Comment on "Mystery of the Alkali Metals: Giant Moments of Fe and Co on and in Cs Films"

In a recent Letter [1] the occurrence of giant magnetic moments of Fe and Co atoms on and in Cs films has been reported. These magnetic moments were detected by employing the anomalous Hall effect, utilizing the fact that the correspondent resistance is known to be proportional to the magnetization of the magnetic atoms [2]. The results were interpreted on the basis of earlier experiments for Fe impurities in Pd films [3] and in bulk Pd [4]. The experiments in Ref. [1] showed a large magnetic response and it was assumed that the magnetic moments of the Fe impurities polarize the surrounding Cs atoms in the same manner as they do in a Pd host [5] so that the total magnetization sums up to the giant moment. As an argument for a possible formation of ferromagnetic moments on Cs, reference is given to a very early Hartree-Fock calculation [6].

In this Comment we argue that this interpretation points in the wrong direction. As for all alkaline metals, the magnetic susceptibility in bcc Cs is small. The density of states at the Fermi energy does not exceed 5 states/Ry/spin which also does not allow for a substantial exchange enhancement. From these considerations any considerable polarization of the Cs neighbors can be ruled out.

The "mystery" of these large moments has actually been resolved much earlier [7] where it was shown that Fe ions implanted in alkali-metal hosts show a fully localized *3d* shell behavior. This means that due to the extremely weak crystal field interaction between the transition-metal host and the alkali-metal host, the transition-metal angular momentum is no longer quenched, as it would be in bulk Fe, so that large orbital moment contributions add to the spin moment. If one assumes that Fe is in a $3d^6$ and Co in a $3d^7$ state, within the framework of Russel-Saunders coupling, the respective ground state is $S = 2$, $L = 2$, $J = 4$, and $g_j = \frac{3}{2}$ for Fe-$3d^6$ and $S = \frac{3}{2}$, $L = 3$, $J = \frac{9}{2}$, and $g_j = \frac{4}{3}$ for Co-$3d^7$ (g_j is the Landé factor). The respective magnetic moments are thus $6\mu_B$ for both Fe and Co. These values for J coincide perfectly with the values derived from the Brillouin function fit performed in Ref. [1] obtaining $J = 4.5$ for Co and $J \approx 4$ for Fe. The magnetic moments given in [1] are too large, since they were derived from the J values using a factor $g = 2$, which of course is valid only for $L = 0$. Our interpretation also explains why the value of the magnetic moment was found to be fairly independent of the thickness or covering of the films. As long as the transition metal is in a completely localized state no changes to its angular momentum state occur. Only for increased concentrations when Fe-Fe interactions become important the angular momentum becomes again quenched by the crystal field and only a spin moment remains. From a calculation of the electronic band structure of the FeCs phase (CsCl structure) we find a large spin moment on Fe ($3.3\mu_B$) but only a minute orbital moment of $0.08\mu_B$.

In summary, the mysterious giant moment found in [1] can be explained on the basis of a fully localized state of the transition metal. The total angular moments detected are in perfect agreement with this assumption. The assumption of polarization effects in the Cs host which should enhance the transition metal moment is not needed and our calculations demonstrate that the polarization of the Cs atoms is very small.

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