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Forced localization in thin K films, investigated with the superconducting proximity effect

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Abstract. – Thin films of alkali metals are forced into an insulating state by being covered with sub-mono-layers of Pb. The superconducting proximity effect is used to investigate the electronic change in the alkali film. On the length scale of the film thickness the electronic properties of the alkali film do not change noticeably during the metal-insulator transition.

In recent years, we have studied thin films of alkali metals with spin-orbit impurities, magnetic impurities and (s,p)-impurities and observed a number of surprising phenomena [1, 2]. Among the different results there was one finding that was particularly puzzling. When we covered a Cs film with sub-mono-layers of impurities the resistance of the film increased dramatically [1]. We tried many different impurities such as Pb, In, Au, Ag, etc. They produced essentially the same result. In fig. 1 the effect of In surface impurities on the conductance of a Cs film is shown ($d_{\mathrm{Cs}} = 8.2\,\mathrm{nm}$). In addition to the resistance, the Hall constant increased as if the density of conduction electrons decreased or the thickness of the alkali film was reduced.

This observation raises the question whether it is possible to transform an alkali film into an non-conducting state by covering it with impurities. Indeed we observed such an effect in thin layers of Cs and K covered with Sb or Pb. We first condensed a 3.2 nm thick insulating film of MgF_2 (at helium temperatures) onto a quartz plate. On top of the fresh substrate a K film of 6.5 nm (about 18 atomic layers) and a resistance of $1082\,\Omega$ was condensed. Then the K was covered with sub-mono-layers of Pb. In table I the resistance is given as a function of the Pb coverage. For a Pb coverage between 1.0 and 1.5 atomic layers, the resistance diverges and the K becomes insulating. This is a remarkable phenomenon. We believe that we observe a forced metal-insulator transition in the alkali film.

A devil's advocate might say that together with the In, some O_2 is introduced into the system, oxidizing the K so that the effective thickness of the K is reduced. However, our quench condensed alkali films are very clean. All the evaporation sources are surrounded with liquid N_2 and the vacuum in our system is better than 10^{-11} torr. If the K is condensed onto a thin (very disordered) Fe film, the K possesses mean free paths up to 100 nm. The fact that the K film condensed on MgF_2 has a resistance of $1082\,\Omega$ indicates that the MgF_2 underneath

TABLE I – *The resistance of a* K *film as a function of the* Pb *coverage.*

$\boldsymbol{d}_{\mathrm{Pb}}$ (a.l.)	$\boldsymbol{R}(\Omega)$
0.0	1082.
0.018	1234.
0.048	1549.
0.1	2110.
0.21	3200.
0.51	5621.
1.0	25860.
1.51	∞

the K has the same effect on the conductance as the Pb on top of the K. In the following experiment we show that the electron density of states remains essentially unchanged during this metal-insulator transition.

This experiment investigates the transport properties together with the superconducting proximity effect. We prepare a sandwich by the following sequence of evaporations: a) An insulating film of 10 atomic layers of amorphous Sb. b) On top of the amorphous Sb, a Pb film with a thickness of 6 nm and a resistance of 33. Ω is condensed (the freshly condensed Sb substrate makes it possible to obtain a homogeneous flat film of Pb, avoiding the formation of islands). This thin layer of Pb has a transition temperature of 6.88 K. c) Then the Pb is covered in three steps with K which is a normal conductor. As one expects for a superconductor-normal conductor (SN) sandwich, the transition temperature decreases with increasing K thickness and reaches the value of 4.79 K for a K thickness of 4.9 nm. This is shown in fig. 2. d) Finally, the K is covered with Pb in steps of $0.02, 0.05, 0.01, 0.3, 1.0, \ldots$ mono-layers. Initially, the Pb has no effect on the transition temperature. Only when the Pb thickness reaches one mono-layer, T_{c} increases slightly. The likely reason for this increase in T_{c} is that one approaches an SNS sandwich.

The observed behavior in T_{c} is typical for an SN sandwich and agrees qualitatively with the transition temperature of PbCu sandwiches [3–5]. What is very unusual is the behavior of the resistance (conductance) of the sandwich. This is shown in fig. 3, where the conductance is plotted *vs.* the coverage with K and (the second) Pb. The evaporation of the first K film decreases the conductance. This means that the K i) increases the resistance of the Pb film

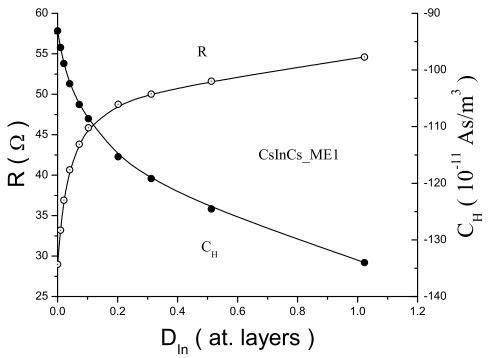

Fig. 1 – The resistance and the Hall constant of a Cs film, covered with sub-mono-layers of In, as a function of the In coverage.

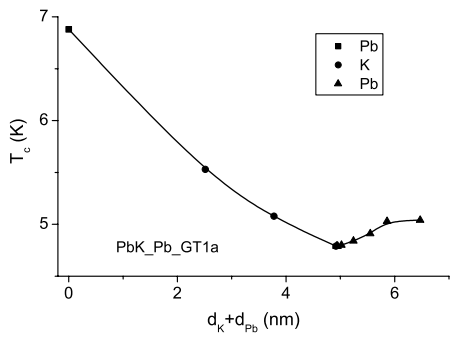

Fig. 2 – The superconducting transition temperature of a sandwich consisting of (Sb)/Pb/K/Pb.

and ii) contributes nothing (or very little) to the conductance. This changes for the second and third K coverage. One observes a clear increase in the conductance due to the K film. (In this experiment, we focussed on the effect of the second Pb film; the first K thickness of about 6 atomic layers lies already beyond the conductance minimum. But similar studies of PbK sandwiches [6] show that the conductance is a smooth function of the K thickness as indicated by the full curve.)

The evaporation of the sub-mono-layers of Pb reduces the conductance of the sandwich. With increasing Pb coverage, the conductance of the sandwich drops not only below the conductance of the pure Pb film, but even below the minimum value of the Pb/K sandwich. The conductance of the sandwich behaves as if the whole K film became insulating.

Again a devil's advocate might say that together with Pb some O_2 is introduced into the system, oxidizing K so that the effective thickness of the K is reduced. Furthermore, one of the referees suggested that the Pb on top of the K forces the K film to "ball-up".

Let us first consider the "ball-up" model. If the K takes the shape of half-spheres, it would still contribute to conductance because its base is in contact with the first Pb film. It is quite difficult to find a structure for the K that would not contribute to the conductance. One would have to assume a column shape of the K where the dimension in the plane is smaller than the height, *i.e.* less than 3-5 nm. Part of the interface between the first Pb film and the K must be free of conducting K. This means that there are parts of the Pb film where T_c is not reduced by the K. Therefore, the balling-up would result in a smaller reduction of T_c, *i.e.* an increase of T_c with the condensation of the second Pb coverage, even if one

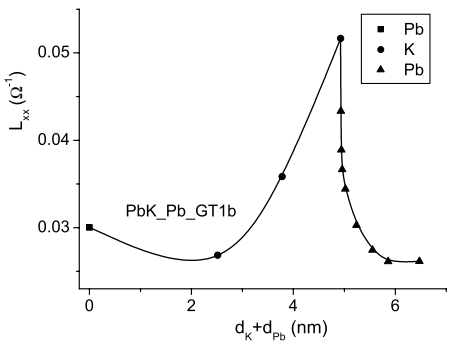

Fig. 3 – The conductance of the same sandwich as in fig. 2 as a function of the K and Pb coverage.

takes the non-locality of the superconductivity into account. Any inhomogeneity in the K thickness increases the transition temperature of the sandwich. Therefore, we believe that we can exclude the ball-up model.

Next we consider what would happen if K is oxidized during the Pb evaporation (although the ultra-high vacuum excludes this possibility). This would result in an increase of the transition temperature of the sandwich: the effective thickness of the normal conductor would be reduced and therefore the T_c would approach the T_c of the first Pb film. However, the T_c of the sandwich remains practically constant.

In the following we want to show that the unaffected transition temperature of the sandwich excludes that

- the effective thickness of the K film is reduced;
- the electron density in the K film is reduced;
- the mean free path perpendicular to the film is noticeably reduced.

The properties of an inhomogenous superconductor are determined by the gap equation [7]. Close to the transition temperature the superconducting gap function $\Delta(\boldsymbol{r})$ is very small and the "gap equation" can be linearized. This linear gap equation has the form [8–10]

$$\begin{aligned}\Delta(\boldsymbol{r}) &= V(\boldsymbol{r})\int \mathrm{d}^3\boldsymbol{r}'\int_{-\infty}^{0}\frac{\mathrm{d}t'}{\tau_T}\sum_{\omega_n} e^{2|\omega_n|t'}F(\boldsymbol{r},\boldsymbol{0};\boldsymbol{r}',t')N(\boldsymbol{r}')\Delta(\boldsymbol{r}'),\\ \tau_T &= \frac{\hbar}{2\pi k_{\mathrm{B}}T}\,, \qquad n_c=\frac{\Theta_D}{2\pi T}\,,\end{aligned}$$

where $\Delta(\boldsymbol{r})$ is the gap function at the position $\boldsymbol{r}$, $\omega_n=(2n+1)\pi k_{\mathrm{B}}T/\hbar$ are the Matsubara frequencies, $1/\tau_T=2\pi k_{\mathrm{B}}T/\hbar$, $V(\boldsymbol{r})$ is the effective electron-electron interaction at the position $\boldsymbol{r}$. $F(\boldsymbol{r},0;\boldsymbol{r}',t')$ gives the probability of a single electron to travel from $\boldsymbol{r}'$ to $\boldsymbol{r}$ during the time interval $|t'|$ (departing at $\boldsymbol{r}'$ at the negative time $t'<0$ and arriving at $\boldsymbol{r}$ at the time $t=0$), $N(\boldsymbol{r}')$ is the (BCS) density of states for one spin direction.

One can describe the physical meaning of the above equation as follows.

- At the (negative) time t' we have $N(\boldsymbol{r}')\Delta(\boldsymbol{r}')$ electrons departing from the position $\boldsymbol{r}'$.
- A fraction $F(\boldsymbol{r},0;\boldsymbol{r}',t')$ of them reaches the position $\boldsymbol{r}$ at the time $t=0$.
- During this propagation, their number is decaying as $\sum_{\omega_n} e^{2|\omega_n|t'}$ (due to the dephasing of the pair amplitude). In this sum the term $\exp[2|\omega_0|t']=\exp[-2\pi k_{\mathrm{B}}T|t'|/\hbar]$ is the most important one because it represents the longest coherence time.
- At the time $t=0$ and position $\boldsymbol{r}$, we integrate over all initial starting positions and over all times $t'<0$.
- The resulting (number of) electrons at $(\boldsymbol{r},t=0)$ multiplied with the attractive electron-electron interaction $V(\boldsymbol{r})$ yields the gap function at the position $\boldsymbol{r}$.

(One of the authors derived the time-dependent Ginzburg-Landau equation from this interpretation by replacing the time 0 by t and $\Delta(\boldsymbol{r}),\Delta(\boldsymbol{r}')$ by $\Delta(\boldsymbol{r},t),\Delta(\boldsymbol{r}',t')$ [11].)

For a sandwich of a superconductor and normal conductor, the gap function vanishes in the normal conductor because the effective electron-electron interaction is zero. Therefore, one has

to consider only electrons which start and arrive in the superconductor, *i.e.* the contribution of $F(\boldsymbol{r}, \mathbf{0}; \boldsymbol{r}', t')$ only counts when $\boldsymbol{r}'$ and $\boldsymbol{r}$ lie in the superconductor. This contribution $F(\boldsymbol{r}, \mathbf{0}; \boldsymbol{r}', t')$ is continuously reduced when a normal-conductor film is condensed onto the superconductor because along the way the electrons can escape into the normal film. This reduces the contribution of the integral over $\int \mathrm{d}\boldsymbol{r}' \int_{-\infty}^{0} \mathrm{d}t'$ and destroys the self-consistency of the gap equation. To maintain the self-consistency, one has to lower the temperature so that the dephasing is reduced.

Since T_c does not change when sub-mono-layers of Pb are condensed on the K film, we conclude that neither the effective thickness of the K nor its density of states is reduced. On the other hand, the resistance measurements show that there is practically no conductance through the K film covered with some Pb. The K film behaves as if its electrons were unable to carry a current in the x-y plane but can easily move in the z-direction. It is the behavior of a thin film with localized electrons where the film thickness is shorter than the localization length.

The question is: If we have localization is the localization

- due to disorder? If the Pb atoms on the K surface are disordered and cause Friedel oscillations in the film, these might impose disorder onto the position of the K atoms. Since the alkali metals are open metals their atoms should be easily displaced by charge oscillations.
- due to the large electron-electron interaction in the alkali metals in combination with the structural disorder?
- due to charge density waves [12]?

Our experimental results raise a large number of questions which require additional experimental and theoretical investigations. Here the superconducting proximity effect is a remarkably powerful tool in the investigations. Further investigations of such T_c measurements in a perpendicular magnetic field should shed additional light on the phenomenon.

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