Induced Spin-Currents in Alkali-Films

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Abstract

In sandwiches of FeK and FeCs the conduction electrons in the alkali metals have a large mean free path. The experiments suggest that the specular reflection for spin up and down electrons is different at the interface yielding a spin current in the alkali film. The spin current is detected by the anomalous Hall effect of Pb surface impurities.

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1. Introduction

Spin currents and the asymmetric scattering of electron spins by magnetic moments and spin-orbit scatterers has attracted great scientific interest in recent years. In the presence of a spin-orbit scatterer spin currents cause an anomalous Hall effect [1], [2], [3]. The whole complex of spin current, spin-orbit scattering and anomalous Hall effect (AHE) is an area with many unsolved and very interesting problems.

In this paper we describe a number of experiments on quench-condensed sandwiches of FeCs and FeK. Because of the low condensation temperature the conduction electrons in the Fe have a very short mean free path of about 1 $\text{nm}$, but the electrons in the alkali films possess a mean free path $l_K$ which can be up to five times the film thickness. An example is shown in Fig.1 where the mean free path of the K electrons is shown as a function of the K thickness $d_K$. For a K thickness of 20 $\text{nm}$ the mean free path is about $l_K = 100 \text{ nm}$, more than four times the thickness. This is rather surprising since the Fe film is very disordered, and its mean free path is only $l_{Fe} = 1 \text{ nm}$. Our conclusion is that the conduction electrons from the K film barely enter the Fe film. The interface mostly reflects the potassium electrons specularly. The detailed mechanism that decouples the two films is not yet well understood. Because $l_K$ is so large we have to assume that the potassium electrons are almost perfectly specularly reflected at the interface. If the electrons would reach the disordered Fe they would be scattered diffusively. This would yield according to Fuchs [4] and Sondheimer [5] an effective mean free path of the order of the film thickness.

There is, however, a small transmission of electrons between the Fe and the K films. In the presence of an electric field the electrons in the Fe do not flow parallel to the electric field, but the current and electric field form an angle due to the anomalous Hall effect (AHE). The current possesses a component perpendicular to the electric field. If electrons cross the interface between the Fe and the alkali film they transfer this "anomalous" current component into the non-magnetic alkali film and introduce there an anomalous Hall conductance (AHC). Such an AHC has been observed in sandwiches of FeCs and other sandwiches of a ferromagnet and an alkali metal [6].

Therefore sandwiches of Fe and Cs (or any other ferromagnet with any alkali metal) represent a rather interesting system; (i) the electrons in the alkali film behave almost as in a perfectly specularly reflecting film, (ii) there is a weak transfer of electrons between the two films.
Since the chemical potentials for spin up and spin down electrons in the Fe are different, one could expect a somewhat different reflection of the spin up and down electrons at the interface. The bulk mean free path of the conduction electrons in the alkali films is fairly large. Therefore the effective mean free path (including surface scattering) is strongly influenced by the degree of specular reflection at the interface. A different specular reflectivity for spin up and down electrons would therefore create a spin current in the alkali film. The question is: How large is the spin current and how can it be detected?

It has been known for a long time that a spin current causes an AHE in the presence of strong spin-orbit scattering. Ballentine and Huberman [1] calculated the correction to the Hall constant in liquid Pb and Bi metals as the consequence of the field-induced spin polarization and the spin-orbit scattering by Pb or Bi atoms. Hirsch [2] introduced a spin Hall effect due to spin-orbit scattering. One of the authors [3] calculated exactly the AHE of a polarized electron gas in the presence of spin-orbit scattering using Friedel phase shifts for the total angular momentum.

The goal of this paper is to check whether sandwiches of FeCs possess a detectable spin current in the Cs (in the presence of an electric field). Our detection tool will be the spin-orbit scattering of Pb impurities which we condense in situ on the free surface of the Cs. If the Cs possesses a spin current close to its free surface then the Pb impurities will cause an AHE. Therefore the anomalous Hall conductance (AHC) of the sandwich will increase when the FeCs sandwich is covered with Pb.

For this investigation we quench condense at helium temperature a thin Fe film with a thickness of about 10 nm and a resistance of about 100 Ω. After the Fe film has been annealed to 40 K it is covered with a Cs film. The thickness of the alkali film is varied between 5 nm and 30 nm. Finally the alkali film is covered with sub-mono-layers of Pb, starting with 0.01 atomic layers and then increasing the Pb coverage to 0.02, 0.05, 0.1, 0.2, 0.5 and 1 atomic layers. Each time the magneto-resistance and the Hall resistance are measured in the field range between -7 and +7 T.

In Fig.2 the anomalous Hall resistance (after subtracting the linear Hall resistance) is plotted for the FeCs sandwich and the same sandwich covered with 2/100 atomic layers of Pb. The thicknesses of the Fe and Cs films are: \( d_{Fe} = 12.4 \text{ nm} \), \( d_{Cs} = 29.6 \text{ nm} \). One recognizes that the coverage with 0.02 atomic layers of Pb introduces a large AHC. We extrapolate the Hall curves back to zero magnetic field. The Hall resistance of the alkali metals is not perfectly linear in the
magnetic field and the extrapolation has to include this slight curvature. The extrapolated Hall resistance $R_{yx}^0$ is the anomalous Hall resistance for the artificial case of a sandwich in field $B = 0$ and the magnetization of the Fe polarized in the $z$-direction.

From the Hall resistance $R_{yx}^0$ and the resistance $R_{xx}$ (per square) of the sandwich we calculate the Hall conductance $L_{xy}^0 = R_{yx}^0 / (R_{xx})^2$. The anomalous Hall conductance is shown in Fig.3 as a function of the Pb coverage (in units of atomic layers). It increases strongly with the first 1/100 of a mono-layer of Pb. (Actually the increase in $R_{yx}^0$ is even stronger than in $L_{xy}^0$ since $R_{xx}$ increases as well with the Pb coverage [7]). Somewhere at a Pb coverage of about 0.03 atomic layers $L_{xy}^0$ reaches its maximum. For increasing Pb coverage the AHC shows a steep decline with a minimum at about 0.3 atomic layers of Pb. Further increase in the Pb coverage yields a soft increase of $L_{xy}^0$ which approaches a saturation value. One appreciates the size of the AHC due to the Pb when one compares it with the AHC of the underlying ferromagnetic Fe film. For the disordered Fe film the AHC $L_{xy}^0$ has the values $6.6 \times 10^{-5}\Omega^{-1}$, for the FeCs sandwich $L_{xy}^0$ is $1.0 \times 10^{-4}\Omega^{-1}$ while the additional contribution due to 2/100 atomic layers of Pb is $5.0 \times 10^{-4}\Omega^{-1}$. Obviously we are dealing here with a large effect.

In the next experimental step we want to make sure that it is the spin-orbit scattering of Pb that produces the observed AHC. The Pb impurities are strong spin-orbit scatterers because of their large nuclear charge. Therefore we have performed additional experiments where similar FeCs sandwiches are covered with sub-mono-layers of Ag and Mg. Ag and Mg impurities possess a much smaller spin-orbit scattering than Pb impurities. The results for the Ag coverage are included in Fig.3. In this case the AHC does not show the pronounced peak. Instead the additional AHE is much smaller and $L_{xy}^0$ approaches its saturation value on the scale of a few hundredth of a mono-layer of Ag. The result for the Mg coverage is similar. This confirms that the spin-orbit scattering of the Pb is essential in the observation of the AHC.

In addition we have investigated the size of the AHC as a function of the Cs thickness. The magnitude of the AHE due the Pb increases with increasing Cs thickness. It also depends on the resistance of the Fe film. For Fe layers with a thickness of 12 nm and a resistance of $R_{xx} = 36\Omega$ the $\Delta L_{xy}^0$ increases roughly linear with the Cs thickness. The shape and in particular the width of the peak is independent of the Cs thickness, the maximum is always at about 0.03 atomic layers of Pb.

It is difficult at the present time to describe the observed AHC quantitatively
since there are too many unknown parameters. But there are a number of qualitative considerations that are useful.

We assume that an electron wave of spin up (down) is mainly specularly reflected at the interface which decreases its amplitude by a factor $a_i^{\uparrow}$ ($a_i^{\downarrow}$). Both spin directions are partially specularly reflected at the free surface with a change of amplitude by $b^s$. Fuchs [4] and Sondheimer [5] derived an expression for the conductivity of a thin film with the coefficient of specular reflection $p$ at the surfaces. By extending this theory to a film with specular reflection $p_i^{\uparrow,\downarrow} = |a_i^{\uparrow,\downarrow}|^2$ on one surface (the interface) and $p^s = |b^s|^2$ on the other surface one obtains for the charge current conductivity and the spin current conductivity

$$
\sigma_c = \sigma_{\uparrow} + \sigma_{\downarrow}
$$

$$
\sigma_s = (\sigma_{\uparrow} - \sigma_{\downarrow})
$$

where $\sigma_{\uparrow,\downarrow} = (n e^2/m) (l_{\uparrow,\downarrow}/v_F)$ are the conductivities for the two spin directions. Here the effective mean free path $l_{\uparrow,\downarrow} = l_{\uparrow,\downarrow} (p_i^{\uparrow,\downarrow}, p^s, l_0)$ is a non-analytic function of the two coefficients of specular reflection $p_i^{\uparrow,\downarrow}$ and $p^s$ and the bulk mean free path $l_0$. (We define here the spin current as the difference between the charge currents for spin up and spin down, $j_s = j_{\uparrow} - j_{\downarrow}$). As long as the spin current is not changed by the condensation of the Pb impurities we expect a monotonic increase of the AHC as a function of the Pb coverage which will be linear for small coverages. In the experiment we see a maximum of $L_{xy}^0$ at about 0.03 atomic layers of Pb. We can see two possible reasons why the spin current is reduced for larger Pb coverages:

(◦) **Spin rotation:** The spin-orbit scattering of the Pb rotates the spin quantization direction. The electrons are no longer in a spin up or down state but can have an arbitrary spin state $[\alpha_{\uparrow} |\uparrow\rangle + \alpha_{\downarrow} |\downarrow\rangle]$. Such an electron is specularly reflected from the interface with an amplitude $[a_{\uparrow} \alpha_{\uparrow} |\uparrow\rangle + a_{\downarrow} \alpha_{\downarrow} |\downarrow\rangle]$. Its coefficient of specular reflection is then $p_i^{\uparrow} = |a_{\uparrow} \alpha_{\uparrow}|^2 + |a_{\downarrow} \alpha_{\downarrow}|^2$. This has two important consequences: (i) the spin current is reduced and (ii) each interaction with the interface tries to restore the quantization in z-direction. If $|a_{\uparrow}^{\uparrow}|^2 > |a_{\downarrow}^{\downarrow}|^2$ then the spin of the specularly reflected part of the electron is rotated towards the up direction, while the diffusively scattered component of the electron is rotated towards the down direction.

Our group has measured the spin-orbit scattering cross section of Pb on the surface of Cs in the past [8]. We found a surprisingly large spin-orbit scattering
cross section of about $\sigma_{so} = 0.5\frac{4\pi}{k_F^2} \approx 1.5 \times 10^{-19} m^2$. This yields a spin-orbit scattering time of about $10^{-12} s$ for 0.01 atomic layer of Pb on the surface of Cs film with a thickness of 30 $nm$. This results in a spin-flip rate is about $2 \times 10^{12} s^{-1}$ which undermines the quatization of the spin in z-direction and tends to reduce the spin current. The spin-dependent reflection at the FeCs interface counteracts the randomization of the spin. Both effects together yield a dynamic spin distribution.

(⊙) **Diffuse surface scattering**: The condensation of (any) impurities onto the Cs surface causes dramatic increase in the resistance of the Cs film [7]. This reduces the coefficient of specular reflection $p^s$ on the free surface. In addition we have strong indications that the (local) conductance close to the surface is strongly reduced. One possible mechanism is that the surface impurities interact with the Cs atoms by Friedel oscillations and cause disorder within 2 - 3 $nm$ of the surface. This reduces strongly the charge and spin currents near the Pb atoms and therefore the AHC would be strongly reduced.

We have investigated which of the two mechanisms destroys the AHC at larger Pb coverages by using instead of Pb another weaker spin-orbit scatterer. If it is the spin-orbit scattering with its spin-flip processes that destroys the spin current then we expect a smaller initial slope of the AHC as a function of impurity coverage with a maximum at larger coverages.

We use Au impurities as an alternative spin-orbit scatterer. The nuclear charge of Au is similar to that of Pb but since it is mainly an s-scatterer its spin-orbit cross section is considerably smaller than that of Pb. We cover an FeCs sandwich ($d_{Cs} = 27.5 nm$) with sub-mono-layers of Au. In Fig.4 its AHC $L_{xy}^0$ is plotted as a function of the Au coverage. The sign of the AHC is reversed. This indicates that the asymmetry of the the Au and Pb scattering have opposite signs. Obviously the AHE yields more information about the spin-orbit scattering than the spin-orbit cross section $\sigma_{so}$.

The Au impurities are less effective by a factor of four than the Pb impurities. This confirms that the spin-orbit scattering of Au is smaller than that of Pb. However, the extremum is for both impurities essentially at the same coverage of about 0.03 atomic layers. This favors the explanation that the impurity coverage reduces the coefficient of specular reflection at the free surface and (or) that the impurities strongly reduce the local conductance close to the surface.

In this paper we have covered sandwiches of FeCs with a few hundredth of a mono-layer of Pb and observed a large anomalous Hall conductance. We can explain the observed phenomena when we assume that the conduction electron in the Cs have a different mean free path for spin up and down because their reflection
at the interface has different degrees of specularity. This yields a spin current in
the Cs and the strong spin-orbit scattering of the Pb detects the spin current
as an AHC. At the present time the Pb impurities represent only a qualitative
indicator of the size of the local spin current. If the different phase shifts $\delta_{j,l}$
with $j = l \pm \frac{1}{2}$ were reliably calculated for Pb impurities on (in) Cs one could
quantitatively determine the size of the local spin-current at the Cs surface using
the results of ref. [3]. This system has the potential for further experiments
with spin currents in a non-magnetic metal. The basic idea of sandwiching a
ferromagnetic and a non-magnetic metal separated by a barrier may be used to
prepare other interesting systems.

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2. Figures

Fig.1: The mean free path of a K film quench-condensed on Fe as a function of the K thickness.
Fig. 2: The anomalous Hall resistance of the FeCs sandwich before and after coverage with 0.02 atomic layers of Pb. (The normal linear Hall resistance is subtracted).

Fig. 3: The anomalous Hall conductance $L_{xy}^0$ of an FeCsPb and an FeCsAg sandwich as a function of the impurity coverage (Pb or Ag given in atomic layers).
Fig.4: The additional anomalous Hall conductance $L_{xy}^0$ of an FeCsAu sandwich as a function of the Au coverage.
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3. Rest

Spin-orbit impurities scatter conduction electrons with a left-right asymmetry in the scattering amplitude. This asymmetry has the opposite sign for opposite spins. As long as both spin orientation contribute equally to the current they do not yield an anomalous "Charge" Hall effect. (However, they yield opposite gradients in the chemical potential for the two spin orientations).

The Fe layer has, of course, a spin polarization and spin current. They cause the AHC of the original Fe film. However, the spin polarization of the Fe should not extend deeply into the Cs film, even in the presence of a current. Furthermore one would expect that any polarization at the free Cs surface by the Fe would decrease with increasing Cs thickness.
4. Theory

The modeling of the observed AHE has to overcome a number of obstacles

- The degree of specular reflection for spin up and down on the FeCs interface and at Cs surface are not known.
- For the calculation of the conductance one has to use the Fuchs-Sondheimer theory which yields non-analytic expression for the conductance.
- Only for zero (or small) concentration of the spin-orbit scatterer Pb the spin of the conduction electrons in the Cs is a good quantum number.
- The structural changes of the Cs surface due to the Pb impurities is not well known and understood.

We strongly simplify the problem in the following way. Since a perfect reflection at the interface or surface of the Cs does not reduce the conductance we replace a slightly imperfect reflection by a scattering lifetime. If $\delta^i_\uparrow$ is the fraction of spin up electrons which is not specularly reflected at the interface then $\tau^i_\uparrow = \delta^i_\uparrow \frac{2d_{Cs}}{v_{Cs}}$ where $d_{Cs}$ is the thickness of the Cs film and $\overline{v_{Cs}}$ is the average of the electron velocity perpendicular to the surface. Similar we define scattering lifetime at the surface (where both spins are equally scattered) $\tau^s = \delta^s \frac{2d_{Cs}}{v_{Cs}}$. The scattering lifetime in the interior of the Cs films is $\tau_0$. Then the lifetime for spin up electrons is

$$\frac{1}{\tau_\uparrow} = \frac{1}{\tau_0} + \delta^i_\uparrow \frac{\overline{\text{v}}}{d_{Cs}} + \delta^s \frac{\overline{\text{v}}}{d_{Cs}}$$

$$\tau_\uparrow = \frac{d_{Cs}/\overline{\text{v}}}{\tau_0 + \delta^i_\uparrow + \delta^s}$$

$$G = \frac{ne^2}{m} d_{Cs} (\tau_\uparrow + \tau_\downarrow)$$

and the spin current density in the presence of an electric field $E$

$$j_{\text{spin}} = \frac{ne}{m} (\tau_\uparrow + \tau_\downarrow) E$$

$$\frac{1}{1+x} - \frac{1}{2+x}$$
Our group has measured the spin-orbit scattering cross section of Pb on the surface of Cs in the past [8]. We found a surprising large spin-orbit scattering cross section of about $\sigma_{so} = 0.5* \frac{4\pi}{k_F^2} \approx 1.5 \times 10^{-19} m^2$. This yields a spin-orbit scattering time of about $10^{-12}$ s for 0.01 atomic layer of Pb on the surface of Cs film with a thickness of 30 nm. This results in a spin-flip rate is about $2.5 \times 10^{12}$ s$^{-1}$ which undermines the quantization of the spin in z-direction and tends to reduce the spin current. The spin dependent reflection at the FeCs interface counteracts the randomization of the spin. If for example the specular reflection of the spin up electrons is larger than the for the spin down electrons then each interface reflection rotates the spin of the specular reflected electrons towards the up direction and the spin of the diffusely scattered part into the down direction.

We assume that the interface with the Fe reduces the amplitude in the reflection by the factor $e_{\pm}$ and $e_{-}$ for spin up and spin down. Now we consider an electron wave $\psi_i = |k\rangle \chi$ with the spin function

$$\chi = \alpha |\uparrow\rangle + \beta |\downarrow\rangle$$

After the reflection one obtains

$$\psi_o = e_+ \alpha |\uparrow\rangle + e_- \beta |\downarrow\rangle$$

Therefore the specular part of the reflection is $[|e_+ \alpha|^2 + |e_- \beta|^2]$ and the spin of the wave changed from $[|\alpha|^2 - |\beta|^2]$ to $[(|e_+ \alpha|^2 - |e_- \beta|^2) / (|e_+ \alpha|^2 + |e_- \beta|^2)]$. If we set $e_{\pm} = e (1 \pm \Delta)$ then we obtain for the probability

$$[|e_+ \alpha|^2 + |e_- \beta|^2]$$
\[ (e (1 + \Delta) \alpha)^2 + (e (1 - \Delta) \beta)^2 = e^2 + 2e^2 (\alpha^2 - \beta^2) \Delta + e^2 \Delta^2 \]

and for the final spin

\[
\frac{((e (1 + \Delta) \alpha)^2 - (e (1 - \Delta) \beta)^2)}{((e (1 + \Delta) \alpha)^2 + (e (1 - \Delta) \beta)^2)} = (\alpha^2 - \beta^2) + 8\alpha^2 \beta^2 \Delta - 16\alpha^2 \beta^2 (\alpha^2 - \beta^2) \Delta^2
\]

\[= (\alpha^2 - \beta^2) + 8\alpha^2 \beta^2 \Delta (1 - 2\Delta (\alpha^2 - \beta^2))\]

We use the linear part.

The electron with the opposite spin

\[ \chi = \beta |\uparrow\rangle - \alpha |\downarrow\rangle \]

is transferred into

\[ = e_+ \beta |\uparrow\rangle - e_- \alpha |\downarrow\rangle \]
\[= e [e_+ \beta |\uparrow\rangle - e_- \alpha |\downarrow\rangle] \]

with the probability

\[ \frac{|e_+ \beta|^2 + |e_- \alpha|^2}{|e_+ \beta|^2 + |e_- \alpha|^2} = (e (1 + \Delta) \beta)^2 + (e (1 - \Delta) \alpha)^2 \]
\[= e^2 - 2e^2 (\alpha^2 - \beta^2) \Delta + e^2 \Delta^2 \]

and the expectation value of \( s_z \) equal to

\[ \frac{|e_+ \beta|^2 - |e_- \alpha|^2}{|e_+ \beta|^2 + |e_- \alpha|^2} \]
\[= \frac{(e (1 + \Delta) \beta)^2 - (e (1 - \Delta) \alpha)^2}{(e (1 + \Delta) \beta)^2 + (e (1 - \Delta) \alpha)^2} \]
\[= - (\alpha^2 - \beta^2) + 8\alpha^2 \beta^2 \Delta + 16\alpha^2 \beta^2 (\alpha^2 - \beta^2) \Delta^2 \]
