about constant for Co (comparing the FeCo result with that for bcc Co), whereas it increases from about $2.2\mu_B$ for pure Fe to about $2.7\mu_B$ for FeCo in accord with neutron-diffraction data.[11]

The local moments assigned to each atom are of course derived from an itinerant-electron picture. Although the energy bands describe delocalized Bloch states, the density of states (DOS) may still be decomposed into local contributions by weighting each energy state with the spin density localized inside the corresponding atomic sphere. Although this decomposition depends to a certain extent on the choice of the atomic sphere radii, the local-spin DOS provide a very useful means of exhibiting, and analyzing important chemical effects; for example, in the transition-metal carbides, nitrides, and oxides[12] where rigid-band models fail. For an understanding of magnetic alloys it is of special interest to investigate to what extent a rigid-band picture is adequate.

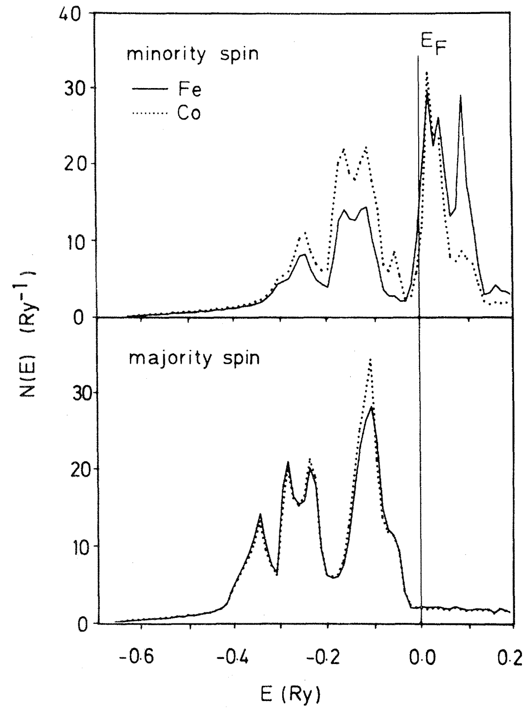

FIG. 2. Local density of states (DOS) per atom and spin of FeCo.

The local-spin DOS of FeCO are shown in Fig. 2. For majority spin (↑) the Fe and the Co DOS are very similar to each other and they both look like a typical bcc DOS.[8] For minority spin (↓), however, the situation is drastically different, since they are not just shifted to higher energies (as is approximately the case for the pure elements), but show additional peaks; one in the Co DOS (↓) below E_F, and one pronounced peak in the Fe DOS (↓) at the top of the band. Furthermore, there is a significant reduction of the Fe DOS (↓) in the lower-energy region of the *d* band in contrast to the Co DOS (↓) which remains comparable in magnitude to the majority spin Co DOS (↑). This last observation is responsible for the fact that the local Co magnetic moment is about the same as in pure bcc Co, whereas the local Fe moment increases to about $2.7\mu_B$. As will be discussed in detail elsewhere,[13] the general features of Fig. 2 can be understood by realizing that Co energy levels are intrinsically more stable than those of Fe (higher electronegativity of Co), whereas the exchange splitting is expected to be larger for Fe. For majority spin these two factors tend to compensate, whereas for minority spin they reinforce each other. Figure 2 shows clearly that a rigid-band model derived from just the bcc structure (of pure Fe for example) yields an inadequate description of FeCo; i.e., chemical bonding effects play an important role. It should be noted that the two extra peaks in the minority spin DOS (↓) are also present in a non-spin-polarized calculation of FeCo.

In summary, the present spin-polarized energy-band calculation have shown the following: (i) the average magnetic moment in the Fe-Co alloys does not vary linearly with composition; (ii) the deviation from linearity is caused by a significant increase of the local Fe moment while the Co moment remains about constant; (iii) this behavior is explained in terms of local spin DOS from which the local moments are derived within an itinerant electron picture; (iv) FeCo deviates strongly from a rigid-band model, since the minority DOS differs qualitatively from the majority DOS, whereas the latter agrees with a typical DOS of a bcc lattice.

Extensions of these ideas to other systems and additional investigations by different theoretical approaches are currently being performed.

ACKNOWLEDGMENTS

We wish to thank Dr. A. R. Williams and Dr. J. Kübler for valuable discussions and for providing us with their ASW program, which was adapted for the present work. We are also grateful to Dr. J. Kaspar and Dr. B. N. McMaster for helpful discussions and to the Natural Sciences and Engineering Research Council of Canada and the Österreichische Forschungsgemeinschaft for financial support.

---

[1] Y. Nakamura, IEEE Trans. Magn. 12, 278 (1976).

[2] M. Shiga, in *Physics of Transition Metals—1980,* edited by P. Rhodes, Inst. Phys. Conf. Ser. No. 55 (IOP, London, 1980), p. 241.

[3] J. Kaspar and D. R. Salahub, Phys. Rev. Lett. 47, 54 (1981).

[4] A. R. Williams, V. L. Moruzzi, C. D. Gelatt, J. Kübler, and K. Schwarz, J. Appl. Phys. (in press).

[5] A. R. Williams, J. Kübler, and C. D. Gelatt, Jr., Phys. Rev. B 19, 6094 (1979).

[6] U. von Barth and L. Hedin, J. Phys. C 5, 1629 (1972).

[7] J. F. Janak, Solid State Commun. 25, 53 (1978).

[8] V. L. Moruzzi, J. F. Janak, and A. R. Williams, *Calculated Electronic Properties of Metals* (Pergamon, New York, 1978).

[9] J. Kübler, Phys. Lett. 79A, 210 (1980).

[10] J. Kübler, Phys. Lett. 81A, 81 (1981).

[11] M. F. Collins and J. B. Forsyth, Philos. Mag. 8, 401 (1963).

[12] A. Neckel, P. Rastl, R. Eibler, P. Weinberger, and K. Schwarz, J. Phys. C 9, 579 (1976).

[13] D. R. Salahub, J. Kaspar, and K. Schwarz (unpublished).