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

By measuring and analyzing the anomalous Hall effect for $\frac{1}{100}$ of a monolayer of Fe or Co on the surface or in the bulk of Cs films, Beckmann and Bergmann [1] deduce unusually large magnetic moments of 7 to 8 Bohr magnetons for both Fe and Co impurities. As an interpretation of these large moments the authors suggest a strong polarization of the conduction electrons in Cs similar to the giant magnetic moment formation of Fe and Co in Pd. In the following we give arguments which support the experimental finding of large moments, but we rely on a different interpretation.

A rather direct comparison with known data can be performed in the case of Fe as a bulk impurity in Cs. As worked out in Ref. [2] the magnetic properties observed for Fe in Cs (and in K, Rb) are completely consistent with a fully localized $3d^6$ configuration of isolated Fe impurities with $L = 2, S = 2, J = 4$ in LS coupling. Using the Landé factor $g_J = \frac{3}{2}$ and $\mu_{\text{eff}} = g_J\,[J(J+1)]^{1/2}\,\mu_B$ results in an effective magnetic moment of $6.7\mu_B$. Furthermore, the data imply possible crystal field splitting as being smaller than the LS coupling.

In the case of Co in Cs we can use an extrapolation. By measuring local magnetic susceptibilities and spin dynamics by the time differential perturbed angular γ-ray distribution method, localized d states and negligible crystal fields have been deduced not only for Fe [2], but also for Ni [3], Sc [4], and even for Mo [5] and Tc and Ru [6] ions; all recoil implanted into Cs and/or Rb, K, Na. Since the spatial extension of the d wave function of Co should be smaller than that of Ni, Sc, Mo, Tc, and Ru impurities, a localized d state can reasonably be expected for Co in Cs, also. By extending the Born-Haber cycle (see Ref. [6]) to Co in Cs, we estimate the ground state as being the $3d^7$ configuration with $S = \frac{3}{2}$, $L = 3$, $J = \frac{9}{2}$, $g_J = \frac{4}{3}$, and $\mu_{\text{eff}} = 6.6\mu_B$.

One important reason for the d shell localization is the extremely large difference in the cell volume of the transition metal atoms and alkali atoms (Cs, Rb, K have the largest metallic radii of all condensed elements), leading to a strongly reduced interaction between the impurity d electrons and host conduction electrons and also leading to strong reduction of crystal field effects [2,5,7]. These features resemble the behavior of certain rare earth impurities in metals.

The trends towards localization are further supported by the systematic predictions of magnetic spin-only moments for all $3d$, $4d$, and $5d$ impurities in Cs and other alkali metals within the frame of local spin density approximations [7], which do not include the calculation of orbital contributions or localized ionic d states. For Fe and Co in Cs and for the other d ions in alkali metal hosts under consideration here, the calculations yield almost saturated spin moments and extremely small linewidths of the local $3d$ and $4d$ density features, which are believed to be indicative for the transition from itinerant to localized d electron behavior and thus being consistent with an analysis in terms of ionic-type configurations [2–6].

In view of all these arguments one is led to the conclusion that (at least) the main part of the large moments analyzed in Ref. [1] can be explained by using the parameters for localized Fe and Co states in Cs.

Finally, we comment on the suggestion (Ref. [1]) of Fe or Co induced large ferromagnetic Cs host conduction electron polarizations. The successful analyses of the positive local susceptibilities for non-S-state d ions with large positive magnetic hyperfine fields arising from intact intraatomic orbital correlations and of the negative susceptibilities (and negative magnetic hyperfine fields) for the $4d^5$ S-state ion Mo in Cs, Rb, and Na [5], all consistently imply negligible contributions arising from ferromagnetic host electron polarizations. Furthermore, for a large part of the systems, including Fe in Cs, Kondo effects are observed via the d spin dynamics or reduced local susceptibilities, which arise from an (weak) *antiferromagnetic* interaction of impurity d electrons with host conduction electrons.

M. Gruyters and D. Riegel
Hahn-Meitner-Institut Berlin
Glienicker Strasse 100
14109 Berlin, Germany

Received 19 October 1999
PACS numbers: 75.20.Hr

[1] H. Beckmann and G. Bergmann, Phys. Rev. Lett. **83**, 2417 (1999).
[2] D. Riegel *et al.,* Phys. Rev. Lett. **57**, 388 (1986).
[3] R. Kowallik *et al.,* Phys. Rev. Lett. **63**, 434 (1989).
[4] J. Kapoor *et al.,* Phys. Rev. Lett. **76**, 1537 (1996).
[5] K. D. Gross, D. Riegel, and R. Zeller, Phys. Rev. Lett. **63**, 1176 (1989).
[6] K. D. Gross and D. Riegel, Phys. Rev. Lett. **61**, 1249 (1988).
[7] N. Papanikolaou *et al.,* Phys. Rev. B **46**, 10 858 (1992); Phys. Rev. Lett. **71**, 629 (1993).

 0031-9007/00/85(7)/1582(1)$15.00