Magnetic structures of fcc systems with nearest-neighbor and next-nearest-neighbor exchange interactions

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The nature of the relationship of the magnetic structure in fcc systems to the values of J_1 and J_2, the nearest-neighbor and next-nearest-neighbor exchange interactions respectively, is examined. Fifteen experimental systems, with ordering varying from ferromagnetism to three different kinds of antiferromagnetism (AFI, AFII, AFIII), fit the predictions of the ter Haar–Lines model. Several other fcc systems where the exchange interactions need to be known more accurately for comparison with the model are noted.

Systems in which magnetic ions occupy sites on a face-centered cubic (fcc) lattice are predicted to have four different kinds of magnetic ordering depending on the relative signs and strengths of J_1 and J_2, respectively the nearest-neighbor (NN) and next-nearest-neighbor (NNN) exchange constants.[1] For $J_1 > 0$ and $J_2 = 0$, the fcc lattice is naturally frustrated in that all exchange couplings cannot be simultaneously satisfied unless some anisotropy is present.[1] For other values of J_2/J_1, three antiferromagnetic (AF) orders, viz. AFI, AFII, and AFIII and a ferromagnetic order F, are possible.[1-3] If exchange interactions among more distance neighbors are appreciable, then additional modifications in the magnetic structures can occur.[1]

By now, a number of experimental fcc systems have been discovered whose magnetic structures and exchange constants are known to a reliable degree of accuracy. The main purpose of this paper is to examine how many of these fcc systems in which J_1 and J_2 are known reasonably accurately from combined magnetic and neutron scattering experiments and where more distance exchange couplings are negligible fit the theoretical predictions. Such an analysis might provide valuable information on how transitions between different magnetic structures may occur as J_1 and J_2 are varied between different systems or because of magnetic doping or change of temperature in the same system.

In Table I, we have listed 15 systems in which magnetic ions occupy the fcc lattice and in which J_1 and J_2 are the dominant exchange interactions. The sign convention used is one in which an exchange interaction is positive for antiferromagnetic interaction and negative for ferromagnetic interaction. In Fig. 1, we have shown the predictions of the theory[1-3] as boundaries (bold lines) between different magnetic structures in the J_1J_2 plane and the positions of the various systems of Table I (shown as dotted lines). Some comments on the observations of Fig. 1 follow.

For the binary transition-metal oxides and a sulfide viz. MnO, FeO, CoO, NiO, and α-MnS, antiferromagnetic order of second kind, AFII, is well established. Since none of these systems are close to the boundaries of the neighboring structures of AFIII and F, the AFII ordering in these systems is apparently quite stable. The series EuO, EuS, EuSe, and EuTe provides a more interesting case in that ordering varies from F to AFII. EuO and EuS are stable ferromagnets, consistent with Fig. 1, since their relative positions are well away from the phase boundaries. Similarly EuTe has a stable AFII structure. On the other hand, for EuSe, the ratio J_2/J_1 is such that it is located close to the boundary between F and AFII phases. Experiments indeed show[4] that EuSe is a metamagnet; it orders in the AF state at 4.6 K but changes to ferromagnetism at 2.8 K. Thus presentation of Fig. 1 has provided insight as to why EuSe has relatively unstable magnetic structures and why the remaining compounds discussed above have stable magnetic structures.

Ordering of the third kind, AFIII, has been observed in β-MnS (Ref. 5) and in diluted magnetic semiconductors such as $M_{1-p}Mn_pTe$ (M=Cd,Zn).[3] In β-MnS, the exchange constant J_2 is not known accurately although $J_2/J_1 = -0.1$ is generally accepted.[6] These materials fall close to the boundaries of AFI (see Fig. 1). However from neutron-diffraction measurements, there is no evidence so far that magnetic ordering is other than AFIII.[3] For the diluted magnetic semiconductors, a recent paper[7] has given a revised listing of the exchange constants. These revised values of J_1 and J_2 are still consistent with the AFIII structure (Fig. 1) for those diluted magnetic semiconductors which have the zinc-blende structure so that the magnetic ions (Mn^{2+}) occupy the fcc lattice.

Magnetic ordering of type AFI has been reported in a number of neodymium systems viz. NdP, NdAs, NdBi, and NdSb.[8] From the known values of J_1 and J_2 these materials fall neatly in line with the predictions of Fig. 1.

There are several other known systems in which magnetic ions occupy fcc lattice. However, in these cases either J_1 and J_2 are not known accurately or more distant neighbor exchange couplings are also significant, making it difficult to include them in Fig. 1 at this time. For ex-

TABLE I. Magnetic constants and magnetic structures of some fcc systems.

System	T_N (K)	T_c (K)	J_1 (K)	J_2 (K)	Type	References
MnO	118		10.0	11.0	AFII	a
α-MnS	152		7.0	12.7	AFII	b
$Fe_{0.95}O$	192		−1.7	16.0	AFII	c
CoO	289		5.5	27.4	AFII	d
NiO	523		34,16	202	AFII	e
EuO		69.4	−0.61	−0.12	*F*	f
EuS		16.5	−0.24	0.12	*F*	f
EuSe	4.6	2.8	−0.11	0.09	AFII,*F*	4
EuTe	9.8		−0.06	0.20	AFII	4
NdP	11		0.01	−0.11	AFI	10
NdAs	13		0.03	−0.10	AFI	10
NdSb	16		0.07	−0.11	AFI	10
NdBi	24		0.09	−0.18	AFI	10
β-MnS	98		28	2.8	AFIII	g,3
$Cd_{0.35}Mn_{0.65}Te$	36		14	1.4	AFIII	3

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ample, in K_2ReBr_6 and K_2ReCl_6 (AFI ordering) and K_2IrCl_6 (AFIII ordering),[9] J_1 and J_2 are not known accurately. The systems, MnS_2, $MnSe_2$, and $MnTe_2$ represent a class of pseudo-fcc systems (pyrite structure) where the magnetic structures are known accurately[10] (MnS_2 has AFIII structure, $MnTe_2$ has AFI structure and $MnSe_2$ has an arrangement intermediate between the two). However, J_1 and J_2 determined from the fit of the Curie-Weiss law of the magnetic susceptibility give values which are not consistent with the prediction of Fig. 1.[11] It was concluded[11] that more distant-neighbor exchange interactions are perhaps important making the predictions

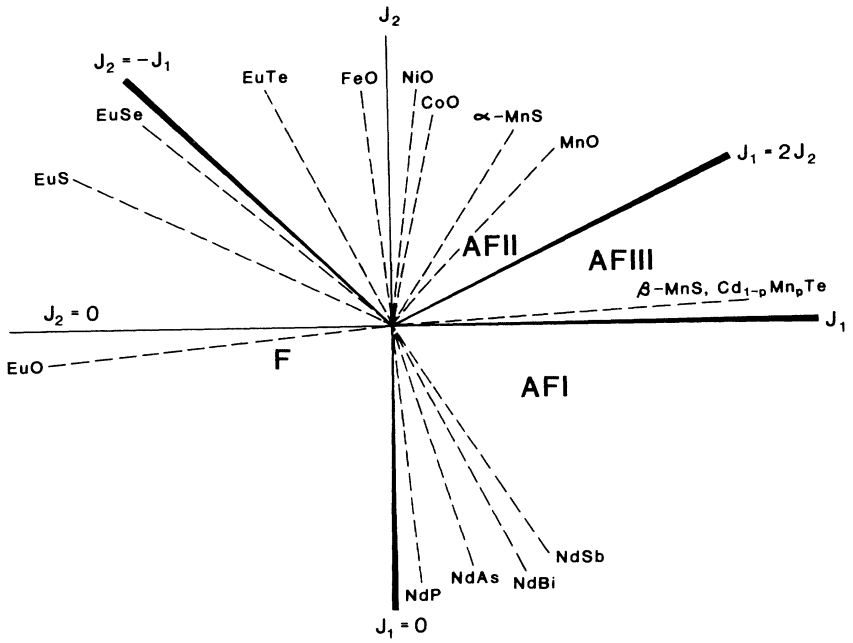

FIG. 1. Phase boundaries of the different magnetic structures of fcc systems in the J_1J_2 plane, where J_1 and J_2 are the NN and NNN exchange constants, respectively. Location of the various systems are shown as dotted lines using the data from Table I.

of the theory[1-3] invalid. However it is also well known that molecular-field treatment of the magnetic susceptibility does not always yield accurate values for the exchange constants. In any case, additional work is needed in these cases to test the validity of the model.

Another way to affect changes in J_1 and J_2 is to change the lattice constant by magnetic doping. Alternately if there is no significant change in the lattice constant on doping, then J_1 and J_2 are not expected to change and hence any changes in the measured properties are then due to other factors. Such a case is represented by the randomly diluted magnetic systems $Co_pMg_{1-p}O$ (Ref. 12) and $Eu_pSr_{1-p}Te$ (Ref. 13) where from Fig. 1, it is evident that both CoO and EuTe have AF II order. Here the lattice constant changes between $p=0$ and $p=1$ are less than 1% and the systems retain the fcc structure. A comparison between the magnetic phase diagrams of these two systems has been reported recently.[14] For $0.45 < p < 1$, the variations of the reduced Néel temperature $t \equiv T_N(p)/T_N(1)$ against p for the two systems coincide. However, for $p < 0.45$, t vs p variations for the two systems are different. These differences then must be due to factors other than the ratio J_2/J_1, e.g., differences in anisotropy or random anisotropy introduced by dilution. Further progress on understanding this difference between the two systems can perhaps be made if theoretical variation of t vs p for a fcc system, with both J_1 and J_2 included, becomes available.

In summary, the 15 fcc systems of Table I, where J_1 and J_2 and the corresponding magnetic structures are known accurately, fit well the phase diagram in the J_1J_2 plane of Fig. 1. Perhaps other systems can be examined in a similar way as their exchange constants and magnetic structures become known more accurately.

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