Spin-orbit scattering of Pb and Bi impurities in Cs, K, and Na films

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The spin-orbit scattering (SOS) of the Pb and Bi impurities in Cs, K, and Na films is measured by means of weak localization. The dimensionless SOS cross section for Pb in Cs (in units of $4\pi/k_F^2$) has the large value of $\sigma_{so}^* = 1/7$. This SOS cross section is the absolute maximum possible for $(s,p)$ impurities (if one neglects $d$ and larger $l$ scattering). The SOS contributes almost 50% to the total elastic scattering. [S0163-1829(99)01147-9]

I. INTRODUCTION

The alkali metals are described in the majority of solid-state text books as the best examples of a nearly free-electron system. DeHaas-van Alphen measurements$^{1,2}$ yield Fermi surfaces for the alkali metals that deviate very little from a perfect sphere, for example for Na, by less 0.1%. Measurements of the transport properties yield, however, a mixed picture. While some can be described with the free electron model, others cannot. Overhauser argues that the Coulomb interaction causes an instability in the electron system of the alkali metals, which results in macroscopic charge-density waves, and he has collected a large body of experimental data, which support his model.$^4$

Our group discovered recently a number of surprising properties of thin Cs films.$^{5,6}$ As an example: if a Cs film, 8.2-nm thick, is covered with 0.1 atomic layers of In, then the resistance increases by 58%, the Hall constant by 15%, and the electron dephasing rate by 15%. The origin of this behavior is unknown. A detailed discussion of various attempts to explain this unusual behavior is given in Ref. 5.

Furthermore, at 4.5 K the magneto-resistance experiments showed an almost linear magnetoresistance for fields between 2 and 7 T, which could not be explained. Measurements of the induced Hall effect, which should measure the ballistic propagation of the conduction electrons, yielded results in Cs, which strongly contradict the free electron model for Cs.

In this paper, we investigate the spin-orbit scattering cross section of Pb in Cs. Since our experiments show an extraordinarily large spin-orbit scattering for this system we also include the neighboring systems, Pb in K and Na, and Bi in Cs and K.

II. EXPERIMENTAL RESULTS AND EVALUATION

The Cs, K, and Na films are evaporated from SAES-Getters evaporation sources. They are quench condensed onto a substrate at He temperature in an ultrahigh vacuum of better than $10^{-11}$ torr. After condensation the films are annealed for several minutes at 40 K. Then the magneto-resistance is measured in the field range between $-7T < B < +7T$ at several temperatures between 4.5 and 20 K. Furthermore the resistance is measured as a function of temperature between liquid helium temperature and 25 K. Then the surface impurities are condensed on top of the alkali film. As an example, we discuss the investigation of a Cs film with Pb impurities.

• A Cs film of about 10.0 nm thickness is condensed and investigated.
• The Cs film is covered with about 0.005 atomic layers of Pb.
• The Cs/Pb “sandwich” is covered with about 5.0 nm of Cs.
• 0.005 atomic layers of Pb are condensed on top of the Cs.
• 5.0 nm of Cs are condensed.
• 0.005 atomic layers of Pb are condensed on top of the Cs.
• Finally, 5.0 nm of Cs are condensed on top of the Cs.

After each condensation the sandwich is annealed to 35 K, and the magneto-resistance of the sandwich is measured. The data of this experiment are collected in Table I. In Fig. 1, the magneto-resistance of the first three films is plotted at 4.5 K. The scales of the magnetic field, the resistance $\Delta R$ and the normalized conductance $\Delta L/L_{00}$ ($L_{00} = e^2/2\pi^2k_B$) are shown individually beside the curves. One recognizes that the Pb impurities introduce a spin-orbit scattering minimum at $B = 0$. The full curves show the theoretical fit with the theory by Hikami et al.$^7$ This fit yields the characteristic fields $H_I$ and $H_{so}$, which represent the dephasing rates $1/\tau_I$ and the spin-orbit scattering rate $1/\tau_{so}$. The relation between the inelastic rate and the field $H_I$ is given by

$$\frac{1}{\tau_I} = H_I \frac{4eD}{\hbar} = H_I \frac{4}{e}\hbar R dN,$$

where $D$ is the diffusion constant of the conduction electrons, $R$ is the resistance of the film, $d$ is the thickness of the film, and $N = dn/dE$ is the electron density of states at the Fermi surface. The dephasing rates of the Cs with different Pb impurities is shown in Table I.

The spin-orbit scattering rate can be similarly calculated from $H_{so}$. For the pure Cs film we find no indication of a spin-orbit scattering. A careful analysis of the data shows that the largest possible value for $H_{so}$ is 0.01 T. This value is smaller by a factor 20 than the $H_{so}$ value with 0.005 atomic layers of Pb on top of the Cs (film number 2). Since the value of the intrinsic spin-orbit scattering field is proportional to the square of the resistivity of the host metal (Cs) it can be neglected for all the investigated sandwiches. For the
Cs films with Pb impurities the agreement between experiment and theory is far from perfect beyond the maximum (in contrast to other quench condensed films). Since we do not know the origin of this derivation at the present time, it introduces an error in the evaluation of the spin-orbit scattering field $H_{so}$. The size of the error is difficult to judge, and we use a conservative estimate of 10% error. Together with the error in the Pb coverage this yields a total error of about 30% for the spin-orbit scattering cross section.

For Cs films with a resistance of the order of 10 Ω or less the contribution of weak localization to the magnetoresistance becomes smaller than other contributions (among others the classical quadratic magnetoresistance). As a consequence the fit with the Hikami theory becomes poorer, even if a quadratic magnetoresistance is included, and the spin-orbit scattering field $H_{so}$ can no longer be fitted reliably.

From the spin-orbit scattering field $H_{so}$ we calculate the dimensionless spin-orbit scattering cross section $\sigma_{so}^*$ as $\sigma_{so}^* = \alpha_{so} k_F^2 / 4 \pi$.

$$\sigma_{so}^* = \frac{\pi \Omega_i H_{so}}{d_i e R}, \tag{2.2}$$

where $\Omega_i$ is the atomic volume of the impurity, $d_i$ is the coverage of the impurity (in metric units), and $R$ is the resistance per square. It is interesting to note that the thickness of the host film does not appear in this formula.

The values of $\sigma_{so}^*$ are collected in Table I for the different stages of the Cs/Pb sandwich. When the Pb is condensed onto the Cs we have Pb surface impurities. After the coverage with 5.0 nm of Cs we have bulk Pb impurities. The values in Table I give always the contribution of the last evaporated Pb atoms. The value of $\sigma_{so}^*$ lies at about 0.6 for the Pb on the surface of the Cs and in the bulk.

The large value of $\sigma_{so}^*$ for Pb impurities in Cs represents another challenge to the free electron model of Cs. Let us for a moment consider Cs as a free electron system. The Pb impurities are $(s,p)$ atoms and their scattering should be mainly $s$ and $p$ scattering. The $d$-scattering amplitude should be rather small, and we neglect the contribution of $d$ scattering and any higher angular momentum.

The spin-orbit scattering cross section is given by the formula

$$\sigma_{so}^* = \sum_{l=1}^{\infty} \frac{l(l+1)}{2l+1} \sin^2(\delta_{l+1/2,l} - \delta_{l-1/2,l}), \tag{2.3}$$

where $l$ is the angular momentum of the scattered wave, $\delta_{l+1/2,l}$ is the phase shift of a spherical electron wave with total angular momentum $j = l \pm \frac{1}{2}$ and orbital angular momentum $l$. The $s$ component of the scattering amplitude yields zero contribution to the spin-orbit scattering (see Eq. (2.3)). Therefore, in this model, only the $p$ scattering contributes to $\sigma_{so}^*$. The maximum scattering cross section for $p$ scattering is $l(l+1)/2l+1 = 2/3$. We find that our experimental result corresponds to the absolute maximum of $p$ scattering. The phase shift between $\delta_{3/2,1}$ and $\delta_{1/2,1}$ has to be of the order of $\pi/2$.

Papanikolaou et al. performed a first principle calculation of the SOS cross section $\sigma_{so}^*$ of $(s,p)$ impurities in Mg, which agreed rather well with the experimental results. It is, however, much harder to extend these calculations to Pb impurities in Cs. The large charge of the Pb nucleus requires a relativistic calculation. Nevertheless, we have serious doubts that the spin-orbit potential can yield such a large value for the phase shift difference.

If we calculate the SOS cross section $\sigma_{so}$ in units of nm$^2$ we find $\sigma_{so} = \sigma_{so}^* 4 \pi/k_F^2 = 0.19$ nm$^2$. This is a remarkably

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**TABLE I.** The parameters of the CsPb film. The thickness for the Cs is given in nm and the Pb coverage is given in atomic layers. The other columns give the resistance of the film, the inelastic field $H_i$, the spin-orbit scattering field $H_{so}$, the inelastic rate $1/\tau_i$ and the dimensionless spin-orbit scattering cross section $\sigma_{so}^*$ of the last Pb layer.

<table>
<thead>
<tr>
<th>Metal</th>
<th>Thickness</th>
<th>$R$ [Ω]</th>
<th>$H_i$ [T]</th>
<th>$H_{so}$ [T]</th>
<th>$1/\tau_i$ [OS$^{-1}$]</th>
<th>$\sigma_{so}^*$</th>
<th>Position</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cs</td>
<td>9.8 nm</td>
<td>150</td>
<td>0.057</td>
<td>$&lt;0.01$</td>
<td>0.17</td>
<td>0</td>
<td>Surface</td>
</tr>
<tr>
<td>Pb</td>
<td>0.0050 a.l.</td>
<td>176</td>
<td>0.051</td>
<td>0.23</td>
<td>0.14</td>
<td>0.49</td>
<td>Surface</td>
</tr>
<tr>
<td>Cs</td>
<td>4.95 nm</td>
<td>22.5</td>
<td>0.0148</td>
<td>0.0405</td>
<td>0.20</td>
<td>0.67</td>
<td>Bulk</td>
</tr>
<tr>
<td>Pb</td>
<td>0.0050 a.l.</td>
<td>24.3</td>
<td>0.0148</td>
<td>0.087</td>
<td>0.19</td>
<td>0.67</td>
<td>Surface</td>
</tr>
<tr>
<td>Cs</td>
<td>5.01 nm</td>
<td>10.8</td>
<td>0.0098</td>
<td>0.038</td>
<td>0.21</td>
<td>0.64</td>
<td>Bulk</td>
</tr>
<tr>
<td>Pb</td>
<td>0.0050 a.l.</td>
<td>11.5</td>
<td>0.010</td>
<td>0.067</td>
<td>0.20</td>
<td>0.83</td>
<td>Surface</td>
</tr>
<tr>
<td>Cs</td>
<td>5.11 nm</td>
<td>6.9</td>
<td>(0.007)</td>
<td>(0.033)</td>
<td>0.18</td>
<td>(0.43)</td>
<td>Bulk</td>
</tr>
</tbody>
</table>

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**FIG. 1.** The magneto-resistance of a pure Cs film (top), Cs with a cover of 0.005 atomic layers of Pb (middle) and a Cs/Pb$_{1-x}$/Cs sandwich (bottom). The symbols are the experimental results, the full curves are the best theoretical fits.
large cross section. The physical cross section of a Pb atom is \( \pi r_p^2 \) whose value lies between 0.045 and 0.07 nm\(^2\) depending on whether one takes the covalent or the ionic radius of Pb\(^{2+}\) for the Pb impurities. \( \sigma_{so}^* \) is roughly three time the physical cross section of the Pb impurities. This demonstrates the remarkable size of the spin-orbit scattering cross section. Another manifestation of the large size of the SOS is the ratio between the SOS cross section and the transport cross section of the Pb. When we condense the Pb on the surface of the Cs the resistance of the Cs increases. We may take a rather naive point of view and attribute the increase in resistance to transport scattering cross section of the Pb impurities. Then, we obtain values for the dimensionless transport cross section of \( \sigma_{tr}^* \) between 1.2 and 2 (with exception of the first condensation of Pb where the Cs might undergo structural changes). This means that the SOS cross section is about \( \frac{1}{3} \) to \( \frac{1}{4} \) of the transport cross section.

We measured recently the SOS cross section of many impurities in Mg.\(^6\) The value of \( \sigma_{so}^* = 0.6 \) for Pb in Cs is about a factor 10 larger than the corresponding value of Pb in Mg. The physical spin-orbit scattering cross section \( \sigma_{so} = (4 \pi/k_F^2)\sigma_{so}^* \) for Pb in Cs is even a factor 40 larger than the corresponding value of Pb in Mg (because of the square of the Fermi wave number in the denominator).

In Table II we have collected the dimensionless SOS cross sections \( \sigma_{so}^* \) for Pb in Cs from five different experiments. The agreement between the different experiments is generally considerably better than the conservative error of 30%.

Similar experiments were performed for the systems: Pb on and in K, Pb on and in Na, Bi on and in Cs, Bi on and in K. The results are also included in Table II.

The Pb impurities on the surface and in the bulk of the alkali metals clearly have a value considerably larger than for Bi impurities. (This is different for a Mg host).

A relativistic first-principle calculation for the SOS cross section of Pb in Cs is very desirable. However, in absence of such a calculation we can at the present time only speculate about the size and origin of the experimentally determined SOS cross section. We doubt very much that a first-principle calculation treating the Cs as a free electron system can reproduce such a large value for the Pb impurity. We rather believe that the Pb impurities displace the Cs atoms so that the Cs itself contributes to the value of \( \sigma_{so}^* \). One possible scenario is that the Pb generates spherical charge density waves. At short distances they correspond to Friedel oscillations. At large distances the Coulomb interaction might stabilize the amplitude of the charge density wave, in analogy to Overhauser’s plane charge densities. In such a case the ‘‘impurity’’ is much more extended, and therefore large angular momenta can contribute to the SOS cross section in Eq. (2.3).

Another interesting question is what the large SOS does to the spin polarization of the conduction electrons. Originally the spin-orbit interaction is spherically symmetric (in the bulk). However, a spontaneous breaking of the rotational symmetry cannot be excluded.

**ACKNOWLEDGMENTS**

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**TABLE II.** The dimensionless spin-orbit scattering cross section \( \sigma_{so}^* \) for Pb and Bi impurities on the surface and in the bulk of Cs, K, and Na.

<table>
<thead>
<tr>
<th>Host</th>
<th>Imp.</th>
<th>Pb</th>
<th>Bi</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cs</td>
<td>Surface</td>
<td>0.6</td>
<td>0.13</td>
</tr>
<tr>
<td></td>
<td>Bulk</td>
<td>0.6</td>
<td>0.13</td>
</tr>
<tr>
<td>K</td>
<td>Surface</td>
<td>0.35</td>
<td>0.15</td>
</tr>
<tr>
<td></td>
<td>Bulk</td>
<td>0.46</td>
<td>0.22</td>
</tr>
<tr>
<td>Na</td>
<td>Surface</td>
<td>0.19</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Bulk</td>
<td>0.21</td>
<td></td>
</tr>
</tbody>
</table>

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