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The formation of localized moments in dilute alloys: a critical behaviour

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Abstract. The formation of localized moments of impurities in simple-metal hosts is investigated in the case of an Mn solute atom in jellia with continuously varying density. The mean-field critical behaviour of the transition from a spin-polarized to a non-spin-polarized state of the impurity, which is deduced from self-consistent local-spin-density (LSD) functional calculations, is analysed assuming a Landau-type expansion of the energy. This analysis is illustrated and supported by constrained LSD functional calculations.

The presence or absence of spin-polarization in itinerant-electron systems is determined by a competition between intra-atomic exchange interactions and interatomic electron motion. The nature of the transition from a spin-polarized to a non-spin-polarized state in such systems has been studied by different authors.

Self-consistent calculations by Stefanou *et al* [1] of the magnetic properties of a single Co impurity in Cu reported that in the vicinity of the transition point there is a square-root dependence of the Co moment on the local lattice relaxation. This behaviour is very similar to the result of the Landau theory for second-order phase transitions as implied by the mean-field character for the LSD functional theory [2, 3] which was employed in the calculations. This result, however, seems to hold also beyond the framework of a mean-field theory, since consideration of quantum fluctuations in the description of the critical behaviour of a three-dimensional system leads to mean-field critical exponents at zero temperature [4].

Moruzzi [5] investigated the transition from magnetic to non-magnetic behaviour in elemental ferromagnets under lattice compression. By examining the simplest possible ways that a system can undergo such a transition he showed that the magnetic moment versus volume curve always exhibits square-root singularities and that it is usually multivalued and discontinuous. These conclusions were also supported by self-consistent LSD calculations. In this case also a Landau-type expansion of the energy can lead to the prediction of the critical behaviour of the magnetic moment.

Recently, Stefanou *et al* [6] studied the formation of local spin-moments of impurities diluted in simple-metal hosts, by means of self-consistent LSD calculations using both the first-principles Korringa–Kohn–Rostoker (KKR) Green function method and the jellium model. In the latter approach, the jellium density $\bar{\rho}$ was varied continuously from a dilute limit corresponding to a Cs host up to the critical value $\bar{\rho}_c$, where the magnetic moment disappears. In the vicinity of the transition from a spin-polarized to a non-spin-polarized impurity state, it was found that the moment varies proportionally to $(1 - \bar{\rho}/\bar{\rho}_c)^{1/2}$ as it is demonstrated here for the specific case of an Mn substitutional impurity.

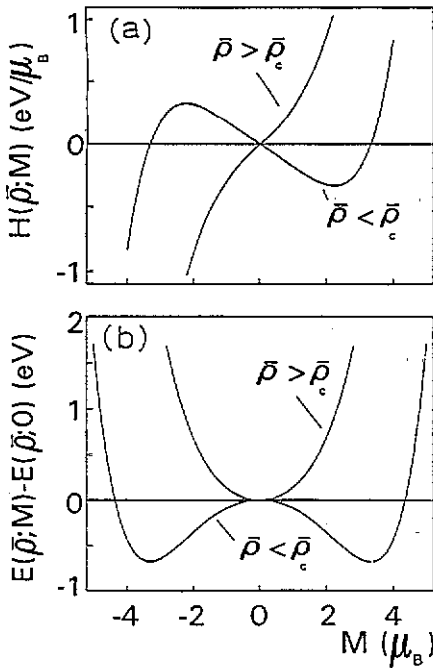


Figure 1. (a) Constraining magnetic field $H(\bar{\rho}; M)$ and (b) resulting energy difference $E(\bar{\rho}; M) - E(\bar{\rho}; 0)$ versus the magnetic moment of an Mn substitutional impurity in a jellium of density $\bar{\rho} = 29.84 \times 10^{-3} \text{ au}^{-3}$ ($\bar{\rho} > \bar{\rho}_c$) and $\bar{\rho} = 12.97 \times 10^{-3} \text{ au}^{-3}$ ($\bar{\rho} < \bar{\rho}_c$). For the Mn impurity $\bar{\rho}_c = 21.34 \times 10^{-3} \text{ au}^{-3}$.

Within the framework of density-functional theory the ground state of a many-electron system is obtained by solving self-consistently the one-particle Kohn–Sham equations. These equations are deduced by minimization of the total-energy functional $E[\rho^\uparrow, \rho^\downarrow]$, subject to particle conservation [2]. In our case we consider the lowest-energy state of an Mn-in-jellium system, constrained to have a given impurity magnetic moment M . This can be obtained by minimization of the generalized functional [7]

$$E[\rho^\uparrow, \rho^\downarrow] - H \left(\int_V d^3r m(\mathbf{r}) - M \right) \quad (1)$$

where H is a Lagrangian multiplier having the dimensions of a magnetic field and acts within the volume V of the system. The impurity magnetic moment M and magnetization density $m(\mathbf{r})$ are defined with respect to a pure jellium without impurity, uniformly polarized by the field H . Here we use for $E[\rho^\uparrow, \rho^\downarrow]$ the LSD functional of von Barth and Hedin [3] with the parametrization proposed by Janak [8]. Minimization of the energy functional (1) leads to the usual one-particle equations, where the term $H\mu_B$ is subtracted (added) in the expression of the effective potential for the spin up (down) states respectively. The lowest energy in this case is

$$E(\bar{\rho}; M) = \min \left[E[\rho^\uparrow, \rho^\downarrow] - H \left(\int_V d^3r m(\mathbf{r}) - M \right) \right]. \quad (2)$$

By taking advantage of the Hellmann–Feynman theorem

$$\partial E(\bar{\rho}; M) / \partial M = H \quad (3)$$

we can calculate the energy difference

$$E(\bar{\rho}; M_2) - E(\bar{\rho}; M_1) = \int_{M_1}^{M_2} dM H(\bar{\rho}; M). \quad (4)$$

We calculate as a function of H the corresponding $M = \int_V d^3r m(r)$ solving self-consistently the one-particle equations deduced from minimization of the energy functional (1) and obtain in this way $H(\bar{\rho}; M)$. Our results for an Mn impurity in a jellium with density $\bar{\rho} = 29.84 \times 10^{-3} \text{ au}^{-3} > \bar{\rho}_c$ and $\bar{\rho} = 12.97 \times 10^{-3} \text{ au}^{-3} < \bar{\rho}_c$ ($\bar{\rho}_c = 21.34 \times 10^{-3} \text{ au}^{-3}$ for the Mn impurity) are shown in figure 1(a). The lower value of $\bar{\rho}$ corresponds to the free-electron density of the Cu host (the $\bar{\rho}$ values for some other simple monovalent metals are: 8.50(Ag), 7.57(Li), 4.39(Na), 2.37(K), 1.91(Rb), 1.43(Cs), in 10^{-3} au^{-3}). In figure 1(b) we show the corresponding energy curves $E(\bar{\rho}; M)$ calculated using (4). It is clear that for $\bar{\rho} > \bar{\rho}_c$ the stable solution is the non-polarized one, whereas for $\bar{\rho} < \bar{\rho}_c$ the minimum energy is obtained for two equivalent spin-polarized solutions, with magnetic moments $\pm \bar{M}$. In the latter case the magnetic solution lies about 0.7 eV below the non-magnetic one.

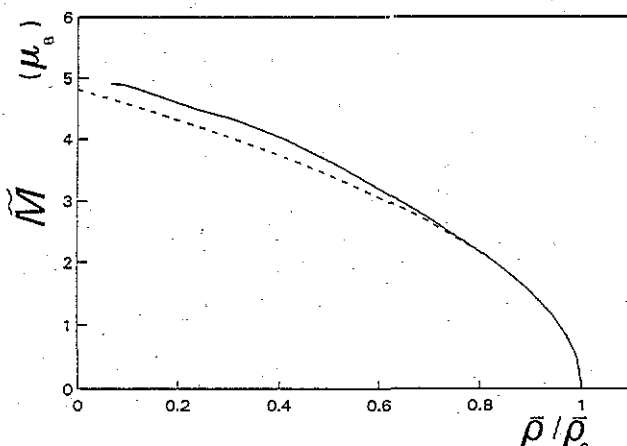


Figure 2. Ground state magnetic moment of an Mn substitutional impurity in jellium as a function of the jellium density (solid curve). The dashed curve shows the result of (7).

In the vicinity of the critical density we can expand $E(\bar{\rho}; M)$ in the manner of the Landau

$$E(\bar{\rho}; M) = E(\bar{\rho}; 0) + \alpha(\bar{\rho})M^2 + \beta(\bar{\rho})M^4 + \dots \quad (5)$$

where only even powers of M occur, since the energy does not depend on the direction of the magnetic moment. With truncation of the expansion (5) at fourth order, the minimum energy condition for a given $\bar{\rho}$ yields the ground state impurity moment

$$\bar{M} = \{-\alpha(\bar{\rho})/2\beta(\bar{\rho})\}^{1/2} \quad \bar{\rho} < \bar{\rho}_c \quad (6a)$$

$$\bar{M} = 0 \quad \bar{\rho} > \bar{\rho}_c. \quad (6b)$$

The coefficient $\alpha(\bar{\rho})$ is a measure of the curvature $\partial E(\bar{\rho}; M)/\partial M$ at $M = 0$ which is negative for $\bar{\rho} < \bar{\rho}_c$ and positive for $\bar{\rho} > \bar{\rho}_c$ (see figure 1(b)). Therefore, due to continuity $\alpha(\bar{\rho}_c) = 0$. In the vicinity of $\bar{\rho}_c$, taking $\alpha(\bar{\rho})$ proportional to $\bar{\rho} - \bar{\rho}_c$, equation (6a) finally gives to leading order

$$\bar{M} = \bar{M}_0(1 - \bar{\rho}/\bar{\rho}_c)^{1/2}. \quad (7)$$

The results of this interpolation formula are shown in figure 2 together with the self-consistently calculated moments. It is seen that (7) is sufficiently accurate and that it also applies for jellium densities away from the critical point.

We note that the transition investigated here belongs to the simplest type (type I) according to the classification of Morruci [5].

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