THEORY OF NEGATIVE MAGNETORESISTANCE IN SMALL SAMPLES AT LOW FIELDS*

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A boundary scattering mechanism is proposed which provides an explanation for low field negative magnetoresistance in metals which have at least one small dimension. Explicit calculations are made for the B field dependence of resistance in thin cylindrical wires and in thin films. It is found, in each case, that as the B field is increased from zero, the resistance decreases in proportion to B^2. An order of magnitude calculation indicates that for metallic films and wires at liquid helium temperature, the change in resistance can be greater than 0.1%.

1. *Introduction*

When the linear size of a metallic crystallite is comparable to the mean free path of an electron in bulk, the electrical conduction becomes distinct from that in a bulk. If a magnetic field is applied, the size effect on the conductivity becomes more apparent. In 1964, Forsvoll and Holwech reported a negative transverse magnetoresistance in a thin film of aluminium subjected to a weak magnetic field directed in the plane of the film [1]. This negative magnetoresistance at low fields appears to have remained unexplained inspite of the fact that a number of theoretical investigations have been reported on the galvanomagnetic effects in thin films and wires [2–6].

Negative magnetoresistance has also been observed in a wide range of carbon (polycrystalline graphite) samples [7]. Recently, one of the present authors (S. F.) proposed to explain this effect by considering the diffuse scattering at crystallite boundaries [8]. A graphite crystal has strong anisotropy, and it is believed that the crystal allows the current to flow mainly in graphitic planes. This makes it possible to consider a two-dimensional, rather than three-dimensional, current flow. The maximum negative magnetoresistance

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due to this mechanism for a carbon is estimated to be 5–10%, which is in agreement with the observation. If a conducting crystal of a three-dimensional character is considered, the effect of a magnetic field on the diffuse scattering will still be present, but smaller in magnitude. This will be shown in the present article.

2. *Diffuse scattering and negative magnetoresistance*

The transverse magnetoresistance is defined by

$$[\varrho(\mathrm{B})-\varrho_0]/\varrho_0 = \Delta\varrho/\varrho_0 \tag{2.1}$$

where $\varrho(B)$ is the electrical resistivity measured in the direction perpendicular to the magnetic field $\boldsymbol{B}$, and ϱ_0 is the resistivity corresponding to the zero magnetic field. The zero-field resistivity ϱ_0 is the inverse of the zero-field conductivity and is given approximately by

$$\varrho_0 \propto m^*\langle v\rangle/(n|e|l) \tag{2.2}$$

according to the simple kinetic theory applied to a single-carrier system, where e, m^*, n, $\langle v\rangle$ and l are respectively, charge, effective mass, density, average speed, and mean free path of the carrier. Among these, the four characteristics e, m^*, n, and $\langle v\rangle$, are unlikely to change appreciably when a weak magnetic field is applied. The change in the mean free path l should then roughly determine the behaviour of the magnetoresistance $\Delta\varrho/\varrho_0$ at low fields. This means that those physical processes which make the mean free path larger for greater values of B should contribute to the negative magnetoresistance. If the increment of the mean free path is denoted by $\Delta l \equiv l(B)-l$, then the magnetoresistance is roughly given by

$$\Delta\varrho/\varrho_0 = -\Delta l/l. \tag{2.3}$$

In most conducting materials the electrons are diffusely scattered at the crystallite boundaries. That is, once electrons arrive at the boundary surface from any direction within a crystallite, they may leave the surface in all possible directions within the crystallite with equal probability. When the linear dimension of a crystallite is of the order 10^2Å, then this diffuse scattering at the boundary becomes one of the predominant processes that influence the mean free path of the conduction electrons.

One can show that if this diffuse scattering is the only mechanism which restricts the free path of a (classical) free electron, the application of a magnetic field B will tend to lengthen the mean free path and hence cause negative magnetoresistance. Consider for simplicity, a rectangular thin layer, a two-dimensional model, in which an electron moves freely. Let's take an electron, starting from the point A (Fig. 1), proceeding on the straight line AC in the absence of a magnetic field and hitting the wall at C. The length EC may be defined as the free path of the electron with respect to the charge transport in the upward direction. If a constant magnetic field of magnitude B is applied in the direction perpendicular to, and into, the paper, the electron will describe a circular orbit of radius $R = mv^*/eB$. Thus, the electron starting from the same point A with the same speed v will travel now on a circular arc AC′ and hit the wall at C′ (OA = OC′ = R). The free path is now EC′ and is shorter than the path EC. $\Delta l_- = \text{EC}' - \text{EC} < 0$.

It is clear from the diagram that in case the electron should preceed so as to hit the wall on the right-hand side in the absence of the magnetic field, its free path will be greatest for $B = 0$ and will become shorter as B increases (and thus R decreases). In a similar manner

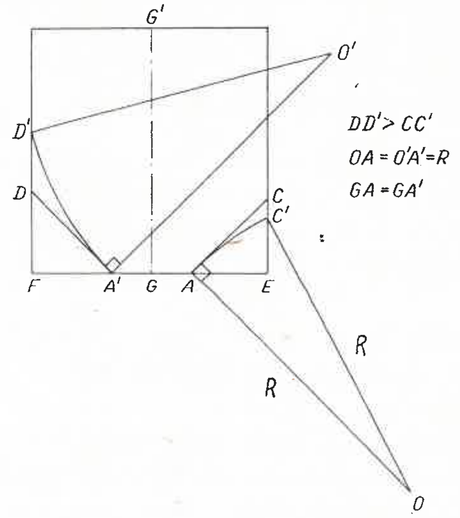

Fig. 1. Graph showing that the mean free path restricted only by the boundary is greater for $B \neq 0$ than for $B = 0$

it is seen that if an electron should hit the wall at the left-hand side for $B = 0$, its free path will become greater as B increases. For example, in the diagram: $\Delta l_+ = \text{FD}' - \text{FD} > 0$. To avoid complications we shall consider the case where D′ can be found on the left wall, which will be assured when B is sufficiently small so that the radius R is sufficiently large compared with the side EF and when the length of the crystal is also sufficiently large.

The balance of the gains and the losses respectively at the left and right walls of the free paths will be now compared. In the absence of magnetic field, to every process in which an electron hits the right wall with an angle θ, there exists a corresponding process in which an electron hits the left wall with the same angle θ after covering the same distance. In general, corresponding pairs can be found by looking at the reflexion symmetry with respect to the axis GG′ which bisects the layer vertically. The two processes indicated in Fig. 1 are just those with this correspondence. From the diagram it is clear that

$$\text{D}'\text{D} > \text{CC}' \quad \text{or} \quad |\Delta l_+| - |\Delta l_-| = \Delta l_+ + \Delta l_- > 0. \tag{2.4}$$

This means that the absolute value of the net gain is greater than that of the net loss.

If the direction of the applied magnetic field is reversed or if the sign of charge of the carrier is reversed, the gains and losses of the free paths occur at the right and left walls respectively. However, it is easily verified that the net change $\Delta l_+ + \Delta l_-$ is again positive.

The argument for the negative transverse magnetoresistance is applicable not only to a small crystallite of graphite but also to a thin film of an isotropic metal. A similar qualitative argument for a negative magnetoresistance in a thin metallic whisker could be developed but will not be described here.

What is the field dependence of the negative magnetoresistance? What is the order of magnitude of such a negative magnetoresistance? These questions will be studied in the following sections.

3. Quantitative analysis for a cylinder and thin film

3.1. General plan

Let us choose a system of coordinates such that the positive x-axis coincides with the direction of an applied constant magnetic field of strength B. An electron will in general describe a helical orbit with the helical axis along the x-axis. The cyclotron frequency ω is given by

$$\omega \equiv \frac{|e|\,B}{m^* c}, \tag{3.1}$$

where c is the speed of light. Let us imagine that an electron starts to move from the point (x_0, y_0, z_0) with a given initial velocity $\boldsymbol{v}$ whose magnitude is denoted by v and whose direction is specified by the polar and azimuthal angles (θ, φ). This situation is depicted in Fig. 2.

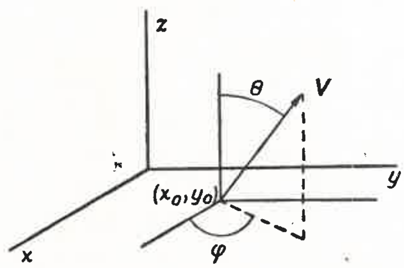

Fig. 2. Diagram showing the initial velocity vector $\boldsymbol{v}$

The helical orbit for such an electron can be described by the equations

$$x(t) = v_x t + x_0 = vt \sin\theta \cos\varphi + x_0 \tag{3.2a}$$

$$y(t) = R\cos(\omega t + \alpha) - R\cos\alpha + y_0 \tag{3.2b}$$

$$z(t) = R\sin(\omega t + \alpha) - R\sin\alpha, \tag{3.2c}$$

where

$$\alpha = \tan^{-1}(-v_y/v_z) \tag{3.3}$$

$$R = v_\perp/\omega = (v/\omega)(\cos^2\theta + \sin^2\theta\,\sin^2\varphi)^{1/2} \tag{3.4}$$

The helical path given by (3.2) is the true orbit followed by the electron if it is disrupted neither by the geometrical boundary nor by scatterers, which are not under consideration at present. Let the equation for the boundary be

$$f(x, y, z) = 0. \tag{3.5}$$

Substitution of (3.2) may yield in general a number of solutions for t. The smallest of these times is the time, t_1, elapsed for the first collision at the boundary. Substituting t_1 in the last equation of (3.2), one can find the distance $z(t_1)$ along the z-direction. When we consider a current flow along the z-direction which is perpendicular to the direction of $\boldsymbol{B}$, this value $z(t_1)$ may be considered as the measure of the free path of an electron.

If we now assume an isotropic velocity distribution at all points, we may define the mean free path to be

$$\langle z(\omega)\rangle_{\text{av}} = \frac{\int d\theta \int d\varphi \int z(\theta, \varphi, x_0, y_0, \omega) \sin\theta \, dx_0 dy_0}{\int d\theta \int d\varphi \int \sin\theta \, dx_0 dy_0} \tag{3.6}$$

where the integration with respect to $dx_0 dy_0$ is over the x, y plane inside the boundary given by $f(x, y, z) = 0$. The limits on the θ and φ integrations will be determined by considering the region of applicability of boundary scattering; this will become clear in the following explicit calculations. The upper limit on the θ integration is $\pi/2$; this is equivalent to the choice

$$v_z(t = 0) \geqslant 0. \tag{3.7}$$

When the magnetic field is not present, the electrons travel along straight lines. The equations of these lines are given by

$$\begin{aligned} x(\omega = 0) &= vt_1 \sin\theta \cos\varphi + x_0 \\ y(\omega = 0) &= vt_1 \sin\theta \sin\varphi + y_0 \\ z(\omega = 0) &= vt_1 \cos\theta, \end{aligned} \tag{3.8}$$

which is the zero field limit of (3.2). The time t_1 for the first boundary scattering is again found through the equation of the boundary, and the average is taken as in (3.6):

$$\langle z(\omega = 0)\rangle_{\text{av}} = \frac{\int d\theta \int d\varphi \int z(\theta, \varphi, x_0, y_0) \sin\theta \, dx_0 dy_0}{\int d\theta \int d\varphi \int \sin\theta \, dx_0 dy_0}. \tag{3.6a}$$

The change in mean free path as a function of ω is, then

$$\Delta l = \langle z(\omega) - z(\omega = 0)\rangle_{\text{av}}. \tag{3.9}$$

Therefore, from (2.3),

$$\frac{\Delta l}{l} = -\frac{\Delta \varrho}{\varrho_0} = \frac{\langle z(\omega) - z(\omega = 0)\rangle_{\text{av}}}{\langle z(\omega = 0)\rangle_{\text{av}}}. \tag{3.10}$$

3.2 Cylindrical wire

With the cylinder of radius r_0 placed with its axis along the z-axis, the boundary equation reads

$$f_c(x, y, z) = x^2+y^2-r_0^2 = 0. \tag{3.11}$$

Given the starting position (x_0, y_0) and velocity vector v of the electron, we may solve (3.2) and (3.11) to find $z(t_1)$. Considering, for example, only those electrons which start at the origin $(x_0 = y_0 = 0)$, we have

$$v^2t^2 \sin^2\theta \cos^2\varphi+R^2 [\cos(\omega t+\alpha)-\cos\alpha]^2-r_0^2 = 0. \tag{3.12}$$

For small magnetic fields the angle ωt will be small (the path differing little from a straight line), and we may solve (3.12) by expansion. Using the expression for R, we obtain

$$(\cos^2\theta+\sin^2\theta\sin^2\varphi)\cdot[\omega t\sin\alpha+(1/2)\omega^2t^2\cos\alpha]^2+$$
$$+\tfrac{1}{2}\omega^2t^2\sin^2\theta\cos^2\varphi-(r_0^2\omega^2/v^2) = 0. \tag{3.13}$$

For the zero field case we have, from (3.8) and (3.11)

$$t_0 = r_0/v\sin\theta. \tag{3.14}$$

We now assume that t_1, the time for the first boundary scattering, differs by only a small amount δt from t_0, the zero field time interval. Substituting $t = t_0+\delta t = r_0/(v\sin\theta)+\delta t$ into (3.13) we find

$$\delta t = (\omega r_0^2/v^2)(\sin\varphi\cot\theta)/[2\sin^2\theta-3(\omega r_0/v)\cos\theta\sin\varphi] \tag{3.15}$$

which becomes upon expansion in powers of $(\omega r_0/v)$

$$\delta t = \tfrac{1}{2}(\omega r_0/v)(r_0/v)(\sin\varphi\cos\theta)/(\sin^3\theta) +$$
$$+\tfrac{3}{4}(\omega r_0/v)^2(r_0/v)(\sin^2\varphi\cos^2\theta)/(\sin^5\theta) +$$
$$+\tfrac{9}{8}(\omega r_0/v)^3(r_0/v)(\sin^3\varphi\cos^3\theta)/(\sin^7\theta). \tag{3.16}$$

The above equation holds only if θ does not approach zero; see below. By expanding the terms on the right-hand side of (3.2c) in powers of ωt, we obtain

$$z(t) = (v/\omega)[\omega t-\tfrac{1}{6}(\omega t)^3]\cos\theta + \tfrac{1}{2}(v/\omega)(\omega t)^2\sin\theta\sin\varphi. \tag{3.17}$$

Using $t = t_0+\delta t$ and $z(\omega = 0) = vt_0\cos\theta$, we obtain

$$[z-z(\omega = 0)] = (v\cos\theta)(\delta t)-\tfrac{1}{6}v\omega^2\cos\theta\,(t_0^3)-\tfrac{1}{2}v\omega^2\cos\theta\,(t_0^2\delta t)+$$
$$+\tfrac{1}{2}v\omega\sin\theta\sin\varphi(t_0^2)+v\omega\sin\theta\sin\varphi(t_0\delta t). \tag{3.18}$$

Using (3.14) and (3.16), we integrate the above expression with respect to θ and φ according to (3.6).

In this integration we restrict the area of interest to those electrons which travel a distance less than the bulk mean free path l_B. Thus, for the zero field case, $vt_0 \leqslant l_B$. Since $t_0 = r_0/(v \sin\theta)$, we have $\sin\theta \geqslant r_0/l_B \equiv 1/k$ which defines the minimum angle θ_{min} to be

$$\theta_{min} = \sin^{-1}(1/k). \tag{3.19}$$

It is noted that the above mentioned restriction of θ is satisfied when we limit the θ-integration in the above fashion. This same θ_{min} will apply very well to the curved paths also, since they differ little from straight lines. The $\langle \Delta z \rangle_{av}$ integral is now given by

$$\langle \Delta z \rangle_{av} = \int_0^{2\pi} d\varphi \int_{\theta_{min}}^{\pi/2} r_0(\omega r_0/v)^2[\tfrac{3}{4}(\cos^3\theta \sin^2\varphi/\sin^5\theta) - \tfrac{1}{6}(\cos\theta/\sin^3\theta) + $$
$$+ \tfrac{1}{2}(\cos\theta \sin^2\varphi/\sin^3\theta)] \sin\theta\, d\theta.$$

Integrating we find

$$\frac{\Delta\varrho}{\varrho_0} = -(1/24)\,(\omega r_0/v)^2\,(3k^4 - 7k^2 + 4k)/(k-1).$$

The speed v is given by the Fermi velocity v_F; setting $\omega = eB/m^*c$ and $F_{cyl} = (1/24)(3k^4 - 7k^2 + 4k)/(k-1)$, we have

$$\frac{\Delta\varrho}{\varrho_0} = -(er_0/cv_F)^2\,(B/m^*)^2 F_{cyl}(k) \tag{3.20}$$

where $k = l_B/r_0$. Knowing the bulk mean free path l_B, we may compute $F_{cyl}(k)$; it is verified that F_{cyl} is positive for all $k > 1$. The geometric factor $F_{cyl}(k)$ will be different if the electrons start at a point other than the origin, but the quadratic B dependence will remain

3.3 Thin film

The analysis of a thin film closely follows that of the wire. Place the film parallel to the x, z plane, with surfaces $y = +a$ and $y = -a$. The field $\boldsymbol{B} = (B, 0, 0)$ is in the plane of the film and will therefore affect the electrons as shown in figure 1. That is, the electrons will follow slightly curved paths in low fields and will collide with either surface $y = +a$ or $y = -a$. The boundary equation reads

$$f_{film} = y - a = 0. \tag{3.21}$$

We again consider only electrons starting at the origin ($y_0 = 0$). Using (3.2b), (3.21) becomes

$$R\cos(\omega t + \alpha) - R\cos\alpha = a$$

or

$$(\omega t + \alpha) = \cos^{-1}[\cos\alpha + (a/R)]. \tag{3.22}$$

The distance traveled along the z-axis is

$$z = R\sin[\cos^{-1}(\cos\alpha + (a/R))] - R\sin\alpha$$
$$= R\{1 - [(a/R)^2 + (2a\cos\alpha/R) + \cos^2\alpha]\}^{1/2} - R\sin\alpha.$$

Assuming low fields we expand the above, and retaining only terms to second order in B we find

$$z = a\cos\theta/(\sin\theta\sin\varphi) + (a^2\omega/2v)[(\sin^2\theta\sin^2\varphi + \cos^2\theta)/$$

$$(\sin^3\theta\sin^3\varphi)] + (a^3\omega^2/2v^2)[(\cos\theta\sin^2\theta\sin^2\varphi + \cos^3\theta)/(\sin^5\theta\sin^5\varphi)]. \quad (3.23)$$

This expansion holds only if both θ and φ do not approach zero. For zero field

$$y(\omega = 0) = vt_0 \sin\theta\sin\varphi = a$$

or

$$vt_0 = a/(\sin\theta\sin\varphi). \quad (3.24)$$

This gives

$$z(\omega = 0) = vt_0\cos\theta = a\cos\theta/(\sin\theta\sin\varphi). \quad (3.25)$$

We again consider only those electrons which travel a shorter distance than the bulk mean free path l_B. That is, $vt_0 \leqslant l_B$, or

$$vt_0 = a/(\sin\theta\sin\varphi) \leqslant l_B.$$

For $\sin\varphi = 1$, $a/\sin\theta \leqslant l_B$ therefore

$$\sin\theta_{\min} = a/l_B \equiv 1/k. \quad (3.26)$$

For given θ, the restriction on φ reads

$$(\sin\varphi)_{\min} = l/(k\sin\theta). \quad (3.27)$$

Expressions (3.26) and (3.27) provide the integration limits and serve to comply with the limitation that neither θ nor φ approach zero. The integral for $\langle\Delta z\rangle_{\text{av}}$ is now

$$\langle\Delta z\rangle_{\text{av}} = \int_{\theta_{\min}}^{\pi/2}\int_{\sin^{-1}[1/k\sin\theta]}^{\pi-\sin^{-1}[1/k\sin\theta]} a(\omega a/v)^2(\cos\theta\sin^2\theta\sin^2\varphi + \cos^3\theta)(\sin^5\theta\sin^5\varphi)^{-1}\sin\theta\, d\theta d\varphi.$$

All of the first order terms in B are zero because of the symmetry of the integrals. This is expected because the change of resistance should be unchanged when the direction of $\boldsymbol{B}$ is reversed (even in B). Upon integration we find

$$\frac{\Delta\varrho}{\varrho_0} = -(1/6)(\omega a/v)^2 \frac{[3k^3(k^2-1)^{1/2} - k(k^2-1)^{3/2} - 3\ln(k+(k^2-1)^{1/2})]}{[\ln(k+(k^2-1)^{1/2}) - (1/k)(k^2-1)^{1/2}]}$$

$$\frac{\Delta\varrho}{\varrho_0} = -(ae/cv_F)^2(B/m^*)^2 F_{\text{film}}(k) \quad (3.28)$$

where

$$F_{\text{film}}(k) = (1/6)\frac{[3k^3(k^2-1)^{1/2} - k(k^2-1)^{3/2} - 3\ln(k+(k^2-1)^{1/2})]}{\ln(k+(k^2-1)^{1/2}) - (1/k)(k^2-1)^{1/2}]}$$

and $k = l_B/a$. The factor $F_{\text{film}}(k)$ is positive for $k > 1$. For the general case $y_0 \neq 0$ we again obtain a negative magnetoresistance, but $F_{\text{film}}(k)$ will change.

3.4 Numerical estimate

To obtain an estimate of the magnitude of negative magnetoresistance, we may use the expressions (3.20) and (3.28). As an example we computed $\Delta\varrho/\varrho_0$ for a film at liquid helium temperature, of thickness $a = 10^{-3}$ cm, $v_F = 10^8$ cm/sec, and $k = l_B/a = 2$; bulk mean free paths, l_B, are often of order 10^{-3} cm in metallic films at liquid helium temperature. For fields of 10, 20, 50, 80, and 100 gauss, we obtain $-\Delta\varrho/\varrho_0$ of 0.003, 0.012, 0.075, 0.192, and 0.3%. Negative magnetoresistance of this order of magnitude should be observable by present-day experimental techniques.

4. Conclusion

For both cylindrical wires and metallic films, we have obtained, at low field intensities, a negative magnetoresistance which varies quadratically with magnetic field. The data of Forsvoll and Holwech appears to be in agreement with our calculation, but their experiments were more concerned with the high field behaviour of magnetorestistance. Further investigation is necessary in order to firmly establish the low field phenomenon.

From expressions (3.20) and (3.28), we notice that $\Delta\varrho/\varrho_0 \propto (1/m^*)^2$; therefore, if the resistance change were measured, it could provide an independent determination of the effective mass of electrons in the specimen.

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