Modest magnetic moments of Ti impurities on the surface and in the bulk of K, Rb, and Cs films

Douglas Garrett and Gerd Bergmann*
Department of Physics, University of Southern California, Los Angeles, California 90089-0484
(Received 30 July 2002; published 16 December 2002)

Thin films of Na, K, Rb, and Cs are quench condensed, then covered with 1/100 of a monolayer of Ti and finally covered with the original host. The magnetization of the films is measured by means of the anomalous Hall effect. An anomalous Hall resistance R^{AHE} is observed for Ti on the surface of K, Rb, and Cs and for Ti inside of Cs. Essentially the R^{AHE} varies linearly with the magnetic field and is inversely proportional to the inverse temperature. A small nonlinearity of R^{AHE} suggests a Ti moment of about $1\mu_B$.

DOI: 10.1103/PhysRevB.66.224407 PACS number(s): 75.20.Hr, 71.20.Dg, 73.20.Fz

I. INTRODUCTION

The properties of magnetic transition metal impurities in alkali metal hosts are a very fascinating field. There are only a few experimental investigations of these systems,[1–5] because the transition metal impurities do not dissolve in the alkali hosts, and special low temperature preparation methods must be applied. Riegel *et al.*[1] investigated the properties of Fe in the alkali hosts Cs, Rb, K, and Li. These authors introduced magnetic *d* impurities by nuclear reactions or recoil from nuclear reactions into the alkali metals. For the investigation of the magnetic properties they measured the hyperfine field $B(0)$ at the Fe nucleus in the temperature range from 20 to 350 K. These authors suggest that the Fe atom dissolved in Cs, Rb, and K possesses its atomic electronic structure for the *d* shell, which is a $3d^6$ configuration for Fe. In the following we call this the ''atomic model.'' The resulting magnetic moment of the Fe is then $6\mu_B$. (We define the magnetic moment as $\mu/\mu_B = gJ$).

Our group used the method of quenched condensation onto a substrate at helium temperature to obtain alkali films with *d* impurities. We investigated the magnetic moments of Fe and Co in thin Cs, Rb, K, and Na films.[2–5] The magnetization of the $3d$ impurities was measured by means of the anomalous Hall effect. We observed giant magnetic moments between 6 and $7\mu_B$ for Fe and Co on the surface of these alkali films and even larger moments in the bulk of these alkali hosts with up to $10\mu_B$ for K and Rb hosts. These observed moments are too large to be explained with the ''atomic model'' which yields a maximum moment of $6\mu_B$. Even Ni impurities on the surface and in the bulk of K, Rb, and Cs showed a moment of about $3-4\mu_B$. The disagreement between our experiments and the atomic model became even more dramatic for the magnetic moment of V impurities in K and Na films.[5] Here the atomic model predicts a very small magnetic moment for the V of $\frac{3}{5}\mu_B$ while our experiments yield magnetic moments for the V in Na and on the surface of K of about $7\mu_B$. From all these measurements we conclude that the moments of the $3d$ impurities in the alkali hosts are enhanced by polarizing the conduction electrons of the host. However, this suggestion is at odds with spin-density functional calculations.[6,7]

Theoretically Henry *et al.*[6] found a d^7 configuration for Fe ions in different alkali hosts in a cluster calculation and Solovyev *et al.*[8] arrived at a d^7 configuration for Fe in Rb (using what they call the LDA+U formalism). Guo[7] obtained about 6.3 *d* electrons for Fe ions in K, Rb, and Cs and 7.4 *d* electrons for Co ions in Cs (in an orbital-polarization corrected relativistic spin-density functional calculation).

Recently in a beautiful experiment Gambardella *et al.*[9] applied x-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism to investigate the electronic structure of Fe, Co, and Ni impurities on the surface of K and Na films. They concluded from their experimental results that Fe, Co, and Ni ions in Na and K have localized *d* states. Their conclusion is that the Fe ion is in a d^7 configuration, the Co ion in a d^8 configuration, and the Ni on K in a d^9 configuration. This conclusion differs from the interpretation by Riegel *et al.*, who found good agreement between their experimental results and a simple calculation of the hyperfine field. Gambardella *et al.* rediscovered our result that Ni on the surface of Na is nonmagnetic, and their XAS spectra suggests a superposition of $3d^9$ and $2p^5 3d^{10}$ configurations. A major claim by Gambardella *et al.* is that there is no enhancement of the $3d$ moment in the alkali hosts, but we did not find anywhere an experimental justification of this claim. Our experimentally observed moments of $6-7\mu_B$ for V ions on the surface and in the bulk of K and Rb cannot be explained by any ionic configuration of the V. A V^{++} ion with d^3 should have a moment of $\frac{3}{5}\mu_B$ while a d^4 configuration should have $J=0$ and therefore zero moment. Without an enhancement the localized model is completely incapable of explaining our experimental results.

So far we observed only one nonmagnetic combination of a $3d$ impurity in an alkali host, the Na/Ni system where the Ni did not show a moment on the surface or in the bulk of Na. There might be a similar chance for the absence of the magnetic moment on the left side of the $3d$ row for Sc and Ti. Therefore in this paper we study the magnetic properties of Ti impurities on the surface and in the bulk of the alkali hosts Na, K, Rb, and Cs.

II. EXPERIMENTAL RESULTS

Since transition metal impurities do not dissolve in alkali hosts one has to prepare the samples at low temperatures. We use the method of quenched condensation. The Na, K, Rb, and Cs films are evaporated from alkali dispensers made by SAES-Getters. The quartz substrate is at He temperature, and

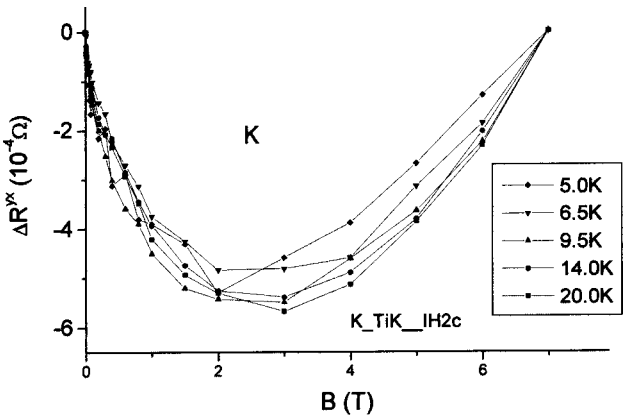

FIG. 1. The nonlinear part of the Hall resistance $\Delta R^{yx} = R^{yx}(B) - B\,[R^{yx}(7\text{ T})/7\text{ T}]$ for a pure K film.

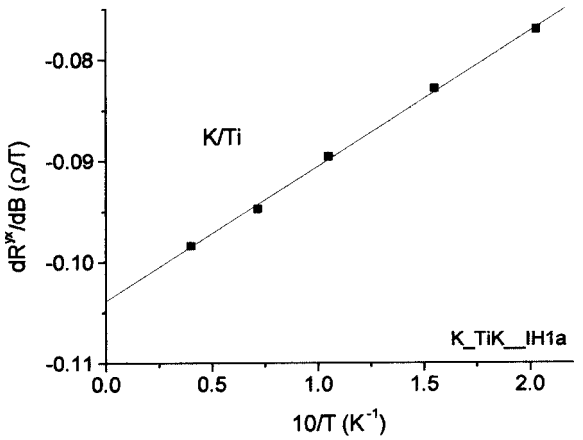

FIG. 2. The initial slope of the total Hall resistance as a function of $10/T$ (squares) for a K film with Ti surface impurities.

the ultra high vacuum is better than 10^{-11} Torr. The preparation of our film samples is generally performed in three steps. First the alkali host film is quench condensed onto a crystalline quartz plate and then annealed to 40 K. The film thickness is chosen so that the resistance per square is of the order of 100Ω after annealing. The thickness lies in the range between 5 and 10 nm. Then the host is covered with about 0.01 atomic layer of Ti. In the third step the Ti is sandwiched with about five atomic layers of the alkali host. In some experiments a submonolayer of Pb (0.01 to 0.2 atomic layers) is condensed prior to the first alkali film or on top of the sandwich. (This is done to introduce a strong spin-orbit scattering. Then the magnetoresistance measurements together with the theory of weak localization permit the determination of the magnetic scattering of the Ti impurities.[10,11]) Details of the film preparation are discussed in former papers.[2,3]

After each condensation the magnetoresistance and Hall resistance of the film are measured in the field range between $-7\text{ T} \leqslant B \leqslant +7\text{ T}$ at several temperatures: 5, 6.5, 9.5, 14, and 20 K. From the Hall resistance we determine the initial slope dR^{yx}/dB at $B=0$.

We first describe the experimental results for the system K/Ti/K. For the pure K film the Hall resistance is nicely proportional to the magnetic field. For the pure K film this initial slope dR^{yx}/dB is temperature independent within 0.5%. There is, however, a small but finite deviation from linearity.

To make the nonlinear part of the (anomalous) Hall resistance visible we subtract the linear part of the Hall resistance through the 7 T point of the Hall curve $B\,[R^{yx}(7\text{ T})/7\text{ T}]$. This yields for the nonlinear Hall resistance $\Delta R^{yx} = R^{yx}(B) - B\,[R^{yx}(7\text{ T})/7\text{ T}]$. This small but finite nonlinear Hall resistance ΔR^{yx} is plotted in Fig. 1 for five different temperatures as a function of the magnetic field. The relative nonlinearity $\Delta R_m^{yx}/R^{yx}(7\text{ T})$ [the maximal deviation from linearity divided by $R^{yx}(7\text{ T})$ at the maximum field of 7 T] is rather small, less than 1.0×10^{-3} for the discussed K film. Furthermore the nonlinearity is almost temperature independent.

When the K film is covered with 0.01 atomic layers of Ti the initial slope changes dramatically, dR^{yx}/dB depends now linearly on the reciprocal temperature $1/T$. In Fig. 2 the initial slope dR^{yx}/dB of the experimental Hall resistance for a K/Ti film is plotted versus the inverse temperature $10/T$. The experimental points lie on a straight line. The extrapolation $1/T \to 0$ yields the derivative of the normal Hall resistance dR_n^{yx}/dB while the temperature-dependent part represents the anomalous Hall resistance (AHR) which is proportional to the susceptibility. The extrapolated value of the (initial slope of the) normal Hall resistance agrees by better than 0.5% with the normal Hall resistance of the pure K film. The extrapolated normal slope allows a separation of the normal and anomalous Hall resistance. We define the normal Hall resistance as a linear function of the magnetic field: $R_n^{yx}(B) = (dR_n^{yx}/dB)B$.

The inverse temperature dependence of the initial slope of the AHR R^{AHE} (which is proportional to the susceptibility) is the typical behavior of a free magnetic moment. The magnitude of the AHR is remarkable; at 5 K the initial slope is of the order of 20% of the normal Hall resistance slope.

In Fig. 3 the anomalous Hall resistance of the K/Ti film is plotted as a function of the magnetic field for five different temperatures. On this scale the AHR and therefore the magnetization are proportional to the applied magnetic field B. The linear field dependence allows the conclusion that (a) the Ti impurities have a magnetic moment and (b) this moment is not a giant moment.

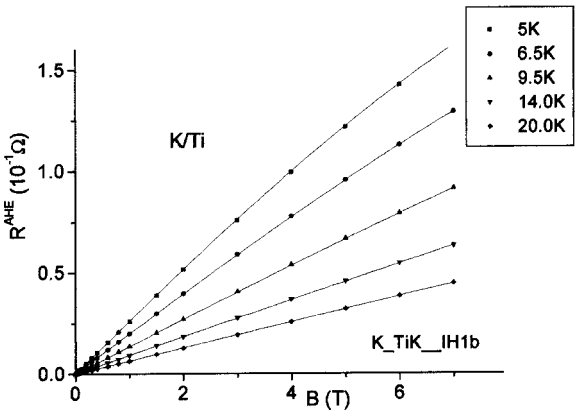

FIG. 3. The anomalous Hall resistance of Ti impurities on the surface of a K film for different temperatures.

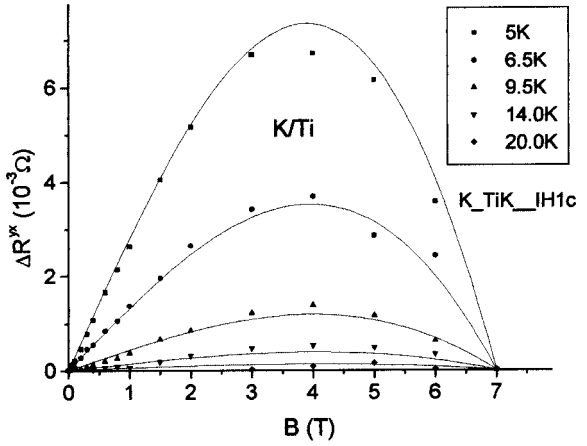

FIG. 4. The nonlinear part of the Hall resistance $\Delta R^{yx} = R^{yx}(B) - B[R^{yx}(7\ \mathrm{T})/7\ \mathrm{T}]$ for Ti impurities on the surface of a K film. The full curves are calculated with $J=1$, $g=0.88$.

However, the linear AHR does not permit one to determine the actual size of the moment. For this we have to turn to the nonlinear part of the AHR. In Fig. 4 we have plotted $\Delta R^{yx} = R^{yx}(B) - B\ [R^{yx}(7\ \mathrm{T})/7\ \mathrm{T}]$ as a function of the magnetic field. According to the definition of ΔR^{yx} the data points return to zero at $B=7$ T. One recognizes that there is a clear nonlinearity in the experimental data, although it is smaller by a factor 30 than the linear AHR. Below we are going to use this nonlinearity to analyze the size of the magnetic moment of the Ti impurities.

When the K/Ti film is covered with additional K the nonlinear behavior of the Hall resistance degenerates. In Fig. 5 the nonlinear Hall resistance is plotted for different temperatures. This ΔR^{yx} is composed of the ΔR^{yx} of the pure K film (see Fig. 1) and an additional Ti contribution. Both terms appear to be of the same order of magnitude. There is a small temperature-dependent contribution but it is only slightly larger than the scattering of the data. The new nonlinearity is at least an order of magnitude smaller than the ΔR^{yx} of the K/Ti film. On the other hand, there is still a large temperature dependence of the initial slope of the Hall resistance. In Fig.

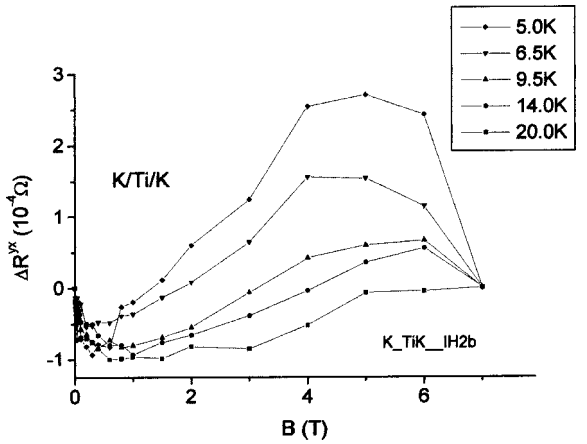

FIG. 5. The nonlinear part of the Hall resistance $\Delta R^{yx} = R^{yx}(B) - B[R^{yx}(7\ \mathrm{T})/7\ \mathrm{T}]$ for Ti impurities sandwiched between two K films (bulk impurities).

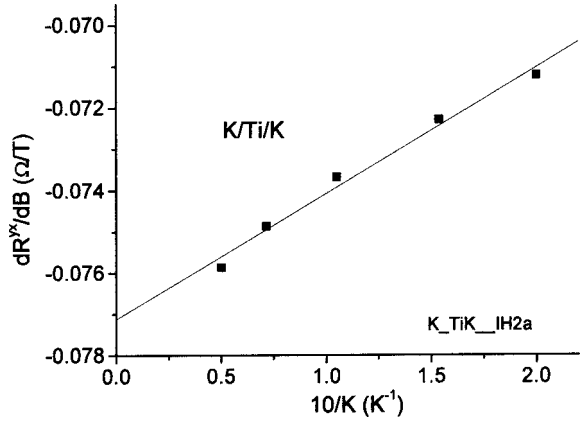

FIG. 6. The initial slope of the total Hall resistance as a function of $10/T$ (squares) for a K film containing bulk Ti impurities.

6 the dR^{yx}/dB is plotted as a function of $10/T$. It yields a reasonably straight line with about half the relative temperature dependence as for the K/Ti film. This demonstrates that the Ti impurities in the bulk of K are also magnetic.

The magnetic character of the Ti impurities is confirmed by magnetoresistance measurements. In Fig. 7 the magnetoresistance of the pure K film, the K/Ti and K/Ti/K are plotted for 5.0 K. The different curves have the same conductance scale (which is the natural scale for weak localization). The points are experimental points. The full curves are numerically calculated and composed of weak localization, a small contribution from a quadratic magnetoresistance and a Lorentzian curve. The latter contributes only at large fields. The width of the magnetoresistance is proportional to the

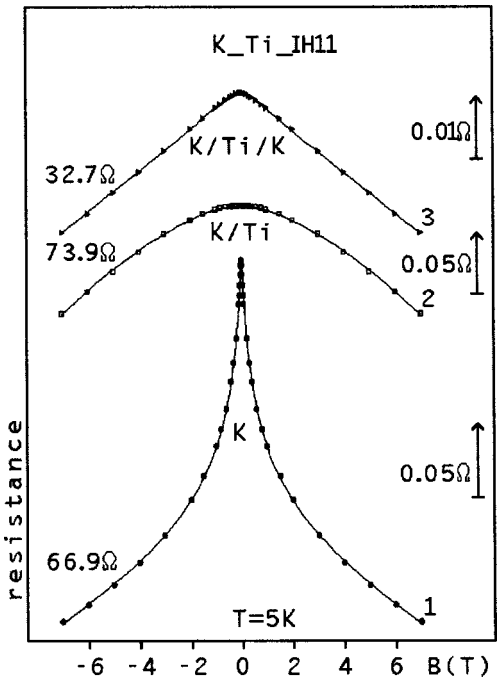

FIG. 7. The magnetoresistance curves of a pure K film, a K/Ti film with 0.01 atomic layers of Ti on the surface, and a sandwich of K/Ti/K where the 0.01 atomic layer of Ti is covered with about 5 atomic layers of K.

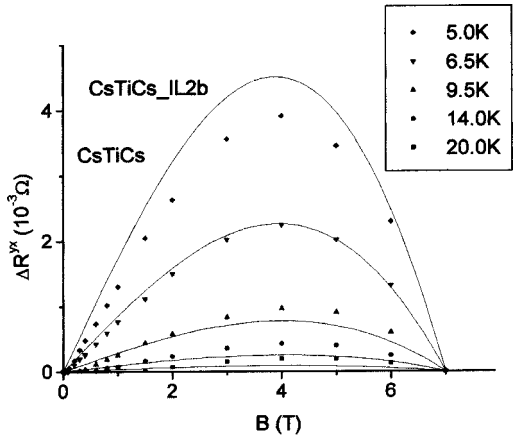

FIG. 8. The nonlinear part of the Hall resistance $\Delta R^{yx} = R^{yx}(B) - B[R^{yx}(7\text{ T})/7\text{ T}]$ for a Cs/Ti/Cs sandwich (bulk Ti impurities). The full curves are calculated with $J=1$, $g=1.0$.

sum of inelastic dephasing and the magnetic scattering rate due the Ti impurities. One can easily detect the dramatic increase in the width of the magnetoresistance due to the Ti impurities. These measurements prove that the Ti is magnetically active on the surface and in the bulk of K.

The experimental results for the Rb host with Ti impurities are qualitatively identical with the results for the K-Ti system. We observe a strong temperature dependence of the initial slope of the Hall resistance which follows a $1/T$ law. The nonlinearity ΔR^{yx} is of the same order of magnitude on the surface. In the bulk the nonlinearity is strongly reduced and the Ti causes a similar broadening of the magnetoresistance curves as in the K host.

For the Cs film covered with 0.01 atomic layers of Ti we again observe qualitatively the same behavior as for the K and Rb films covered with Ti. However, bulk Ti impurities behave differently in Cs than in K and Rb. In Cs/Ti/Cs sandwiches the nonlinearity of the Hall resistance ΔR^{yx} does not collapse as for the K and Rb hosts, but it is similar to that for Ti on the surface of Cs. This is shown in Fig. 8 where ΔR^{yx} is plotted as a function of the applied magnetic field. One recognizes the same pattern as in Fig. 4 for Ti on the surface of K. It appears that in the Cs host the magnetic state of the Ti impurities survives the coverage with the host better than in K and Rb.

The Na host on the other hand does not support the magnetic state of the Ti impurities, neither on the surface nor in the bulk. The Ti impurities do not induce a temperature dependence of the Hall resistance. The Ti impurities cause only a slight broadening of the magnetoresistance. The magnetoresistance curves for Na, Na/Ti, and Na/Ti/Na are shown in Fig. 9. The experimental data are represented by the points, and the full curves are theoretical curves calculated with the theory of weak localization.[10,11] They will be discussed below.

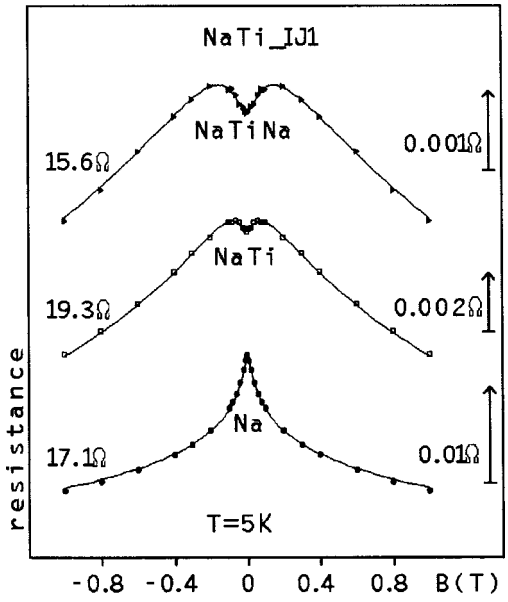

FIG. 9. The magnetoresistance curves of a pure Na film, a Na/Ti film with 0.01 atomic layers of Ti on the surface and a sandwich of Na/Ti/Na, where the 0.01 atomic layer of Ti is covered with about five atomic layers of Na. The full curves are calculated with the theory of weak localization.

III. EVALUATION AND DISCUSSION

The electronic structure and the magnetic properties of magnetic impurities in a solid can be very complex. The simplest case is the ''free spin model'' having a free magnetic moment with a total angular moment J and a Lande factor g, where J is a multiple of $\frac{1}{2}$. When the $3d$ electrons strongly hybridize with the conduction electrons one obtains the resonance model of a magnetic impurity. Its properties are very complex because at sufficiently low temperature it forms a nonmagnetic singlet state due to the Kondo effect. Above the Kondo temperature one often uses the mean field solution of the Friedel-Anderson resonance model as a guide.[12,13] Further complications can arise from crystal field effects (The effect of the crystal field in alkali hosts has been ruled out by Riegel *et al.*[1])

Since the AHR (and therefore the magnetization) are essentially linear in the magnetic field the magnetic Ti impurities are only partially aligned, even in the field of 7 T. (The Ti impurities are in the low field limit.) As we see below, the experiments yield for each system two key pieces of information dR^{yx}/dB and ΔR_m^{yx}, the (temperature dependence of the) initial slope and the nonlinearity. Since one parameter is needed to determine the amplitude of the AHR, i.e., the proportionality constant between the individual moment and its contribution to the AHR, there is only one additional parameter left to be determined. Therefore in our evaluation we use the ''free spin model'' of a free magnetic moment. Its properties are described by J and g where J can take the values $\frac{1}{2}$, 1, $\frac{3}{2}$, 2,.... It will turn out that our evaluation yields a relatively well determined value for the magnetic moment. If future theoretical or numerical structural investigations yield a different model then it will be relatively easy to translate the results of our evaluation into such a model.

It should, however, be emphasized that our experimental results yield a strong support for the free moment behavior because of the inverse temperature dependence of dR^{yx}/dB.

Theoretically the normalized magnetization (and AHR) of free magnetic moments are given by the Brillouin function

$$B_J(x)=\frac{2J+1}{2J}\coth\left(\frac{2J+1}{2J}x\right)-\frac{1}{2J}\coth\left(\frac{1}{2J}x\right),$$

where

$$x=\frac{gJ\mu_B B}{k_B T}$$

and g is the Lande factor, μ_B the Bohr magneton, J the total angular momentum, and k_B the Boltzmann constant. For small x the Brillouin function can be expanded in powers of x. The linear part is

$$B_J^{(1)}(x)=\left(\frac{1}{3}\frac{J+1}{J}\right)x=\frac{g(J+1)\mu_B B}{3k_B T}$$

and the cubic term is

$$B_J^{(3)}(x)\approx-\frac{1}{90}\frac{(J+1)(2J^2+2J+1)}{J^3}x^3.$$

Our maximum field of $B=7\,[\mathrm{T}]$ corresponds to an x_0 value with $x_0=gJ\mu_B(7\,[\mathrm{T}])/k_BT$ which depends on the temperature. (The units for T and K are inclosed by square brackets to avoid confusion with the temperature).

For a comparison with our experimental nonlinearity ΔR^{yx} we have to subtract from $B_J^{(3)}(x)$ the linear contribution $(x/x_0)B_J^{(3)}(x_0)$. This yields

$$\Delta B_J(x)=\frac{1}{90}\frac{(J+1)(2J^2+2J+1)}{J^3}(x_0^2x-x^3).$$

This function vanishes at $x=0$ and $x=x_0$ as required. It has its maximum at $x_m=x_0/\sqrt{3}$. The value at the maximum is

$$\Delta B_J(x_m)=\frac{\sqrt{3}}{405}\frac{(J+1)(2J^2+2J+1)}{J^3}x_0^3.$$

The maximum of the nonlinearity at $x_0/\sqrt{3}$ corresponds to a magnetic field of $B_m=7\,[\mathrm{T}]/\sqrt{3}\approx4.04$ T.

Inserting x_0 yields

$$\Delta B_J(B_m)=\frac{\sqrt{3}}{405}(J+1)(2J^2+2J+1)g^3\left(\frac{\mu_B}{k_B}\right)^3(7\,[\mathrm{T}])^3\frac{1}{T^3}.$$

The value of the maximum should vary with temperature as T^{-3}.

The magnetization and the experimental AHR are both proportional to the Brillouin function, each with a constant (temperature and field independent) factor. For the AHR this means that

$$R^{\mathrm{AHE}}(B,T)=a_{\mathrm{AHE}}B_J\left(\frac{gJ\mu_B B}{k_B T}\right).$$

We define a linear and a nonlinear factor f_l and f_{nl}. Both can be determined experimentally. The two expressions

$$f_l=T\frac{dR^{\mathrm{AHE}}}{dB}=a_{\mathrm{AHE}}\frac{g(J+1)\mu_B}{3k_B},$$

$$f_{nl}=T^3\Delta R^{\mathrm{AHE}}(B_m,T)$$
$$=a_{\mathrm{AHE}}\frac{\sqrt{3}}{405}(J+1)(2J^2+2J+1)g^3\left(\frac{\mu_B}{k_B}\right)^3(7\,[\mathrm{T}])^3$$

are temperature (and field) independent and therefore constant. In the ratio

$$\frac{f_{nl}}{f_l}=\frac{T^3\Delta R^{\mathrm{AHE}}(B_m)}{T\left.\frac{dR^{\mathrm{AHE}}}{dB}\right|_{B=0}}=\frac{T^3\Delta B_J(x_m)}{T\left.\frac{dB_J(x)}{dx}\right|_{x=0}\frac{dx}{dB}}$$

the proportionality factor a_{AHE} cancels. We form the ratio of the two constant (temperature independent) terms

$$\frac{f_{nl}}{f_l}=\frac{\frac{1}{90}(J+1)(2J^2+2J+1)\frac{2\sqrt{3}}{9}g^3\left(\frac{\mu_B}{k_B}\right)^3(7\,[\mathrm{T}])^3}{\frac{1}{3}(J+1)g\frac{\mu_B}{k_B}}$$

$$=\frac{1}{135}\sqrt{3}(2J^2+2J+1)g^2\left(\frac{\mu_B}{k_B}\right)^2(7\,[\mathrm{T}])^3$$

$$=\frac{1}{135}\sqrt{3}(0.67171)^2 7^3(2J^2+2J+1)g^2[\mathrm{T\,K^2}]$$

$$=(2J^2+2J+1)g^2(1.9856[\mathrm{T\,K^2}])$$

$$\approx(2J^2+2J+1)g^2 2[\mathrm{T\,K^2}],$$

where the factor 2 $[\mathrm{T\,K^2}]$ approximates the value $\frac{1}{135}\sqrt{3}(\mu_B/k_B)^2\,(7\,[\mathrm{T}])^3=1.9856\,[\mathrm{T\,K^2}]$. We obtain a relation between J and g:

$$2(2J^2+2J+1)g^2=\frac{f_{nl}}{f_l},$$

$$g=\sqrt{\frac{f_{nl}/f_l}{2(2J^2+2J+1)}}.$$

A. Amplitude

The amplitude of the AHR can be obtained from the linear expansion of the Brillouin function

$$T\frac{dR^{\mathrm{AHE}}}{dB}=a_{\mathrm{AHE}}\frac{g(J+1)\mu_B}{3k_B}=f_l,$$

$$a_{\mathrm{AHE}}=\frac{3f_l}{g(J+1)\frac{\mu_B}{k_B}}.$$

Experimentally for the K/Ti system with the Ti impurities on the surface of the K we find that the derivative of the AHR with respect to temperature is indeed inversely proportional to the temperature $dR^{\mathrm{AHE}}/dT\propto1/T$, as Fig. 2 demonstrates. To demonstrate the constancy of the term

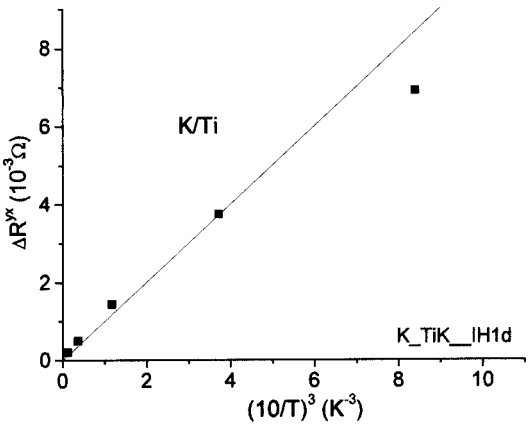

FIG. 10. The maximum value of ΔR^{yx} at different temperatures for a K film covered with 0.01 atomic layers of Ti as a function of $(10/T)^3$.

$T^3\Delta R^{\mathrm{AHE}}(B_0)$ we plot in Fig. 10 the maximum of the nonlinear Hall resistance $\Delta R^{\mathrm{AHE}}(B_0)$ versus $(10/T)^3$. One obtains a reasonably straight line through the origin. The two plots give the following values: $f_{nl}=T^3\Delta R^{\mathrm{AHE}}(B_0)=3.7\times10^{-3}\times6.5^3=1.0\,[\Omega\,\mathrm{K}^3]$ and $f_l=T(dR^{\mathrm{AHE}}/dB)|_{B=0}\approx0.13\,[\Omega\mathrm{K/T}]$. This yields

$$g=\sqrt{\frac{7.7}{2(2J^2+2J+1)}}.$$

B. Magnetic moments

We obviously obtain for a given J a Lande factor and a magnetic moment. We evaluate the Lande factor, the total moment $\mu=Jg\mu_B$ and the amplitude of the AHR for half integer values of J in the range from $\frac{1}{2}$ to 2. In Table I the resulting g values and magnetic moments for different values of J are collected.

Larger values of J yield smaller values for g. For J values between 1 and 2 the magnetic moment is of the order of one Bohr magneton and depends only weakly on the total angular momentum. We conclude that our experimental results suggest a magnetic moment of the Ti impurities of about one Bohr magneton. The full curves in Fig. 4 are calculated with $J=1$ and $g=0.88$. If one considers the agreement between the experimental points and the full curves not as perfect, one has to keep in mind that the relative deviation is very small, only 5×10^{-3}.

TABLE I. The total angular momentum, Lande factor and magnetic moment of the Ti impurities on the surface of the K film. a_{AHE} is the saturation amplitude of the anomalous Hall resistance.

J	g	μ	a_{AHE} [Ω]
1/2	1.24	0.62	0.31
1	0.88	0.88	0.33
3/2	0.67	1	0.35
2	0.54	1.08	0.36

TABLE II. For $J=1$ the Lande factor g, the magnetic moment μ, and the amplitude of the AHR are calculated from linear and nonlinear behavior, f_l and f_{nl}, of the AHR.

	K/Ti	Rb/Ti	Cs/Ti	Cs/Ti/Cs
g	0.88	0.74	1.1	1
μ/μ_B	0.88	0.74	1.1	1
a_{mp} (Ω)	0.67	0.65	0.57	0.16
f_l [Ω K/T]	0.13	0.217	0.28	0.0707
f_{nl} [Ω K^3]	1.0	1.3	3.4	0.7

The atomic model predicts for Ti^{++} impurities with two d electrons the following values: $J=2$, $g=\frac{2}{3}$, and $\mu=\frac{4}{3}\mu_B$. The resonance model of Friedel and Anderson (with suppressed orbital angular momentum) allows for a magnetic moment between 0 and $2\mu_B$ and a Lande factor of 2. In this case the atomic model is in reasonable agreement with the experimental results.

Our experimental results show a well developed nonlinear Hall resistance for Ti on the surface of K, Rb, and Cs. For Ti within Cs we obtain similar results. For all these systems we perform the above evaluation. They show very similar results. Again the moment depends only a little on the choice of g. In Table II we collect the data for $J=1$ which yield the characteristic magnetic moment.

C. The Na/Ti system

In the Na/Ti system we do not observe an AHR.

D. Sign of the anomalous Hall resistance

For all the alkali host with $3d$ impurities that we investigated so far the sign of the AHR was negative. This includes Cs/Fe, Cs/Co, K/Fe, K/Co, K/V, Rb/Fe, Rb/Co, Na/Fe, Na/Co, and Na/V. Only in the Na/Ni system there was no AHR at all, not even a dephasing of weak localization. In contrast we observe for Cs/Ti, Rb/Ti, and K/Ti a positive AHR.

E. Ti inside of K and Rb films

As a comparison between surface and bulk effects, Figs. 4 and 5 show that the nonlinearity of the Hall resistance ΔR^{yx} strongly reduces when the Ti impurities on the surface of K are covered with additional K. Our interpretation of Fig. 5 is that the nonlinear part of the anomalous Hall resistance is reduced by a factor 10 or more and is therefore no longer much larger than the nonlinear part of the Hall resistance of the pure K (which is reduced by about 25% due to the increase of the thickness). What we see in Fig. 5 is the superposition of the (reduced) ΔR^{yx} of Fig. 1 and a much smaller nonlinear AHR. The reduction of the latter can only be estimated; it is reduced by a factor of about 10 to 20. This means that the Lande factor and therefore the magnetic moment of the Ti is reduced by roughly a factor of 4. This may not be the only possible interpretation of the experimental observation but it appears to be the most natural one.

IV. CONCLUSIONS

The properties of Ti impurities on the surface and in the bulk of Na, K, Rb, and Cs films are investigated by means of the anomalous Hall effect and the magneto-resistance. The AHR is proportional to the magnetization of the Ti impurities perpendicular to the film surface. Ti impurities on the surface and in the bulk of K, Rb, and Cs films possess a free magnetic moment as demonstrated by the inverse temperature dependence of the initial slope of the anomalous Hall resistance. Ti on the surface and in the bulk of Na films does not possess a magnetic moment but does show a small dephasing of weak localization. The latter is probably due to spin fluctuations. From the nonlinearity of the (anomalous) Hall effect we obtain an estimate of the size of the magnetic moment. Within the ''free spin model'' of magnetic moments we obtain about one Bohr magneton for Ti impurities on the surface of K, Rb, and Cs and for Ti impurities in the bulk of Cs. In the hosts of K and Rb the (bulk) Ti impurities produce a much smaller nonlinearity of the AHR. We interpret this difference as a reduction of the Ti moment by roughly a factor of 4.

This behavior of Ti impurities on the surface and in the bulk of alkali films is another example that the electronic and magnetic properties of $3d$ impurities are not the same on the surface and in the bulk of a host. For K films with V impurities we observed a similar behavior. It shows that one has to be careful in extrapolating from the surface to the bulk behavior.

If these experiments had been the first measurements of a $3d$ impurity in alkali hosts then the observed moments would be essentially in line with our expectations. Ti impurities have no moment in the noble metal hosts.[14] Since the electron density is reduced in the alkali hosts this could explain the finite moment for the K, Rb, and Cs hosts. The observation of two rather different moments on the surface and in the bulk of K and Rb might be somewhat unusual but not out of line. The fact that a magnetic moment forms easier on the surface than in the bulk is well established.[15,16] The increased tendency toward magnetism in going from Na to Cs is confirmed. However, the properties of the Ti impurities in the alkali hosts differ quite dramatically from those of other $3d$ impurities. For V, Fe, and Co impurities we observed in the past magnetic moments of $6\mu_B$ or more. For Ni in Cs, Rb, and K the evaluated moments were of the order of $3-4\mu_B$. (The only exception was Ni on the surface and in the bulk of Na which was nonmagnetic.) Therefore the Ti results are now rather an exception than the rule.

Another significant difference between the Ti impurities and V, Fe, Co, and Ni in the systems which have been investigated so far is that the sign of the anomalous Hall resistance is different. Ti impurities yield a positive AHR. This represents an important clue about the electronic structure of the $3d$ impurities in the alkali hosts. Unfortunately our microscopic understanding of the electronic and transport properties of alkali hosts with $3d$ impurities is not yet sufficiently developed to extract this information.

Our results add to the theoretical challenge. At the present time the theoretical and numerical calculations cannot explain this diverse magnetic behavior of $3d$ impurities in the alkali hosts.

ACKNOWLEDGMENTS

The research was supported by NSF Grant No. DMR-9814260.

*Email address: bergmann@usc.edu

[1] D. Riegel, H. J. Barth, and L. Buermann, Phys. Rev. Lett. **57**, 388 (1986).
[2] H. Beckmann and G. Bergmann, Phys. Rev. Lett. **83**, 2417 (1999).
[3] Mohamed Hossain and Gerd Bergmann, Eur. Phys. J. B **26**, 7 (2002).
[4] G. Bergmann and M. Hossain, Phys. Rev. Lett. **86**, 2138 (2001).
[5] Funing Song and Gerd Bergmann, Phys. Rev. Lett. **88**, 167202 (2002).
[6] M. E. McHenry, J. M. MacLaren, D. D. Vvedensky, M. E. Eberhart, and M. L. Prueitt, Phys. Rev. B **40**, 10 111 (1989).
[7] G. Y. Guo, Phys. Rev. B **62**, R14 609 (2000).
[8] I. V. Solovyev, P. H. Dederichs, and V. I. Anisimov, Phys. Rev. B **50**, 16 861 (1994).
[9] P. Gambardella, S. S. Dhesi, S. Gardonio, C. Grazioli, P. Ohresser, and C. Carbone, Phys. Rev. Lett. **88**, 047202 (2002).
[10] S. Hikami, A. I. Larkin, and Y. Nagaoka, Prog. Theor. Phys. **63**, 707 (1980).
[11] G. Bergmann, Phys. Rep. **107**, 1 (1984).
[12] J. Friedel, Philos. Mag. **43**, 153 (1952); Adv. Phys. **3**, 446 (1954); Philos. Mag., Suppl. **7**, 446 (1954); Can. J. Phys. **34**, 1190 (1956); Nuovo Cimento, Suppl. **7**, 287 (1958); J. Phys. Radium **19**, 38 (1958).
[13] P. W. Anderson, Phys. Rev. **124**, 41 (1961).
[14] G. Bergmann and H. Beckmann, Phys. Rev. B **52**, 15 687 (1995).
[15] H. Beckmann, R. Schaefer, W. Li, and G. Bergmann, Europhys. Lett. **33**, 563 (1996).
[16] P. Lang, V. S. Stepanyuk, K. Wildberger, R. Zeller, and P. H. Dederichs, Solid State Commun. **92**, 755 (1994).