Magnetism in bcc cobalt

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Local-spin-density-approximation-based calculations, performed using a general-potential linearized augmented-plane-wave method, are presented for bcc Co. The ground-state properties and magnetization energies are reported. It is found that the moment is strongly suppressed in constrained antiferromagnetic calculations, indicating that a local-moment picture is less appropriate for this material than for bcc iron.

I. INTRODUCTION

The growth of single-crystal films of bcc Co on GaAs substrates[1,2] has provided a very interesting transition metal ferromagnet for study. Further, growth of bcc Co has been reported on Cr (Ref. 3) and Fe (Ref. 4) substrates, making possible studies of interface magnetism between bcc Co and other magnetic materials.[5] Photoemission experiments[6] and local-spin-density-approximation (LSDA) band-structure calculations[6-11] both indicate that the electronic structure of bcc Co is similar to that of bcc Fe, with a rigid shift corresponding to the addition of one electron. Significantly, like Fe, but unlike Ni, bcc Co has a substantial exchange splitting of approximately 1.6 eV.[6,12] However, unlike Fe, the majority-spin d bands in bcc Co are fully occupied. In bcc Fe-Co alloys, the Co moment remains nearly fixed at $1.7\mu_B$ over the composition range for which the bcc structure is stable, while the Fe moment varies from $2.2\mu_B$ to approximately $3\mu_B$ as the Co concentration is increased.[7,13,14] Other evidence for the stability of the Co magnetic moments comes from Brillouin scattering experiments.[15] Although magnetization measurements for bcc Co films grown on GaAs were taken as an indication that the Co moment in pure bcc Co is significantly smaller ($1.4\mu_B$–$1.5\mu_B$) than in Fe-Co alloys, hcp Co, and LSDA calculations for bcc Co, recent polarized neutron experiments have shown that this discrepancy could be a result of the diffusion of substrate As atoms into the interface region of the films.[16]

Transition-metal magnetism is normally treated in terms of an itinerant Stoner model. On the other hand, magnetism in more strongly correlated materials (characterized by a large ratio of the intra-atomic Coulomb integral to the bandwidth, U/W), such as the $4f$ band materials, is normally treated as a system of interacting local moments. Further, even in Fe, moments are known to persist well above the Curie temperature.[17] The position of Co on the right side of the $3d$ series leads to the possibility of relatively strong correlations for a transition metal. While estimates of values of U for transition metals vary considerably depending on the approach used, it is generally thought that hcp Co is more strongly correlated than bcc Fe and perhaps as strongly correlated as fcc Ni.[18-20] The bandwidth W of bcc Co (Refs. 5 and 6) is approximately 1.5 eV smaller than that of hcp Co,[21] so that bcc Co may be the most strongly correlated ferromagnetic transition metal.

The relatively large value of U/W, and the experimentally observed stability of the Co moments in bcc Co, raises the question of to what extent a local moment picture of the magnetism in this material might be useful. The strongest evidence for the utility of such a picture comes from the Brillouin scattering study of Karanikas *et al.*,[15] in which the exchange stiffness D of bcc Co was compared with that of hcp Co. As noted in Ref. 15, the bcc and hcp phases of Co have similar nearest-neighbor distances (2.44 and 2.50 Å, respectively) but different coordination numbers, z, of 8 and 12, respectively. Karanikas *et al.* found that D for hcp and bcc Co scales with z. This is a characteristic of a local-moment system. On the other hand, it is known that certain properties of itinerant ferromagnets, including the exchange stiffness measured by low-momentum probes (such as Brillouin scattering), may be well described by a fictitious local-moment Heisenberg Hamiltonian.[22]

Here, highly converged, full potential LSDA-based calculations of the ground-state properties and electronic structure of bcc Co as well as calculations with constraints on the spin density are presented. I note that these are full-potential total-energy calculations for bulk bcc Co (in Ref. 11, full potential calculations are reported for the electronic structure of thin bcc films). The constrained calculations were performed in order to address the question of the stability of the Co moments in bcc Co and the utility of a local-moment picture of this material. The particular calculations performed were for a constrained paramagnetic (zero-spin) system and for an antiferromagnetic spin configuration. In order to provide a reference, parallel calculations were performed for bcc Fe. In addition, the spin susceptibility of ferromagnetic bcc Co, for fields aligned with the magnetization, is reported.

II. METHOD

The calculations reported here were performed using the general-potential (i.e., no shape approximations to the

charge densities or potentials) linearized augmented-plane-wave (LAPW) method.[23] This technique has been discussed in detail elsewhere and accordingly only details particular to the present calculation are given here. The calculations were performed fully self-consistently, with relativistic core states and scalar relativistic valence states, using the von Barth–Hedin local exchange correlation (xc) function, parametrized to reproduce the Hedin-Lundqvist xc function for a non-spin-polarized system.[24] The Brillouin-zone samplings were performed using a set of 240 special **k** points[25] in the irreducible wedge of the Brillouin zone for the bcc unit cells, and 120 special **k** points for the two-atom simple-cubic cells. The electronic density of states (DOS) was calculated using a uniform mesh of 506 **k** points in the irreducible wedge. A sphere radius of $R_{MT}=2.15$ a.u. was used for both Fe and Co. For Co, a basis set cutoff of $R_{MT}K_{max}=9.5$ was used yielding approximately 110 LAPW basis functions per atom at the equilibrium lattice constant. For Fe, convergence tests indicated that a smaller cutoff, $R_{MT}K_{max}=9.0$, which yielded approximately 95 basis functions per atom, was adequate. Test calculations were also performed using an extension of the LAPW method which reduces any residual errors due to linearization of the d bands.[26] This test indicated that the error due to linearization was not significant. The static properties (lattice parameter and bulk modulus) were determined by fitting the Murnaghan equation of state[27] to calculated total energies for a range of lattice parameters between 5.0 and 5.4 a.u. The spin susceptibility for fields parallel to the magnetization, χ, was obtained through self-consistent calculations of the magnetic moment as a function of the applied field. Calculations were performed for nine field values between 0 and 17.5 mRy.

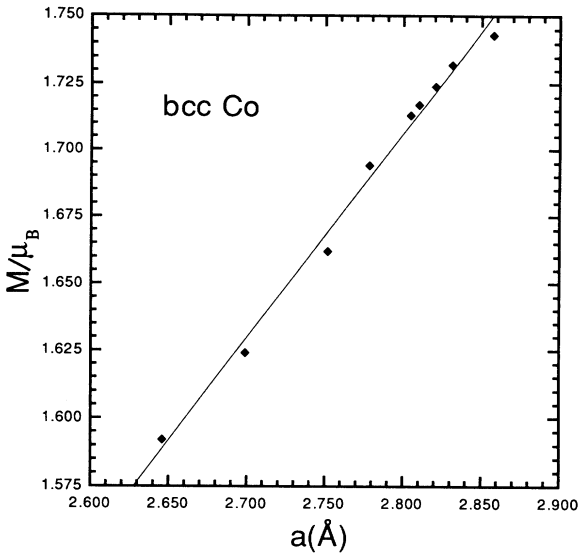

FIG. 1. Spin magnetization per atom, M, of bcc Co as a function of the lattice parameter **a**. The solid symbols are calculated values, while the curve is a quadratic fit to the calculated points.

III. RESULTS AND DISCUSSION

A. Ferromagnetic bcc Co

The principal results of our calculations for ferromagnetic bcc Co are given in Table I, where the ground-state properties and spin susceptibility are reported and compared with experiment and previous theoretical calculations where available, in Fig. 1, where the variation of the spin moment with atomic volume is shown and in Fig. 2, where the DOS is given. As may be noted from Table I, the calculated lattice parameter **a**=2.76 Å is 2% smaller than the value obtained by extrapolating room-temperature data for bcc Fe-Co alloys to pure bcc Co.[1] This sort of discrepancy is typical when well-converged LSDA calculations for transition metals are compared with experiment and is significantly smaller than the 3.5% error found for bcc Fe.[29] Previous calculations,[9–11] which employed various spherical approximations, yielded somewhat larger lattice parameters in better agreement with experiment.

The calculated spin magnetization, $M=1.726\mu_B$/atom, at **a**=2.82 Å is in reasonable agreement with previous calculations which yielded values ranging from $1.65\mu_B$/atom to $1.73\mu_B$/atom. As mentioned, until quite recently experiments for bcc Co films grown on GaAs substrates yielded significantly smaller magnetizations. Based on the disagreement of the experimental value with LSDA calculations, Moruzzi *et al.* suggested that the moment in the film was probably being suppressed by de-

TABLE I. Calculated properties of ferromagnetic bcc Co. Here **a** is the lattice parameter, B is the bulk modulus, M is the spin magnetization per atom, and χ is the spin susceptibility for fields in the direction of the magnetization. M and χ were calculated at a lattice parameter of 2.82 Å (derived from extrapolation of the lattice parameters of Fe-Co alloys) (Ref. 1). Expt. denotes experimental values.

	a (Å)	B (GPa)	M/μ_B	χ (μ_B/Ry)
This study	2.76	278	1.726	8.4
Expt. (GaAs substrate)	2.82[a]		1.3–1.7[b]	
Min *et al.*, LMTO[c]	2.82	245	1.73	
Hermann *et al.*, LMTO[d]			1.72	
Schwarz *et al.*, ASW[e]	2.773		1.65	
Moruzzi *et al.*, ASW[f]	2.81		1.68	

[a]Reference 2.
[b]References 15, 16, and 28.
[c]Reference 11.
[d]Reference 9.
[e]Reference 8.
[f]Reference 10.

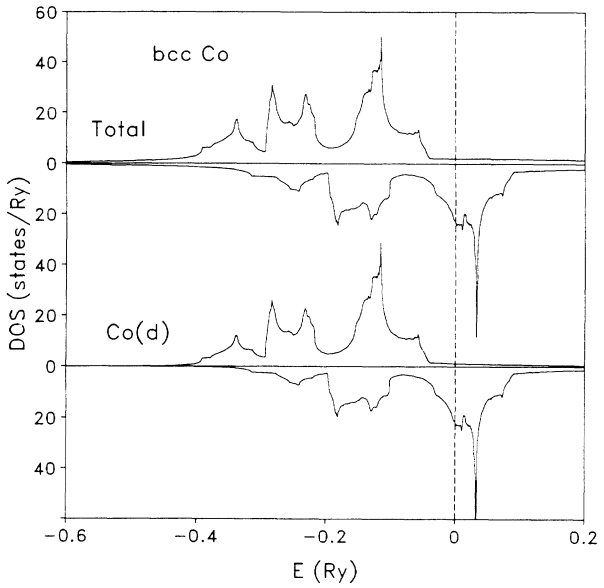

FIG. 2. Electronic DOS of bcc Co at $a=2.82$ Å. The top panel shows the majority- (positive-) and minority-spin DOS. The lower panel shows the corresponding components of the DOS projected onto the d orbitals in the spheres. The Fermi energy is denoted by the dashed vertical line.

fects.[10] Recently, Bland *et al.* have reported polarized neutron data, which can be reasonably fit by assuming a magnetization profile in which the Co moments are suppressed near the interface with the substrate and reach a maximum value of $1.7\mu_B$/atom away from the interface, leading to the conclusion that the discrepancy between the measured and calculated magnetizations could be due to As impurities in the bcc Co films.[16] Similar effects have been observed in thin bcc Fe films grown on GaAs.[15]

As shown in Fig, 1, the variation of the Co moments with volume is predicted to be small. Similarly, the calculated variation in the magnetization with applied field is relatively weak. This can be understood in terms of the DOS (Fig. 2), which shows that majority-spin d bands of bcc Co are fully occupied with only a small majority-spin DOS at the Fermi level. (The calculated majority- and minority-spin DOS are 2.1 and 22.7 states/Ry, respectively.) Because the majority-spin d bands occur well below the Fermi energy, perturbations which affect the bandwidths or exchange splitting, such as magnetic fields or changes in the lattice parameter, will need to be quite strong to significantly change the occupation of the majority-spin d orbitals. Provided that the majority-spin d states remain below the Fermi level, changes in the spin moment are inhibited by the requirement of charge neutrality. On the other hand, as is often found in calculations for nonequilibrium structures, the Fermi energy lies near a peak in the total DOS. This provides an indication that charge transfer may be energetically favorable in alloys based on bcc Co, and this may affect the moments. It is possible that charge transfer between Co and As impurities is important in producing the lower moments measured in films grown on GaAs substrates. Calculations investigating this possibility are in progress.

B. Magnetic energies and local moments

As mentioned, in order to assess the applicability of a local-moment picture for bcc Co, constrained local-spin-density calculations have been performed. The characteristic of a system which is describable by a local-moment picture is that the moments are approximately independent of the spin configuration. In order to investigate this, self-consistent calculations were performed for two spin configurations at the equilibrium lattice parameter. These are ferromagnetic bcc Co and an antiferromagnetic spin configuration in which nearest-neighbor moments are constrained to be opposite. In order to facilitate comparison of the results of these two calculations, they were both performed in the same simple-cubic unit cell. The antiferromagnetic constraint was imposed using a suggestion of Mehl,[30] by calculating wave functions for one spin only and constructing the wave functions for the other spin using the assumed antiferromagnetic symmetry. In order to investigate the energetics, a non-spin-polarized calculation was also performed. Parallel calculations were performed for Fe. The results of these calculations are given in Table II. It is interesting that in both bcc Fe and bcc Co the antiferromagnetic configuration is lower in energy than the non-spin-polarized paramagnetic system.

In the case of Fe, a substantial spin moment is found to persist in the antiferromagnetic calculation. Forcing the antiferromagnetic symmetry leads to a substantial increase of 30 mRy/atom in the total energy, but a reduction in the moment of only $0.75\mu_B$ or approximately 35%. This is in qualitative agreement with the results of Luchini and Heine[17] who, using a different approach, obtained a suppression of the moment of approximately 45% and an energy increase of 25 mRy/atom for the antiferromagnetic ($1-\cos\theta=2$, in their notation) configuration.

TABLE II. Magnetic energy, E_{mag}, and moment, μ_s, of bcc Co at a lattice parameter of 2.82 Å and of bcc Fe at its equilibrium lattice parameter of 2.86 Å. μ_s is the spin moment in the Co or Fe spheres. This differs from M (see Table I) which is the total spin moment per unit cell.

	bcc Co		bcc Fe	
	E_{mag} (mRy/atom)	μ_s/μ_B	E_{mag} (mRy/atom)	μ_s/μ_B
Paramagnetic	0	0	0	0
Ferromagnetic	−23.0	1.78	−33.6	2.19
Antiferromagnetic	−3.2	0.82	−3.7	1.44

The present calculations indicate that the moments in bcc Co are substantially less robust. Although, like Fe, imposing the antiferromagnetic spin configuration involves a substantial energy cost (20 mRy/atom in this case), in bcc Co there is a considerably larger suppression of the moment. In particular, the spin moment for the antiferromagnetic configuration is found to be $0.82\mu_B$ or approximately 54% smaller than for the ferromagnetic configuration. This difference (54% for bcc Co versus 35% for Fe) shows that even though U/W in bcc Co is larger than in Fe, bcc Co is considerably less well described by a local-moment picture than Fe.

IV. CONCLUSIONS

Well-converged general-potential LAPW calculations have been presented for bcc Co, with both ferromagnetic and antiferromagnetic spin configurations. For the ferromagnetic configuration, results in reasonable agreement with experiment are obtained, with the possible exception of the spin moment. The DOS indicates that the spin moment may be significantly changed in bcc Co-based alloys in which there is the possibility of charge transfer. The suppression of the moments for the antiferromagnetic configuration is substantially larger than in Fe, showing that a local-moment picture is not likely to provide a good description of the magnetism in bcc Co.

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