

## OBSERVATION OF A RESONANCE IN THE SPIN–ORBIT SCATTERING OF $5(s, p)$ IMPURITIES IN Mg AND Cu

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(Received 8 March 1993 by G. Guntherodt)

The spin–orbit scattering (SOS) cross section  $\sigma_{so}$  of the  $5(s, p)$  impurities is investigated in two different metal hosts. In the simple Mg host we observe a maximum at half filled  $p$  shell. This is the first experimental observation of an impurity  $p$  resonance and confirms theoretical predictions. For the transition metal host Cu the maximum in  $\sigma_{so}$  is shifted towards Te, the impurity with four  $p$  electrons. This is due to the hybridization of the Cu  $d$  states with the Te  $p$  level. The comparison between experimental and theoretical SOS cross sections represents a compact test of the quality of modern self-consistent electronic structure calculations.

THE SPIN–ORBIT interaction plays an important role in solid state physics. It leads to a coupling between the coordinates in real space and in spin space, and its strength determines whether the electron spin is a good quantum number.

Slichter and coworkers [1] studied the spin-orientation lifetime of conduction electrons in dilute Li and Na based alloys. Measuring the effect of various concentrations of impurities in the linewidth of the conduction-electron spin resonance (CESR) they determined the impurity contribution to the spin-lattice relaxation rate and deduced the SOS cross section. Their results were analyzed in the light of model calculations [1] and a simple resonance formula [2] was found to fit reasonably well the experimental data for impurities within a given  $(s, p)$  series.

Recent experimental investigations of the SOS of  $(s, p)$  impurities in Mg, using the method of weak localization, showed a strong dependence of the SOS cross section  $\sigma_{so}$  on the valence and the atomic number  $Z$  of the impurity [3]. For impurities within a given group of the periodic table the results followed a power law  $\sigma_{so} \propto Z^p$  ( $p \simeq 5$  for the noble metal impurities) and a strong increase of  $\sigma_{so}$  with increasing valence was observed.

Theoretical calculations of the SOS cross section of these systems were performed within the framework of the self-consistent local density functional (SCLDF) theory [4]. The jellium model was used for the Mg host and the spin–orbit interaction was treated as a perturbation of the non-relativistic Hamiltonian. Therefore, these calculations are restricted to the  $4(s, p)$  and  $5(s, p)$  impurities where relativistic effects are of minor importance. The agreement between the theoretical and the experimental results was surprisingly good, the experimental cross sections lying about 30% below the theoretical results with the exception of the impurities Cu and Ag. The noble metal impurities have a full  $d$  resonance just below the Fermi energy  $E_F$  and it is well known that the SCLDF theory has difficulties to give the correct position of (almost) full bands [5] and localized resonance states [6].

Furthermore the theory made an interesting prediction. For impurities with more than a half-filled  $p$  shell the SOS cross section should decrease, since a  $p$  resonance crosses through the Fermi level of the host and the SOS cross section depends quadratically on the impurity local density of states (LDOS) at  $E_F$ . Stimulated by this theoretical prediction we extended the investigation of the

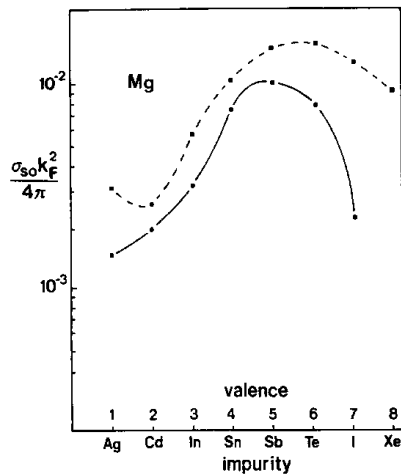


Fig. 1. The spin-orbit scattering cross section of  $5(s, p)$  impurities in Mg. The full points are the experimental results, and the squares give the theoretical results using a jellium for Mg.

$5(s, p)$  impurities to the valence seven, i.e. the impurity *I*. In addition we measured and calculated the SOS cross section of the  $5(s, p)$  impurities in a Cu host.

The experimental procedure is essentially the same as reported in [3]. First we condensed a thin film of about 30 at. layers of Mg onto a quartz plate which is kept at helium temperature. Then we evaporated on top of the Mg film a fraction of a monolayer of the impurity. The thickness of the impurity layer was between 0.1 and 0.05 at. layers. To obtain the desired coverage of the impurity, the impurity material was heated in a tantalum or tungsten boat and its evaporation rate was determined over a period of several minutes. An evaporation rate of a few tenths of a monolayer per minute was chosen. Then the shutter was electronically opened for the appropriate time so that the desired coverage was achieved. The accuracy of the impurity coverage is estimated to be about  $\pm 20\%$ .

In Fig. 1 we have plotted the renormalized SOS cross section  $\sigma_{so} k_F^2 / 4\pi$  for the  $5(s, p)$  impurities in Mg. This dimensionless quantity corresponds to a sum over phase shifts [3] and is well suited for a comparison between experiment and theory. The full points represent our experimental results (some of these data have already been published earlier). The dashed curve gives the theoretical results. One recognizes in Fig. 1 that the agreement between the experiment and theory is restricted to the valence range between 2 and 5. For valences 6 and 7, i.e. Te and *I* the experimental results are much smaller than the theoretical prediction.

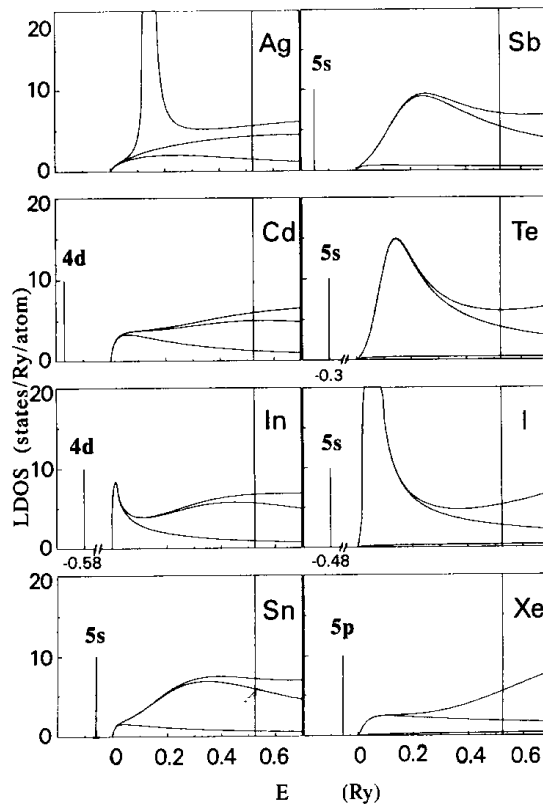


Fig. 2. The local density of states ( $s, s+p, s+p+d$  from bottom to top) of  $5(s, p)$  impurities in a Mg-jellium. The bound semicore states are shown as vertical bars and the attached number gives their position. The vertical lines on the abscissa indicate the Fermi energy ( $E_F = 0.524$  Ry).

We believe that this reduced agreement for higher valence indicates that the SCLDF theory is less accurate for higher valences. As shown in Fig. 2 the  $p$ -resonance of Te and *I* lie clearly below the Fermi energy of Mg and in both cases it is almost completely filled. This situation is somewhat similar to the case of a Ag impurity in Mg where the  $d$  resonance is almost completely filled. It is documented from other examples that the SCLDF theory tends to misplace the position of full bands [5] and localized resonance states [6]. The disagreement between the experimental and theoretical SOS cross section for full or almost full resonances appears to indicate that the position of the  $p$  resonance might be slightly lower than the SCLDF theory suggests. On the other hand also lattice relaxation effects around the  $5(s, p)$  impurities should be important due to their large size. An outward displacement of the host neighbor atoms would lead in this case to sharper  $p$  impurity resonances and the  $p$  local density of states at  $E_F$  would further decrease.

In addition we wished to investigate another host

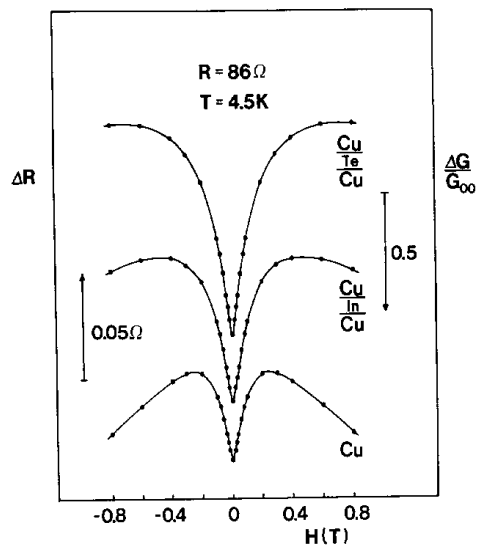


Fig. 3. The magneto-resistance curves of a pure Cu film, a Cu film with 0.4 at. layers of In and a Cu film with 0.2 at. layers of Te (both covered with 5 at. layers of Cu).

metal besides Mg as well. However, the choice is somewhat restricted since the main requirement is that the intrinsic SOS of the host must be small. That restricts us to metals below or at most in the  $n = 4$  row. Although several choices of the host metal might be interesting a noble metal host is the simplest transition metal and will most likely introduce some new physics compared with the simple (*s, p*) hosts. Therefore, we selected Cu as a second host in which we investigated the SOS cross section of the 5(*s, p*) impurity series.

However, the SOS of quench condensed Cu films with a thickness of about 60 Å and a resistance per square of the order of 100–120 Ω is already about 10 times larger than that in Mg. To obtain optimal accuracy in the additional SOS of the impurity we preferred to have a similar additional SOS from the impurity. This requires, however, an almost ten times larger concentration of the impurity atoms than in Mg. Figure 3 shows the magnetoresistance curves of a pure Cu film, a Cu with 0.4 at. layers of In and a Cu film with 0.2 at. layers of Te. Since the interlayer of In with a thickness of 0.4 at. layers can hardly be considered as single non-interacting impurities we became concerned that the SOS of the individual In atom might be altered by the contact with other In atoms. Therefore, we performed a control experiment in which an In interlayer of only 0.1 at. layers was used. Of course, in this experiment the SOS contribution of the In was much smaller than that of the Cu background. The resulting SOS cross section agreed within 3% with the former In

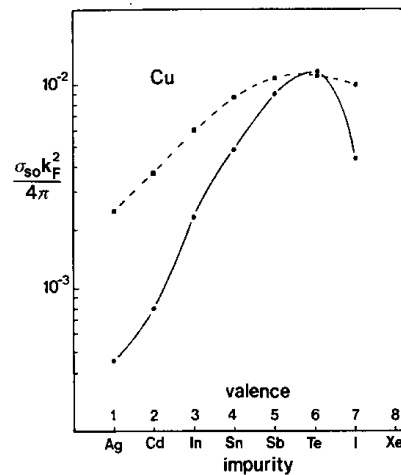


Fig. 4. The spin-orbit scattering cross section of 5(*s, p*) impurities in Cu. The full points are the experimental results, and the squares give the theoretical results using a jellium for Cu.

experiment (interlayer thickness of In 0.4 at. layers). For Ag, where  $\sigma_{so}$  is much smaller we investigated in one experiment a Cu film with an Ag interlayer of 0.75 at. layers, and in a second experiment an Ag interlayer of 0.2 at. layers. Again the agreement was better than 3%. This test reassures us that the proximity of other impurity atoms does not change  $\sigma_{so}$  of the impurity.

In Fig. 4 the renormalized SOS cross section  $\sigma_{so} k_F^2 / 4\pi$  of the 5(*s, p*) impurity series in Cu is plotted as a function of the valence of the impurity. The full circles are the experimentally determined values. The SOS cross section shows a clear maximum, but for the Cu matrix the maximal value of  $\sigma_{so}$  lies at the valence of 6, i.e., at the Te impurity. The dashed curve gives the result of a first theoretical calculation. Here the Cu matrix is treated as a jellium and the intrinsic spin-orbit interaction in the Cu host is neglected. (As we will discuss below a realistic calculation is much more demanding and is planned for the future). The experimental curve shows a much stronger dependence of  $\sigma_{so}$  on the valence than the theoretical calculation. In particular the SOS cross section for Ag and Cd impurities is much smaller than for the other impurities and far below the theoretical results.

While the impurities in Mg can be essentially treated as being dissolved in a jellium the situation is considerably more complicated in a Cu host. First, in Cu one has to include the properties of the Cu *d* electrons. As a result of the hybridization between the low lying Cu *d* band and the impurity *p* states the *p* resonance is repelled to higher energies. This presumably explains why the maximal value of  $\sigma_{so}$

occurs at the valence of 6 instead of 5 as reported above. Furthermore, the Cu atoms themselves have a considerable spin-orbit interaction. Therefore, the effective SOS potential at an impurity atom is actually the difference between the SOS scattering potential of the impurity and that of the replaced Cu atom. (The reason is that the Cu atoms even in a disordered film are arranged to a first approximation in a periodic lattice. In an ideal crystal the electron states are described by Bloch states which are eigenfunction of the fully relativistic Hamiltonian. Therefore, the Bloch-functions experience no spin-orbit scattering; only deviations from the periodicity cause the observed background SOS of the Cu film.) For example, if we replace one Cu atom by a light atom with a weak spin-orbit scattering potential then the Bloch wave-functions of the host experience the deviation from the periodic potential as the scattering potential. With respect to the spin-orbit scattering the deviation from the periodic potential is just the full spin-orbit scattering potential of a single Cu atom. Therefore we expect in this approximation for the light atom roughly the same SOS cross section as a single Cu atom would have in a jellium.

To verify this analysis we investigated the SOS of Mg impurities in a Cu host. Mg is a very light metal and a Mg impurity in a "Cu-jellium" should have a very small SOS cross section. We calculated  $\sigma_{so}k_F^2/4\pi$  for Mg in a jellium having the electron density of Cu and obtained a very small value of  $7 \times 10^{-6}$ . Furthermore we performed a similar experiment as above with an interlayer of 0.2 at. layers Mg in a Cu film. We obtained an effective SOS cross section of  $3.5 \times 10^{-23} \text{ m}^2$  and the corresponding value of  $\sigma_{so}k_F^2/4\pi$  is  $5.1 \times 10^{-4}$ . (This is even larger than the value for Ag impurities in a Cu host.)

A further (necessary) improvement would be to

include the change of the Bloch wave-functions at the Cu neighbors of the impurity. A modified wave-function at the Cu neighbors will be scattered by the spin-orbit potential of the Cu neighbors. In addition, as we mentioned above, lattice relaxation effects around the 5(*s, p*) impurities in Cu should be important due to the extremely large size differences and therefore they should be taken into account in a theoretical calculation. Unfortunately this is a very demanding theoretical program. But the experimental results and their comparison with the jellium model indicate that one has to attempt this approach. If one succeeds theoretically then the measurement of the SOS cross section and the comparison with the theory provides us with a promising method to check the modification of the electronic wave-function in a direct vicinity of an impurity

*Acknowledgement* — The research was supported by National Science Foundation Grant No. DMR-8521662.

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