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Size effect in metallic sandwiches

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The condensation of a metal M onto a metallic substrate F avoids the formation of islands and yields a homogeneous structure of M. The theory of the geometrical size effect in such a sandwich is developed and magnetic field effects are included. The size effect in sandwiches allows a series of new and informative experiments. Three experimental examples are presented in which Cu, Pd, and In are quench condensed onto an amorphous substrate at He temperature. The conductance of these films follows the theory exactly, which suggests that they possess a homogeneous structure. The experimental data yield both the mean free path l and the product $\rho\ l$ for Cu, Pd, and In, where ρ is the resistivity.

I. INTRODUCTION

The mean free path of the conduction electrons plays the dominant role in the transport properties of metals. Therefore several experimental methods have been developed to determine the mean free path. They are collected under the name "size effects." A detailed discussion of these size effects: cyclotron-resonance, anomalous skin effect, geometrical size effect is given by Chambers.[1] In this paper we want to consider the geometrical size effect. One usually investigates a metallic film or foil whose surface scatters the conduction electron more or less diffusely. For thick samples the effective film thickness is reduced by $\frac{3}{8}l$ (l is the mean free path of the conduction electrons) due to a diffuse scattering at both surfaces. For thin films the conductance depends much more sensitively on the ratio D/l. Unfortunately, the application of the geometrical size effect is often problematic. On the one hand, it is difficult to prepare a film which is both thinner than the mean free path and homogeneous. It is difficult to avoid the formation of islands. On the other hand, one has for thick films the problem that the structure changes with increasing film thickness.

We are going to consider in this paper a modified geometrical size effect, which offers several advantages. We condense the metal of interest M on top of a metallic substrate F. This avoids in most cases the formation of islands. In addition one can vary the substrate and can choose, e.g., an amorphous metal or a ferromagnet. This allows, as we shall see later, quite new and interesting investigations.

In Sec. II we present the theoretical model and its evaluation. We discuss the broad spectrum of possible applications. In Sec. III we show experimental results for Cu, Pd, and In films which are quench condensed onto an amorphous metal at He temperature. The experimental results demonstrate that these films have a homogeneous structure and allow a determination of l and ρl (ρ is the resistivity) which contains the area of the Fermi surface.

II. THEORY

We consider the following sandwich which is sketched in Fig. 1. The semi-infinite metal F fills out the half-space $z<0$. It is covered with a film of the metal M in the region $0<z<D$ (D is the thickness of the metal film M). Later on, we apply a magnetic field B perpendicular to the film plane in the z direction. For the calculation of the conductance we make the following assumptions: (i) the conduction electrons in both metals are free and possess the same Fermi momentum, (ii) the conduction electrons are diffusely scattered at the upper surface of M, and (iii) the conduction-electron wave is damped by a factor of r when it crosses the interface between F and M in either direction. We make the last assumption in order to be able to also treat the case when two metals with a rather ideal structure possess an interface with structural disorder. r describes the transparency of the interface for conduction electrons.

In calculating the conductance of this inhomogeneous system (in square geometry) we use the vector mean-free-path method as described by Chambers.[1] As in a simple metal the Fermi distribution function of the conduction electrons is changed by the application of an electrical field:

$$f_{\vec{k}}(\vec{r})=f_{k}^{0}-\left(\frac{\partial f}{\partial \epsilon}\right)\Delta \mathcal{E}_{\vec{k}} . \tag{1}$$

Here $\Delta\mathcal{E}_{\vec{k}}$ is the energy that the conduction electron $\vec{k}$ gained on its path through the metallic system:

$$\Delta\mathcal{E}_{\vec{k}}=e\vec{E}\cdot\vec{\lambda}_{\vec{k}} , \tag{2}$$

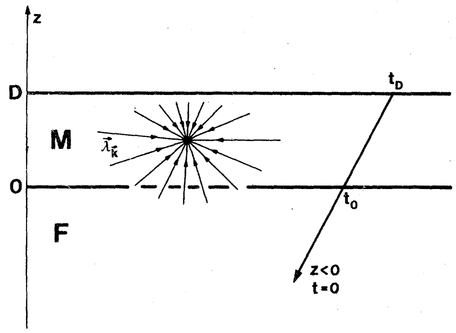

FIG. 1. Sandwich of the semi-infinite metal F ($z<0$) and the metal film M ($0<z<D$) of thickness D. The vector mean free path $\vec{\lambda}_{\vec{k}}$ for conduction electrons with different momenta $\vec{k}$ at one position is drawn, as well as the path of one conduction electron $\vec{k}$.

$\vec{\lambda}_{\vec{k}}$ is the vector mean free path of the conduction electron with momentum $\vec{k}$, i.e., the path that the conduction electron $\vec{k}$ traveled (without collision) before it reached the position $\vec{r}$. It is drawn in Fig. 1 for one position and different $\vec{k}$.

We need in the following calculation only the x and y components of the vector mean free path and denote them by the complex mean free path $\lambda_c = \lambda_x + i\lambda_y$. It is calculated in the Appendix. We have to distinguish four different cases: $z \gtrless 0$ and $v_z \gtrless 0$. We obtain

$$\begin{aligned}\lambda_c(z<0, v_z<0) &= l_c^F[1-\exp(-|z|/\tau_F|v_z^F|)]\\ &\quad + r l_c^M \exp(-|z|/\tau_F|v_z^F|)\\ &\quad \times[1-\exp(-D/\tau_M|v_z^M|)],\end{aligned} \tag{3}$$

$$\begin{aligned}\lambda_c(z<0, v_z>0) &= l_c^F,\\ \lambda_c(z>0, v_z<0) &= l_c^M\{1-\exp[-(D-z)/\tau_M|v_z^M|]\},\\ \lambda_c(z>0, v_z>0) &= l_c^M[1-\exp(-z/\tau_M v_z^M)]\\ &\quad + r l_c^F \exp(-z/\tau_M v_z^M),\end{aligned}$$

where

$$l_c^{M,F} = v_c^{M,F}\tau_{M,F} = v_\rho^{M,F}\tau_{M,F}e^{i\alpha},$$

$$v_\rho = (v_x^2+v_y^2)^{1/2}, \quad \tan\alpha = v_y/v_x .$$

Now we can easily calculate the energy shift of the conduction electrons due to an applied electrical field $\vec{E}=(E,0,0)$ in the x direction. One obtains the conductance in square geometry (not the conductivity) by integrating over the Fermi sphere of M and F and over the z direction. The contribution of the metal film M ($0<z<D$) is

$$L_c^M = L_{xx}^M + iL_{xy}^M = \frac{e^2}{4\pi^3\hbar}\int_0^D dz \int_M dS\, \lambda_c(z, v_z)\hat{k}_E , \tag{4}$$

where $\hat{k}_E = (\hat{k}\cdot\hat{E}) = \sin\theta\cos\phi$, and θ and ϕ are the angles in the polar coordinates. The contribution of the metal F to the conductance is obtained in the same manner. The integrations over z and ϕ can be easily performed and there remains only an integration over $u=\cos\theta$. For a comparison with the experiment we are only interested in the change of the conductance due to the second film M. We obtain after a straightforward calculation for the change in the (complex) conductance:

$$\begin{aligned}L_c &= A_M[l_M D - l_M^2 T(D/l_M) + \tfrac{1}{2} r l_M l_F T(D/l_M)]\\ &\quad + A_F \tfrac{1}{2} r l_M l_F T(D/l_M),\end{aligned} \tag{5}$$

where

$$A_{F,M} = \frac{e^2 k_{F,M}^2}{3\pi^2\hbar} = \frac{e^2}{12\pi^3\hbar}S_{F,M}$$

(S being Fermi-surface area) and

$$T(s) = \frac{3}{2}\int_0^1 du\,(1-u^2)u(1-e^{-s/u}),$$

$$\frac{dT(s)}{ds} = 1, \quad T(\infty) = \tfrac{3}{8}.$$

One can easily understand this expression for the changes in conductance due to the film M. If we set $r=0$ then the conduction electrons cannot cross the interface. The additional conductance, proportional to $l_M D - l_M^2 T(D/l_M)$, is just the result obtained by Fuchs[2] for the conductance of a thin film. The third term describes the change of conductance which is due to those conduction electrons which passed through the interface from F to M. The fourth term is due to those conduction electrons which passed through the interface from M to F and changed the conductance in F. Therefore it is proportional to A_F. Although we assumed in our model that the Fermi momenta k_F and k_M are identical, we distinguish them in order to demonstrate the origin of the change in conductance. If one condenses the same metal, $M=F$, on the metallic substrate with identical properties, then one has $r=1$ and $k_M=k_F$. Then the third and fourth terms compensate for the second one, and the change in conductance is, as it has to be proportional to the thickness. Those conduction electrons, which flow from F into M, originate from a thickness l_F in F and propagate in M over a distance which has about the minimum value of l_M or D, and which is of the order of $l_M T(D/l_M)$. Its contribution to the conductance is therefore proportional to $\frac{1}{2}l_F l_M T(D/l_M)$.

If we want to generalize the calculation for the presence of a magnetic field B in the z direction one has to replace the mean free path l by $\tilde{l}$ in both metals with

$$\tilde{l}_{M,F} = l_{M,F}/[1-i(\omega\tau)_{M,F}],$$

where l_M and l_F are the usual scalar mean free path and $(\omega\tau)_{M,F}$ are the Hall angles in M, F.

If the metals F and M have different Fermi momenta a contact voltage occurs at the interface and the direction of the velocity is changed after the penetration of the interface. This problem requires, in principle, a new calculation. One might obtain an approximate description by using the individual Fermi-surface areas and by adjusting the transparency r to the experimental data.

We now suggest some applications of the size effect in sandwiches.

(a) *Transparency at the interface*. If we evaporate first the metal F, its conductance increases with the slope $A_F l_F$. If we continue with the metal M, the conductance grows further with the slope $\frac{1}{2}r(A_M+A_F)l_F$. Therefore, such an experiment enables us to determine the transparency of the interface.

(b) *Amorphous substrate*. If we use for the first film an amorphous metal, its mean free path is of the order of a few angströms. Therefore, the contribution of electrons passing the interface is very small ($\sim l_F$). Any uncertainty of r, l_F, or A_F is not relevant and one obtains almost the results of an isolated film with well known corrections. The great advantage of such an experiment is the avoidance of islands on top of the amorphous film. We are going to present such experiments in Sec. III.

(c) *Anomalous Hall effect*. We may use for the first metal a ferromagnet which possess an anomalous Hall effect. For a ferromagnet the anomalous Hall angle corresponds to an internal field of megagauss proportions. This again causes in the second film M an anomalous Hall conductance proportional to

$$\tfrac{1}{2}r(A_F+A_M)l_F l_M(\omega\tau)_F T(D/l_M),$$

where $(\omega\tau)_F$ is the Hall angle in the ferromagnet. Such an experiment allows the direct determination of the mean free path l_M and will be published elsewhere.[3]

III. EXPERIMENT

We now give one example for the usefulness of the sandwich size effect. It is often claimed that the structure of a quench-condensed film is very inhomogeneous as a function of thickness.[4] We want to demonstrate for three examples that under optimal evaporation conditions: metallic substrate, ultrahigh vacuum, and constant evaporation rate, the structure is so homogeneous that the conductance of the quench-condensed film accurately follows the theoretical formula, yielding the correct product $\rho \cdot l$. We evaporate Cu and

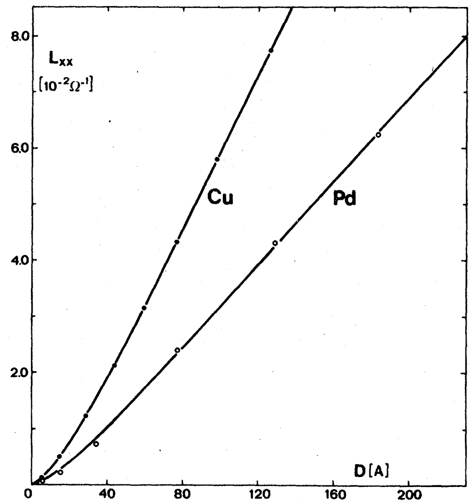

FIG. 2. Change in conductance due to Cu and Pd quench condensed on top of an amorphous metallic substrate is plotted as a function of thickness. The full curves are calculated according to Eq. (5), and yield the values l and $(\rho l)^{-1}$.

Pd on top of an amorphous film. Since we want to condense the metals in several steps we need very good vacuum conditions. We use an apparatus which we described recently[5] and which has a vacuum of the order of 10^{-11} Torr. In this apparatus the substrate (a quartz plate) is at 10 K. We condense amorphous Fe with a resistivity of 1.00×10^{-6} Ω m which corresponds to a mean free path of about 6 Å. On top of the amorphous Fe, we condense in several steps the metal M of interest. The thickness is measured with a quartz balance with an accuracy of 0.1 Å. Figure 2 shows the increase of conductance with increasing film thickness for Cu and Pd. The full curves are calculated according to Eq. (5). We assumed $A_F = A_M$ which is reasonable because the number of conduction electrons is about one for the three metals. We set $r=1$ (transparency approximation). By extrapolation of the linear part of the curves towards zero we obtain $\frac{3}{8}(l_M - l_F)$. This yields for $l_{Cu} = 48$ Å. Together with the linear slope of the curve we obtain $(\rho l)^{-1}_{Cu} = A_{Cu} = 1.45 \times 10^{15}\ \Omega^{-1}\,m^{-2}$. Chambers[6] gave for Cu the value $(\rho l)^{-1}_{Cu} = (1.54 \pm 0.05) \times 10^{15}\ \Omega^{-1}\,m^{-2}$ while the free-electron value is $1.52 \times 10^{15}\ \Omega^{-1}\,m^{-1}$. Since $A = (e^2/12\pi^3\hbar)$, S is proportional to the Fermi-surface area and the latter is reduced in Cu compared with the free-electron sphere—this result appears very reasonable. We state that the agreement between experiment and the theoretical formula is excellent, the parameter ρl agrees well with otherwise determined values, and quench-condensed films have a homogeneous structure.

For Pd we obtain $l_{Pd} = 47$ Å and $(\rho l)^{-1}_{Pd} = A_{Pd} = 0.79 \times 10^{15}\ \Omega^{-1}\,m^{-2}$. From A we obtain the Fermi-surface area. It corresponds to a free-electron sphere with $z \approx 0.5$ conduction electrons per atom.

For the Cu film we examined whether the upper surface scatters the conduction electrons diffusely. We superimposed the total Cu film with amorphous $Pb_{75}Bi_{25}$. Now we expect that the surface scatters completely diffusely. Indeed the conductance was reduced by $7\times10^{-4}\ \Omega^{-1}$. This is the conductance of a Cu film of 1 Å thickness and corresponds to a specular reflectance of 10%.

For Cu and Pd the mean free path was rather small and most of the experimental data were such that $D > l_M$. Figure 3 shows the experimental results for the other extreme case. For In we find a good fit between experimental data and the theory if we set $l_{In} = 240$ Å and $A_{In} = (\rho l)^{-1}_{In} = 1.03\times10^{15}\ \Omega^{-1}\,m^{-2}$, which is about 55% of the free-electron value. This is in good agreement with the value by Lyall and Cochran[7] who obtained $(0.90 \pm 0.16) \times 10^{15}\ \Omega^{-1}\,m^{-2}$. The theoretical curve agrees exactly with the experimental data points. Because of the restricted thickness region ($D < l_{In}$) one obtains also a good agreement if one changes l_{In} by 5%. In this experiment the mean free path of M is that much larger than in the amorphous substrate that one can neglect any influence of the substrate.

If one quench condenses a metallic film onto a nonmetallic substrate, the first layers have a strongly reduced mean free path (in addition to the formation of islands). Our experiments exclude such an inhomogeneous structure. If we assume, for example, that in the first layer of M with a thickness l_M the mean free path is reduced by a factor of 2, this would shift the asymptotic straight line at large thicknesses by $\sim\frac{3}{8} l_M$. Apart

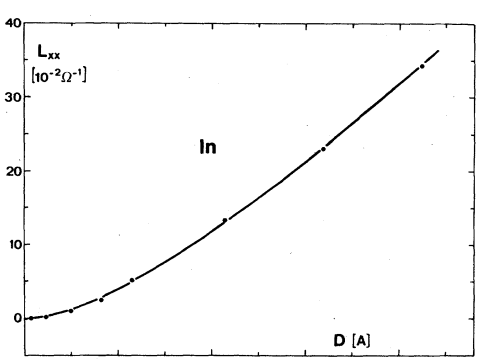

FIG. 3. Change in conductance due to In quench condensed on top of an amorphous metallic substrate is plotted as a function of thickness. The full curve is calculated according to Eq. (5), and yields the values l and $(\rho l)^{-1}$.

from the difficulty in fitting the total experimental curve by a theory, this would produce a (false) mean free path of about $2l_M$. The good agreement between our experimental results for ρl and the otherwise obtained values demonstrates the absence of an inhomogeneous structure.

Bassewitz and Minnigerode[8] investigated metal films quench condensed onto a quartz substrate at He temperature in a vacuum better than 10^{-6} Torr. They concluded from their experimental results that several layers of adsorbed gases were condensed between the quartz plate and the film. In addition, the structure of Cu, for example, was found to be porous. Obviously we can avoid these problems by means of a freshly condensed metal surface as substrate and the ultrahigh vacuum.

IV. CONCLUSION

We have suggested the investigation of geometrical size effects in sandwiches. A theoretical model calculation is performed. This method avoids the serious difficulty of island formation during the condensation of the film. Several new applications offer (a) the investigation of the transparency of the interface between two metals, (b) the use of amorphous metals as substrate, and (c) the direct observation of the penetration of conduction electrons from a ferromagnet into a normal metal due to the anomalous Hall effect in the ferromagnet.

For the examples of quench-condensed Cu, Pd, and In films, it is demonstrated that these films possess a homogeneous structure. The mean free path and ρl are determined.

APPENDIX

According to Chambers the vector mean free path $\vec{\lambda}_{\vec{k}}$ is given by

$$\vec{\lambda}_{\vec{k}} = \int_{-\infty}^{0} \langle \vec{v}(t) \rangle_{\vec{k}}\, dt\,. \tag{A1}$$

Here $\langle \vec{v}(t) \rangle_{\vec{k}}$ is the average velocity at time t of an assembly of electrons all of velocity $\vec{v}_{\vec{k}}$ at $t = 0$.

In the absence of a magnetic field the average velocity is given by

$$\langle \vec{v}(t) \rangle_{\vec{k}} = \vec{v}(t)_{\vec{k}} \exp\left(- \int_t^0 \frac{du}{\tau(u)_{\vec{k}}} \right)\,. \tag{A2}$$

$\tau(u)_{\vec{k}}$ is the relaxation time that the conduction electron $\vec{k}$ experiences at its position at the time u. It is equal to τ_M in the metal M or τ_F in the metal F, respectively. At the upper surface of M, $\tau = 0$. $\vec{v}(t)_{\vec{k}}$ is the velocity of the electron at the time t if the conduction electron does not experience a collision in the time interval between t and 0. The exponential *increase* of $\langle \vec{v}(t) \rangle_{\vec{k}}$ for $t < 0$ with time t has the physical meaning that the amplitude of the electron wave, which has the momentum $\vec{k}$ at the position $\vec{r}$ and the time $t=0$, increases at negative times because other conduction electrons are scattered into the state $\vec{k}$. Since the physical situation and the mathematical analysis are discussed in detail in Ref. 1 we restrict ourselves to this short summary.

We now have to determine the vector mean free path of the conduction electrons in the sandwich. It is a function of the position and the velocity. We consider first the case without a magnetic field ($B=0$). One has to calculate four different situations which are determined by the conditions $z>0$ or $z<0$, and $v_z>0$ or $v_z<0$. We perform the calculation explicitly for the most complicated case $z<0, v_z<0$. In Fig. 1 the path of a conduction electron is plotted which is located at the time $t=0$ at a position $z<0$. It crosses the interface at the time $t_0=-|z|/|v_z^F|$ and started from the upper surface at the time $t_D=-D/|v_z^M|-|z|/|v_z^F|$. The x and y components of the velocity $v_c(t)=v_x(t)=v_x(t)+iv_y(t)=v_\rho e^{i\alpha}$ are given by[9]

$$v_c(t)=\begin{cases} v_c^M & \text{for } t_D<t<t_0 \\ v_c^F & \text{for } t_0<t<0 . \end{cases} \tag{A3}$$

(We distinguish between v^M and v^F although they have the same value in our model.) Therefore, we obtain for $\langle v_c(t)\rangle=\langle v_x(t)\rangle+i\langle v_y(t)\rangle$

$$\langle v_c(t)\rangle=\begin{cases} r v_\rho^M \exp\left(i\alpha+\frac{t_0}{\tau_F}+\frac{t-t_0}{\tau_M}\right), & t_D<t<t_0 \\ v_\rho^F \exp(i\alpha+t/\tau_F), & t_0<t<0 . \end{cases} \tag{A4}$$

The factor r expresses the partial reduction of the electron wave by crossing the interface. Now we obtain the vector mean free path $\vec{\lambda}_{\vec{k}}$ for the considered conduction electron. We calculate the x and y components from $\vec{\lambda}$: $\lambda_c=\lambda_x+i\lambda_y$,

$$\begin{aligned}\lambda_c(z<0, v_z<0) &= \int_{t_0}^{0} dt\, v_\rho^F \exp\left(i\alpha+\frac{t}{\tau_F}\right)+r\int_{t_D}^{t_0} dt\, v_\rho^M \exp\left(i\alpha+\frac{t_0}{\tau_F}+\frac{t-t_0}{\tau_M}\right) \\ &= l_c^F\left[1-\exp\left(-\frac{|z|}{\tau_F|v_z^F|}\right)\right]+r l_c^M \exp\left(-\frac{|z|}{\tau_F|v_z^F|}\right)\left[1-\exp\left(-\frac{D}{\tau_M|v_z^M|}\right)\right].\end{aligned} \tag{A5}$$

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