

## Weak localization in thin Cs films

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Thin, quench condensed films of Cs change their resistance and Hall effect dramatically when covered with surface impurities. In this paper we investigate the quantum interference corrections to the resistance (weak localization) and determine the inelastic dephasing rate of the conduction electrons. The dephasing rate is proportional to the temperature-dependent resistance. For pure Cs films the magnetoresistance curves show a rather poor agreement with the theory, which is exceptional for quench condensed metal films. In particular, at 4.5 K a linear magnetoresistance is observed at large magnetic fields, which defies explanation. Sandwiches of AgCs yield a much better agreement between the experimental results and the theory. However, the dephasing rate of Cs in Ag/Cs and Au/Cs sandwiches has only half the value of that in pure Cs films with the same thickness and mean free path. [S0163-1829(99)04611-1]

### I. INTRODUCTION

The alkali metals have long been considered as the best representation of free-electron systems. The free-electron Fermi surface does not touch or intersect any Bragg plane and lies well inside of the first Brillouin zone. As a matter of fact, that is how the alkali metals are still described in many text books. There have, however, been early suggestions by Overhauser that simple metals form spin-density waves (SDW's) (Refs. 1 and 2) or charge-density waves (CDW's).<sup>3</sup> Overhauser has since collected a large body of evidence for the existence of CDW's in K and Na.<sup>4</sup>

Our group has recently observed some unusual properties in thin Cs films.<sup>5</sup> Thin Cs films show a dramatic increase of the resistance and the Hall effect when covered with submonolayers of Ag, Au, In, and Pb. As shown in Fig. 1, 1/100 of a monolayer of In increases the resistance of a Cs film (thickness  $D = 82 \text{ \AA}$ ) by approximately 15% while the Hall constant increases by approximately 3%. The size of the effect decreases with thicker Cs films. For thicker Cs films the saturation is essentially reached for an impurity coverage of 0.1 atomic layers. This indicates that the impurities act from the surface and do not penetrate the film. We attempted a number of explanations with the traditional free-electron picture. In particular, we considered the possibility that the impurity atoms introduce diffuse surface scattering. However, a detailed analysis showed that this explanation required a scattering cross section for the impurities that was far too large and it could not explain the increase of the Hall effect. Another hypothesis was that the impurities passivate a finite thickness of the Cs at the surface. This yields a thickness of the passivated layer of approximately 30  $\text{\AA}$  for thin Cs films and about 60  $\text{\AA}$  for thicker ones. However, this requires that one impurity atom would passivate approximately 30 Cs atoms, a rather unlikely scenario. Therefore we feel that we have to consider the more interesting models of spin-density and charge-density waves suggested by Overhauser.

In this paper we investigate the weak localization of the conduction electrons in thin quench condensed Cs films. In thin disordered films one observes quantum interference of the conduction electrons. This yields a distinctive magnetoresistance at low fields (see, for example, Refs. 6–8). These magnetoresistance curves correspond to time-of-flight

measurements and their evaluation yields characteristic fields such as  $H_i$  and  $H_{so}$ , which can be easily transferred into the dephasing time  $\tau_i$  and the spin-orbit scattering time  $\tau_{so}$  [by means of Eq. (3.1)].

### II. EXPERIMENT RESULTS AND EVALUATION

The Cs films are evaporated from SAES-Getters Cs 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 magnetoresistance is measured in a field range of  $-7 < B < +7$  T at several temperatures between 4.5 and 20 K. Furthermore, the resistance is measured as a function of temperature between liquid-helium temperature and 30 K.

#### A. Weak localization of films of Cs and Cs with In surface impurities

The magnetoresistance (MR) of a pure Cs film ( $D = 82 \text{ \AA}$ ,  $R = 28.8 \Omega$ ) is shown in Fig. 2 at different tempera-

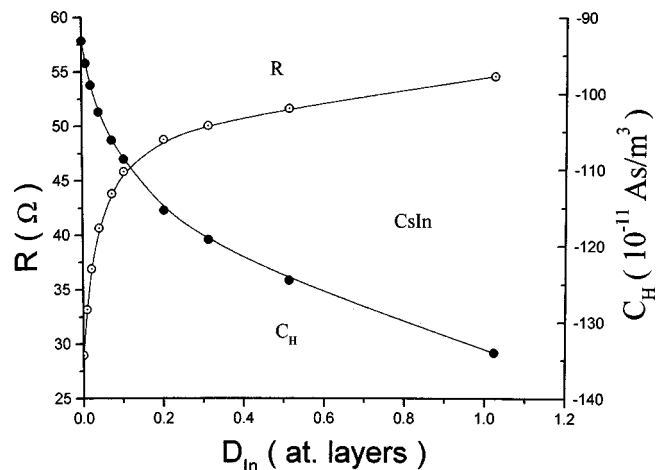


FIG. 1. A thin Cs film (82  $\text{\AA}$  thick) is covered with submonolayers of In. The resistance (left scale) and the Hall constant (right scale) is plotted as a function of the In coverage (in atomic layers).

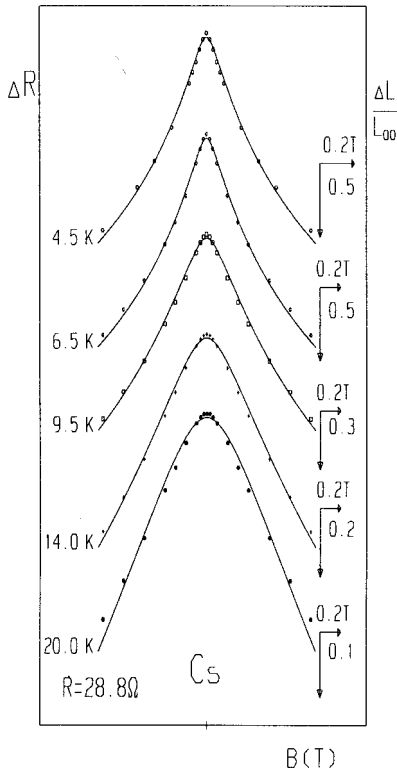


FIG. 2. The magnetoconductance of a Cs film ( $D_{cs}=82.4 \text{ \AA}$ ,  $R=28.8 \text{ }\Omega$ ) for different temperatures. The points are the experimental results and the full curves are fits with the theory. The right scale is the conductance in units of  $e^2/2\pi^2\hbar$  and the left scale gives  $\Delta R$  in  $\Omega$ .

tures. The symbols represent the experimental results. The theoretical evaluation is performed with the theory of weak localization by Hikami, Larkin, and Nogaoka.<sup>9</sup> This fit between experiment and theory requires two characteristic fields  $H_i$  and  $H_{so}$ , which represent the dephasing rate  $1/\tau_i$  and the spin-orbit scattering rate  $1/\tau_{so}$ .

For simple quench condensed metals one generally observes an excellent agreement between the experimental MR curves and the theory.<sup>10,11</sup> However, for the quench condensed Cs films we find large deviations between the experimental results and the theory at larger magnetic fields, in particular, at low temperatures. That is shown in Fig. 3. Here one finds an almost linear MR at 4.5 K for  $B > 1 \text{ T}$ . This behavior is definitely not due to weak localization. There is another MR mechanism present on top of weak localization. Since these Cs films are very disordered, even in a field of 7 T the product  $\omega\tau$  is only of the order of 0.02, normal magnetoconductance effects should be suppressed.

When one covers the Cs film with a monolayer of In then its resistance almost doubles. In Fig. 1 the dependence of the resistance is plotted as a function of the In coverage. The latter is given in units of atomic layers. We repeat the MR measurement for the In coverage of 0.1 atomic layers. The results are shown in Fig. 4. For temperatures between 4.5 and 10 K the film has a sufficiently strong spin-orbit scattering that one can observe a minimum at  $B=0$ . However, the fit with the theory of weak localization is even less satisfactory than for the pure Cs film. Even the low-field portion of the curves can only be fitted in the central part (which yields the dephasing field  $H_i$ ). Above 9.5 K the fitted values of  $H_i$

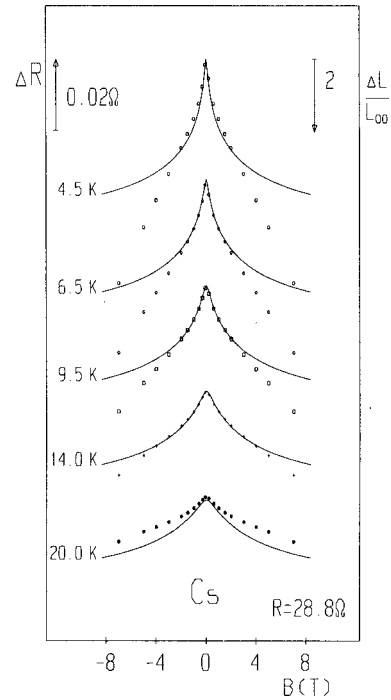


FIG. 3. The same magnetoconductance curves as in Fig. 2 but in a larger field range.

and  $\tau_i$  are no longer reliable. In Fig. 5 the MR of the Cs film with increasing coverage of In is plotted for a temperature of 6.5 K and the full field range. Here the full curves are only a guide to the eye. Clearly the linear slope of the conductance at large fields decreases with increasing In coverage.

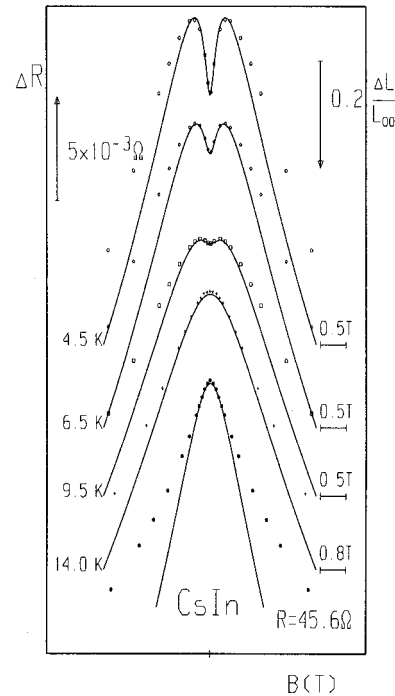


FIG. 4. The magnetoconductance of a Cs film covered with 0.1 atomic layers of In for different temperatures. The points are the experimental results and the full curves are fits with the theory. The right scale is the conductance in units of  $e^2/2\pi^2\hbar$  and the left scale gives  $\Delta R$  in  $\Omega$ .

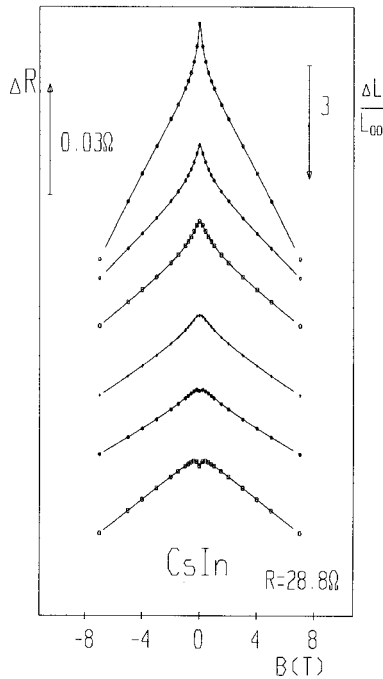


FIG. 5. The magnetoresistance of a Cs film with increasing cover of In at 6.5 K. The In coverages are 0, 0.01, 0.02, 0.05, 0.1, and 0.2 atomic layers of In. The right scale is the conductance in units of  $e^2/2\pi^2\hbar$  and the left scale gives  $\Delta R$  in  $\Omega$ . The full curves are a guide to the eye.

## B. Proximity effect of weak localization for sandwiches

### 1. Ag/Cs sandwiches

In the next step we investigate the weak localization of Ag/Cs sandwiches. For a sandwich of two thin films the weak localization averages perfectly over both films and can be described (for each temperature) by an effective dephasing field  $H_i$  and an effective spin-orbit scattering field  $H_{so}$ .<sup>12</sup> From the latter one can derive the dephasing rate and spin-orbit rate of the second film (see below).

In the experiment we first prepare a thin Ag film with a thickness of 68 Å and a resistance per square of  $R = 106.5 \Omega$ . We measure the magnetoresistance of the Ag film. Then we cover this film with Cs at liquid-He temperature in several steps where the total film thickness is increased to 92, 116, 140, and finally 183 Å. Since the Cs has a Debye temperature of approximately 38 K we anneal the sandwich only to 12 K to maintain a maximum of disorder. The resistance of the last sandwich drops down to a value of 12  $\Omega$ . The Ag introduces a spin-orbit scattering in the Ag/Cs sandwich that allows us to determine the characteristic fields  $H_i$  and  $H_{so}$  for each sandwich (each sandwich behaves as a perfect two-dimensional conductor and has a well-defined value for  $H_i$  as well as for  $H_{so}$ ). As we see below we can easily determine the characteristic fields of the Cs film on top of the Ag as a function of the Cs thickness. In Fig. 6 we have plotted the MR curves of the pure Ag film (on the top) and the Ag/Cs sandwiches for 4.5 K. The points represent the experimental data while the full curves give the theoretical fit using the theory by Hikami, Larkin, and Nagaoka. We observe a very good agreement between experiment and theory that is typical for quench condensed films.

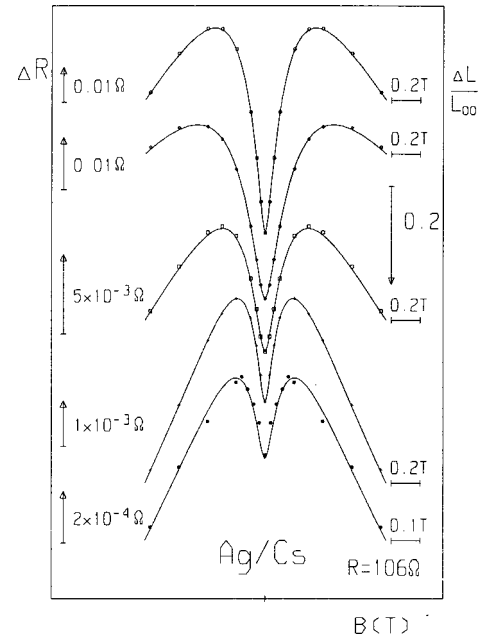


FIG. 6. The magnetoresistance of AgCs sandwiches at 4.5 K. The top curve is for a pure Ag film with a resistance of 106  $\Omega$ . The right scale is the conductance in units of  $e^2/2\pi^2\hbar$  and the left scale in  $\Omega$ . The total thicknesses are given in the text.

This good agreement between experiment and theory extends to large fields. In Fig. 7 the same MR curves are shown in the full range of the magnetic field. One can see this good agreement with the exception of the lowest curve. Here the resistance (per square) is reduced to 12  $\Omega$  and one observes, on top of the contribution of weak localization, a quadratic field dependence. Such a classical MR is expected when the

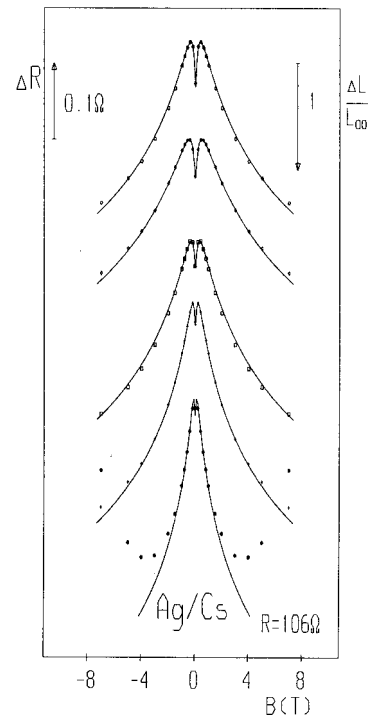


FIG. 7. The same magnetoresistance curves as in Fig. 6 of the AgCs sandwiches at 4.5 K but in a larger field range.

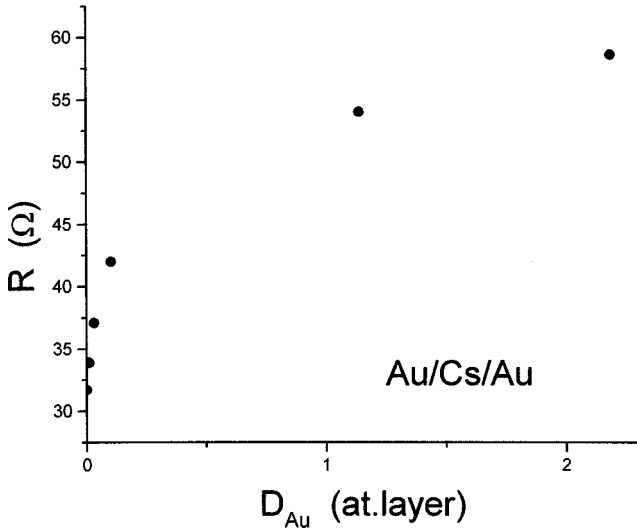


FIG. 8. A sandwich of Au/Cs is covered with sub-mono-layers of Au. The resistance is plotted as a function of the Au coverage (in atomic layers).

product  $\omega_{\mathbf{k}}\tau_{\mathbf{k}}$  ( $\omega_{\mathbf{k}}$  is the cyclotron frequency,  $\tau_{\mathbf{k}}$  is the relaxation time at the wave number  $\mathbf{k}$ ) varies as a function of the position on the Fermi surface or the position in real space. The product  $\omega_{\mathbf{k}}\tau_{\mathbf{k}}$  is different in the two films of the sandwich. Furthermore, in the presence of charge density waves  $\omega_{\mathbf{k}}\tau_{\mathbf{k}}$  also varies as a function of  $\mathbf{k}$  in the Cs film.

### 2. Au/Cs sandwiches

In a similar experiment we covered a Au film ( $R = 160 \Omega$ ,  $D_{\text{Au}} = 65.6 \text{ \AA}$ ) with increasing thicknesses of Cs. This time the sandwiches were annealed to 35 K. The first Cs film (on top of Au) had a thickness and mean free path of  $D_{\text{Cs}} = 45.7 \text{ \AA}$  and  $l_{\text{Cs}} = 40 \text{ \AA}$ . The magnetoresistance curves of this sandwich showed a nice agreement with the theory of weak localization. In the next step the Cs thickness was increased ( $D_{\text{Cs}} = 78 \text{ \AA}$ ,  $l_{\text{Cs}} = 100 \text{ \AA}$ ). Now we find again deviations from the theory.

In this sandwich of AuCs we covered the Cs again with submonolayers of Au. We observed a strong increase of the resistance. Despite the fact that the Cs had a substrate of Au the qualitative behavior was the same as in the condensation of In on pure Cs. In Fig. 8 the resistance is plotted versus the Au coverage (in units of atomic layers).

### C. Temperature dependence of the Cs resistance

Cs has a very low Debye temperature of  $\Theta_D \approx 38 \text{ K}$ . Above  $\Theta_D/5$  the increase in resistance is essentially linear with temperature. Figure 9 shows the temperature dependence of the pure Cs film (left scale) and the Cs film covered with 0.1 atomic layers of In (right scale). The slope of the temperature dependent resistance is 12% larger for the Cs with In surface impurities.

## III. DISCUSSION

### A. Dephasing rate in Cs

#### 1. Pure Cs

The magnetoresistance in disordered thin films is described by the theory of Hikami, Larkin, and Nagaoka.<sup>9</sup> A fit

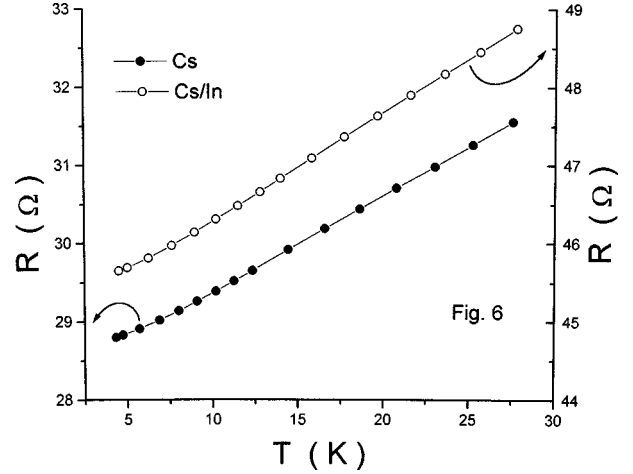


FIG. 9. The resistance of the pure Cs film and the Cs with an In coverage of 0.1 atomic layers as a function of the temperature.

between the experimental magnetoresistance and the theory yields two characteristic fields, the inelastic field  $H_i$  and the spin-orbit scattering field  $H_{so}$ . From these fields one obtains the characteristic rates, the dephasing rate  $1/\tau_i$  and the spin-orbit scattering rate  $1/\tau_{so}$  by the relation

$$\frac{1}{\tau_n} = \frac{4}{e\hbar\rho N} H_n, \quad (3.1)$$

where  $\rho$  is the resistivity of the film and  $N$  is the density of states of the investigated metal.

The magnetoresistance curves of the pure Cs films do not show a spin-orbit scattering minimum at zero field. We set the spin-orbit scattering field  $H_{so}$  tentatively equal to zero (we will return to this point). Then the evaluation of the magnetoresistance curves in Fig. 2 yields the dephasing rate of the pure Cs film. The results are plotted in Fig. 10. The dephasing rate depends almost linearly on the temperature. Since Cs has a very low Debye temperature of 38 K this linear dephasing rate can be explained by electron-phonon processes. Below we will see that the temperature dependent resistance confirms this picture.

### 2. Sandwiches with Cs

From the experiments with sandwiches of Ag/Cs and Au/Cs we can also derive the dephasing rate of Cs. The sandwiches consist of two films with resistances  $R_1$  and  $R_2$  and the characteristic fields  $H_{i,1}$ ,  $H_{so,1}$  and  $H_{i,2}$ ,  $H_{so,2}$ . One of the authors<sup>12</sup> developed the theory for weak localization in two-dimensional sandwiches. The characteristic field  $H_n$  ( $H_n$  representing either the dephasing field  $H_i$  or the spin-orbit scattering field  $H_{so}$ ) of the sandwich can be obtained from the characteristic fields of the individual films  $H_{n,1}$  and  $H_{n,2}$ :

$$\frac{H_n}{R} = \frac{H_{n,1}}{R_1} + \frac{H_{n,2}}{R_2}. \quad (3.2)$$

Since in the experiment all the parameters of the first film ( $R_1, H_{i,1}, H_{so,1}$ ) and all the parameters of the sandwich ( $R, H_i, H_{so}$ ) are determined one can use Eq. (3.2) to determine the properties of the second film, in particular,  $R_2, H_{i,2}, H_{so,2}, \rho_2$ . From these data we obtain the dephasing

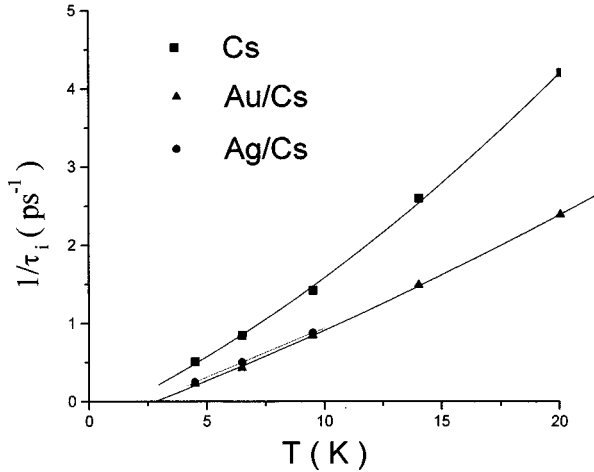


FIG. 10. The electron dephasing times in a pure Cs film (full circles) and similar Cs films on top of Ag and Au as a function of temperature.

rate of the Cs films. The results are shown in Fig. 10 together with the data for pure Cs. In Table I the parameters of the Cs films are collected. The properties of the three films are rather similar. The inelastic rates are quite different for the sandwiches as opposed to the single Cs film. A possible explanation could be a change in the phonon spectrum or the electronic structure of the Cs on top of the noble metal. At the present time we have not yet systematically investigated this deviation.

### 3. Influence of surface impurities

When we cover the Cs with impurities then the resistance increases dramatically as shown in Fig. 1. At the same time one's ability to fit the magnetoresistance curves becomes greatly reduced. As shown in Fig. 4 the inner part of the magnetoresistance curves (which yield the inelastic rate) can still be fitted for 4.5 and 6.5 K. The resulting dephasing rate shows an increase due to the impurity coverage. For a coverage of 0.1 atomic layers of In at 6.5 K the increase is approximately 15%.

### B. Deviations from weak localization theory

In the majority of quench condensed metal films one observes an excellent agreement between the experimental magnetoresistance and the theory.<sup>13</sup> (Only in metal films with magnetic effects such as Kondo impurities or spin fluctuations as in Pd have we observed high-field deviations between experiment and theory). The reason for this universal agreement is that weak localization is a great equalizer. It does not matter what the scattering properties of the elastic impurities are.<sup>14</sup> It does not matter which and how many

TABLE I. The Cs parameters (thickness  $D_{Cs}$ , resistivity  $\rho$ , and mean free path  $l$ ) in the pure Cs film and the Ag/Cs and Au/Cs.

	$D_{Cs}$ (Å)	$\rho$ ( $10^{-6}$ Ωm)	$l_{Cs}$ (Å)
Cs	82.4	0.24	127
Ag/Cs	72.1	0.29	102
Au/Cs	78	0.31	98

electron bands are participating in the conduction process.<sup>15</sup> The only requirement is that the transitions between different bands are fast compared with the characteristic time of weak localization. This characteristic time is the smallest of the following times: the inelastic dephasing time  $\tau_i$ , the spin-orbit scattering time  $\tau_{so}$  and the magnetic dephasing time  $t_H = (1/H)(1/4eD)$ , where  $H$  is the applied magnetic field and  $D$  the diffusion constant. Even spacial inhomogeneities are averaged out if the conduction electrons diffuse during their characteristic time over larger distances than the inhomogeneities. Weak localization yields a motional averaging.

Therefore the observed deviations for Cs films and CsIn sandwiches carry a clear message. Either the MR contains other contributions that have nothing to do with weak localization or some transitions in the films are so slow that the motional averaging process is lost.

### C. The negative linear magnetoresistance at high fields

The pure Cs film shows, at low temperatures and high magnetic fields, a *negative* linear MR as is shown in Fig. 3. The first question is whether this deviation from the theory of weak localization is in itself a two-dimensional quantum correction. Quantum corrections such as weak localization, the Coulomb anomaly of the resistance, and the Aslamzov-Larkin and the Maki-Thompson fluctuations in superconductivity, to name a few, are specific interference contributions to the current that are represented by specific Kubo diagrams. They yield a resistance independent (and often universal) contribution to the conductance of a thin film. On the other hand, everything that causes an additional scattering (such as impurities, electron-phonon interaction, etc.) contributes additively to the *resistance*. We investigated Cs films with different resistances between 100 and 1 Ω. We found that the deviation from the weak localization theory was essentially constant in a resistance plot while in a conductance plot it decreased roughly as  $1/R^2$ . The decrease of the linear slope with increasing In coverage in Fig. 5 demonstrate the same effect since the resistance increases with the In coverage. (For the very low film resistance the analysis was obscured by an increasing (classical) quadratic MR). This strongly suggests that the linear MR is *not* a quantum correction but due to an additional scattering mechanism. This scattering must decrease with increasing magnetic field to yield a negative MR.

There have been observations of a linear MR in pure alkali metals in the past (see, for example, Ref. 16). These have been interpreted by Overhauser and co-workers<sup>17</sup> as a proof that the alkali Fermi surface is not simply connected so that the electrons can travel on open orbits. This explanation does not apply in our experiments for the linear MR since the product  $\omega\tau$  is very small in our disordered Cs film and has a value of approximately 0.02.

The origin of this scattering mechanism is rather mysterious. We have to search for a mechanism that (a) is magnetic-field dependent, (b) shows a strong temperature dependence below 20 K, and (c) disappears if the Cs film is condensed onto a Ag film.

Since we consider any phase coherence effect as an unlikely origin of this linear MR we have only a few mechanism by which the magnetic field changes the resistance in

these disordered films. (i) the presence of magnetic impurities together with spin-flip scattering, (ii) a magnetic breakdown in the Fermi surface, and (iii) impurities with an extended electronic wave function or an extended screening cloud. None of these mechanisms appears to be the likely origin of the linear MR.

#### D. Spin-orbit scattering

Our Cs films do not show a spin-orbit scattering (SOS) minimum at zero magnetic field. From a comparison between the dephasing rate and the electron transport rate below we conclude that the spin-orbit scattering rate must be small compared with the dephasing rate.

Cs belongs to the same row in the periodic system as Au. Thin films of quench condensed Au show a very strong SOS and are essentially in the strong SOS limit. Both metals possess one  $s$ -conduction electron per atom. Therefore the different behavior is at first rather puzzling. We suggest the following explanation. Let us first consider the SOS of a single Cs atom in a free-electron jellium. Its SOS cross section  $\sigma_{so}$  is given by<sup>18</sup>

$$\sigma_{so} = \frac{4\pi}{k_F^2} \sum_l \frac{l(l+1)}{2l+1} \sin^2(\delta_{l+(1/2),l} - \delta_{l-(1/2),l}),$$

where  $\delta_{l\pm(1/2),l}$  is the scattering phase shift for an electron wave with total angular momentum  $l \pm \frac{1}{2}$  and orbital angular momentum  $l$ . For a Cs metal with filled  $p$  and  $d$  bands and with a relatively large distance between neighboring atoms we expect that only the phase shift for  $l=0$  will be important. This  $s$  scattering does not contribute to the SOS cross section. In the case of Au impurities the situation is more complicated. Because of the close proximity of the  $d$  band to the Fermi energy one cannot neglect the  $d$ -phase shift. A stronger SOS appears reasonable in Au.

In the Ag/Cs sandwich experiment we condensed Cs on top of Ag. The interface introduces an asymmetrically distorted electronic wave function at the Cs atoms, i.e., a wave function with nonzero angular momentum. As a consequence we observed an increase of the SOS rate of the sandwich. In Fig. 11 the additional SOS rate due to the Cs film is plotted as a function of the Cs thickness. The evaluation yields a much larger spin-orbit scattering rate in the Cs on top of the Ag than the rate for pure Cs. This result is presently not well understood. The SOS rate of the Ag film is  $\tau_{so}^{-1} = 0.26 \text{ ps}^{-1}$ .

#### E. Weak localization and the temperature-dependent resistance

The measurement of the magnetoresistance yields the dephasing rate  $1/\tau_i$  of the conduction electrons. In the experimental temperature range the dephasing rate is caused by the inelastic electron-phonon processes and essentially given by  $1/\tau_{ep}$ .

We have also investigated the temperature-dependent resistance of the Cs film. In the free-electron model the resistance is given by the Drude formula

$$R(T) = \frac{1}{D_{Cs}} \frac{m}{ne^2} \frac{1}{\tau_{tr}}, \quad (4.3)$$

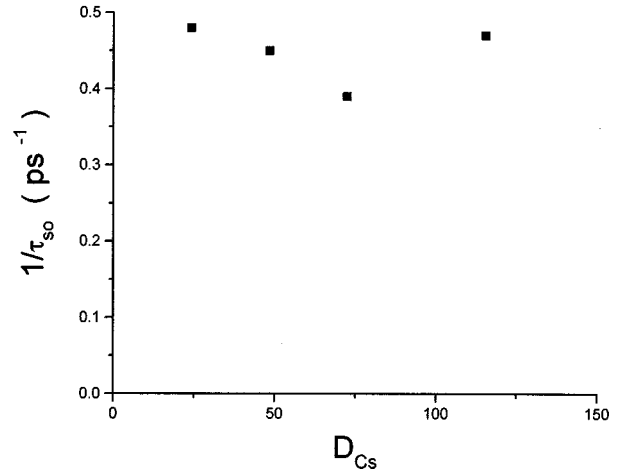


FIG. 11. The additional spin-orbit scattering rate  $\tau_{so}^{-1}$  of the AgCs sandwich due to the Cs coverage as a function of the Cs thickness.

where  $D_{Cs}$  is the thickness of the Cs film. For isotropic or catastrophic scattering the transport relaxation rate is

$$\frac{1}{\tau_{tr}} = \left[ \frac{1}{\tau_0} + \frac{1}{\tau_{ep}} \right],$$

where  $1/\tau_0$  is the elastic scattering rate. For anisotropic scattering one has to include the factor  $\langle 1 - \cos \Theta \rangle_{av}$  between the transport relaxation time and the mean relaxation time. We ignore this complication here because it is not a serious problem at sufficiently high temperatures (with respect to the Debye temperature).

Under these simplifying assumptions we expect that a plot of the transport relaxation rate in the resistance  $1/\tau_{tr} = RD_{Cs}ne^2/m = 1/\tau_0 + 1/\tau_{ep}$  versus the dephasing rate in weak localization  $1/\tau_{ep}$  yields a straight line. Figure 12 shows this plot for the pure Cs film. The slope of the straight line through the points is 1.14 for the pure Cs film. This is remarkably close to the value 1 if one considers that we neglected any dependence on the scattering angle.

The above evaluation shows that even at 4.5 K the fitted dephasing rate and the transport rate lie on the straight line. This suggests that the neglect of the spin-orbit scattering rate yields the correct dephasing rate which means that  $1/\tau_{so}$  is quite small compared to  $1/\tau_i$ . Furthermore, we see that the same mechanism that causes the temperature-dependent resistance also causes the dephasing. For Cs with its low Debye temperature of 38 K this mechanism is expected to be the electron-phonon interaction.

#### F. Charge-density waves and disordered films

As pointed out in the Introduction Overhauser<sup>2</sup> had suggested that the electron system in alkali metals forms charge-density waves. In this model the resulting Fermi surface is very complicated. The charge-density vector  $\mathbf{Q}$  generates an energy gap at the  $\mathbf{k}$  plane defined by  $\mathbf{Q}$ . This  $\mathbf{k}$  plane is perpendicular to  $\mathbf{Q}$  and intersects  $\mathbf{Q}$  at  $\mathbf{Q}/2$ . The direction of  $\mathbf{Q}$  is roughly parallel to  $\mathbf{G}_{110}$  and its length is about  $1.11 \times 2k_F$  for potassium. For thin films of alkali metal  $\mathbf{G}_{110}$  should be perpendicular to the film plane.<sup>19</sup> In addition,

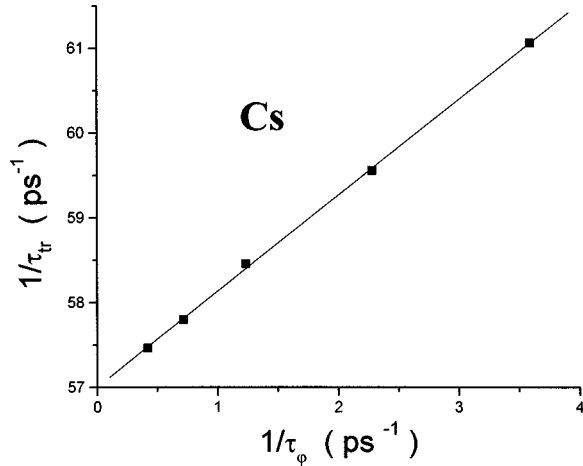


FIG. 12. The (temperature-dependent) transport relaxation rate is plotted vs the the dephasing rate of weak localization for the pure Cs film.

Overhauser obtained new gap planes defined by  $\mathbf{Q} \pm n(\mathbf{Q} - \mathbf{G}_{110})$ , the so-called “minigaps,” and at  $\pm n(\mathbf{Q} - \mathbf{G}_{110})$ , which Overhauser calls the “heterodyne gaps.” Here  $n$  is an integer. With increasing  $n$  the magnitude of the gap decreases. For Na Overhauser and his co-workers obtained a charge-density gap of magnitude 0.4 eV. The first minigap is 0.1 eV and the first heterodyne gap is 0.02 eV.

For thin films of alkali metals Overhauser<sup>4</sup> concluded that the  $\mathbf{Q}$  vector lies essentially perpendicular to the surface, i.e., it forms only a small angle with  $\hat{\mathbf{z}}$ , the unit vector perpendicular to the film. If the alkali metal film has a large mean free path and specular surface reflection then one has to consider the quantization of  $k_z = \nu(\pi/D_f)$ , where  $D_f$  is the thickness of the film. Now the Fermi surface has to accommodate the formation of charge-density waves and the  $k_z$  quantization at the same time. Without having performed a quantitative analysis we expect that the  $k_z$  quantization forces the  $\mathbf{Q}$ -vector to lie parallel to  $\hat{\mathbf{z}}$  and all the planes of energy gaps perpendicular to  $\hat{\mathbf{z}}$ . In this case the charge-density waves lie parallel to the film plane, an intuitively reasonable assumption.

The charge density wave in the alkali film forbids the  $\mathbf{k}$  vectors within a certain cone about the  $z$  direction, i.e., they have a smallest  $k$  vector  $\mathbf{k}_p$  within the  $k_x$ - $k_y$  plane. We expect that this has two consequences: (a) all conduction electrons hit the surface under minimum angle (with respect to  $\hat{\mathbf{z}}$ ), and (b) we will have surface states with  $\mathbf{k}$  vectors parallel to the film surface the  $\mathbf{k}$  values of which are less than  $k_p$ .

In thin disordered films one has a mean free path  $l$ . For example, our Cs film yields an electron elastic relaxation time  $\tau_0 = l/v_F$  and an energy uncertainty in any  $k$  state at the Fermi surface of  $\hbar v_F/l$ . This means that a Cs film with a mean free path of 100 Å has a smearing of the energy by about 50 meV. This smearing of the  $\mathbf{k}$  vector and the energy acts on both the  $k_z$  quantization and the charge-density energy gaps. It eliminates all secondary gaps except for the first minigap.

At the present time we have not yet collected enough experimental results to connect our findings in a unique way with the charge-density model. In particular, the strong change in resistance and Hall effect due to surface impurities represents quite a challenge.

We can only contribute an unproven suggestion: Imagine that in a pure alkali film the charge-density waves lie parallel to the surface. Then an electrical current in the film plane is hardly affected by their presence. We can think of two (quite different) effects of surface impurities.

(a) When surface impurities are condensed on the surface of the Cs they introduce points of high charge. This attracts the charge-density-wave maxima. (Similar to the attraction of superconducting flux lines by small normal conducting regions). In the surface region with the bent charge-density waves the electron flow is strongly hindered and the current flows in a reduced thickness of the film. This would yield an increased resistance as well as an increased Hall constant. But it leaves still a number of questions unanswered. For example, one might expect that the pinning of the charge-density wave would reach a maximum for a certain impurity coverage and decrease for larger coverages. (In a monolayer of surface impurities the pinning would cancel out).

(b) The surface impurities modulate the charge in their direct vicinity due to Friedel oscillations. These Friedel charge oscillations interact with the charge-density waves (both have the same wavelength  $\lambda_F/2$ ) and deform their shape in the vicinity of the impurity. This yields additional scattering of conduction electrons in the vicinity of the surface.

It should be pointed out that these two ideas exclude each other. In model (a) the charge density crest goes radially through the impurity and in model (b) the crest aligns perpendicular to the radius. A detailed evaluation of the involved energies is required to see which of the two arrangements is energetically more favorable.

#### IV. CONCLUSION

In this paper we have investigated the weak localization of thin films of pure Cs, Cs with In surface impurities and sandwiches of Ag/Cs and Au/Cs. The dephasing rate of pure Cs was in good agreement with the temperature-dependent transport relaxation rate of the resistance, showing that both processes are governed by the same inelastic processes. For larger fields the experimental magnetoresistance curves showed clear deviations from the theory of weak localization. The deviations increased when the Cs films were covered with 0.1 atomic layers of In. Such deviations are not observed in quench condensed films of other simple metals and appear to be due to a new unknown mechanism. They occur despite the disorder ( $\omega\tau < 0.02$ ) and are not due to open orbits.

The drastic effect of small concentrations of surface impurities on the properties of Cs extends to the dephasing rate and the temperature-dependent resistance. The superposition of 0.1 atomic layers of In on the discussed Cs film increases the resistance by 58%, the Hall constant by 17%, the slope of the linear part of the temperature-dependent resistance changes by 12%, and the dephasing rate by approximately 15%. This is another indication that the surface impurities not only increase the residual resistance but also alter the dynamics of the electron (and phonon) system. Mattiessen’s rule is definitely violated.

The experimental results of weak localization in Ag/Cs and Au/Cs sandwiches were evaluated with the sandwich theory of weak localization. They yield the surprising result

that the dephasing in the Cs sandwiches was roughly by a factor 2 smaller than for pure Cs films.

We believe that the observed properties cannot be explained within the (nearly) free-electron model of the alkali metals. Some more complex properties such as Overhauser's charge-density model has to be included. We first made attempts to discuss the interplay of charge-density waves and thin films, including the effect of disorder. Additional experimental research is required to shed more light on the unusual properties of the Cs and the research has to be extended to other alkali films. At the present time we are investigating the propagation of the conduction electrons perpendicular to

the film plane. Here we use the induced anomalous Hall effect in Cs in contact with thin ferromagnetic Fe films. Again the results are quite unexpected, indicating an internal reflection of the conduction electrons in the Cs film. These results will be published soon.

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