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## LETTER TO THE EDITOR

# Magnetoresistance of single-crystal potassium

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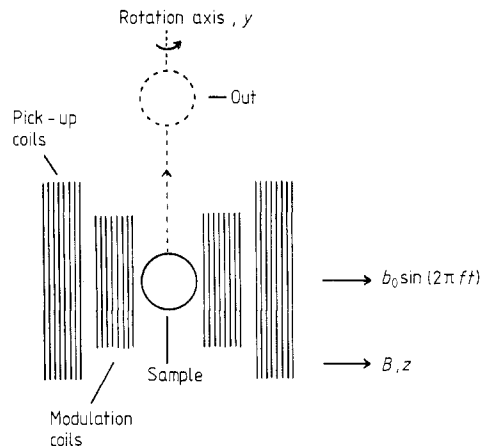
**Abstract.** We report direct measurements of the low-temperature transverse magnetoresistance in potassium crystals in magnetic fields of up to 6 T using an induced-eddy-current method. No evidence for the presence of open orbits is found. These results disagree with Overhauser's charge-density-wave interpretation of the high-field induced torque in potassium made by Coulter and Datars.

During the past two decades several experiments on the electronic properties of potassium have yielded conflicting results. While some experiments appear to be in agreement with conventional views of the electronic structure of the alkali metals, some 'anomalous' results appear only to find explanation in terms of Overhauser's charge-density-wave (CDW) model. Possibly the most dramatic example of such conflicting experiments is de Haas–van Alphen (DHVA) effect measurements and high-field induced torque ( $\text{IT}$ ).

As is well known, DHVA effect studies (Lee and Falicov 1968, O'Shea and Springford 1981, for example) clearly indicate that the Fermi surface of potassium is simply connected and nearly spherical. The small deviations from sphericity which exist, a few parts in a thousand, have been measured with great precision using the DHVA effect. This free-electron-like Fermi surface is in accord with conventional theory, but in complete disagreement with the CDW model.

A free-electron-like Fermi surface should lead to an isotropic and field-independent magnetoresistance. However, measurements of magnetoresistance in potassium by Coulter and Datars (CD) (1980, 1985) using a contactless  $\text{IT}$  method showed a highly anisotropic  $\text{IT}$  upon rotating the sample, with sharp peaks characteristic of open orbits as are seen in the case of copper.  $\text{IT}$  peaks are known to be a consequence of a strong directional dependence of magnetoresistance when open-orbit directions exist in a metal. The apparent observation by CD of open orbits in potassium is therefore in complete disagreement with conventional models, but it has been convincingly explained within the CDW model (see Huberman and Overhauser 1981, 1982 for example). In addition, more recent four-terminal measurements of magnetoresistance in potassium (Soethout *et al* 1987) have also been interpreted using the CDW model (Overhauser 1987).

It is clearly of great importance to a theoretical understanding of the alkali metals that the disagreement between DHVA and  $\text{IT}$  data is resolved. It has been suggested (Overhauser 1987) that pre-selection of samples in DHVA effect studies may explain the discrepancy. On the other hand, helicon resonance experiments in potassium (de Podesta and Springford 1986, 1987) have detected no open orbits and it has been



**Figure 1.** Schematic diagram of the induced-eddy-current method. Modulation coils provide an alternating magnetic field  $b_0 \sin(2\pi ft)$  parallel to the main magnetic field  $B$ . The induced magnetic moment of the sample is detected with the pick-up coils. An extra set of modulation and pick-up coils (not shown) connected in series opposition reduces direct pick-up between the modulation and pick-up coils. The sample may be rotated about the vertical axes and removed from within the coils.

suggested (Elliott *et al* 1988) that the IT results may be an experimental artefact, rather than intrinsic to potassium. We feel that it is therefore valuable to make an independent verification of the IT results. In this Letter we report new direct measurements of the high-field magnetoresistance of potassium, made using a contactless induced-eddy-current method.

The experiments were performed at temperatures in the range 1.3–4.2 K in transverse magnetic fields of up to 6 T provided by a split-pair magnet. Vertical access to the transverse field allows the sample to be rotated to examine any field direction in a plane. Samples may be removed and replaced without warming the system above liquid helium temperature.

In figure 1 the induced-eddy-current method for measuring magnetoresistance is shown. An alternating magnetic field at a frequency  $f$  of the form  $b_0 \sin(2\pi ft)$  is superimposed upon the main magnetic field  $B$  by modulation coils. Both  $B$  and the modulation field are along the  $z$  axis. The modulation field induces eddy currents in the sample which therefore has an induced alternating magnetic moment  $m$ . This alternating moment is detected by the voltage induced in a pick-up coil in surrounding the sample. The sample can be withdrawn from the coil in order to determine a reference level. This is necessary because although an astatically wound pair of modulation and pick-up coils was used to prevent direct coupling between the modulation and pick-up coils, in practice it is impossible to achieve perfect balancing to give zero signal with no sample within the coils.

The induced magnetic moment for a spherical sample of radius  $a$  is given at low frequency (neglecting longitudinal–transverse mixing) by

$$m = -\frac{2}{15}\pi a^5 b_0 2\pi f \cos(2\pi ft) / \rho_T \quad (1)$$

where  $\rho_T$  is the transverse magnetoresistivity  $(\rho_{xx} + \rho_{yy})/2$ . This formula is most easily derived from equations (2.8) and (2.15) of Visscher and Falicov (1970), where the changing magnetic field must now be taken parallel to the main magnetic field, instead

of at right angles to it as in the case of the IT geometry. The reader is also referred to the discussion by Delaney and Pippard (1972), § 4.1. Our eddy-current method thus provides a simple and direct measurement of transverse magnetoresistance.

For a free-electron metal,  $\rho_T$  should be isotropic and independent of magnetic field and given by

$$\rho_0 = m^*/ne^2\tau \quad (2)$$

where  $m^*$ ,  $n$ ,  $e$  and  $\tau$  are the electronic effective mass, number density, charge, and scattering lifetime, respectively. The other case of interest is when the field is perpendicular to an open-orbit direction. It is simplest to model this case in terms of a conductivity tensor arising from a cylindrical portion of Fermi surface pointing along the open-orbit direction, plus that due to a spherical Fermi surface (Fletcher 1982, Huberman and Overhauser 1982). The cylinder is supposed to contain a small fraction  $\eta$  of the total electron number. It is straightforward to show that this model yields a transverse magnetoresistivity

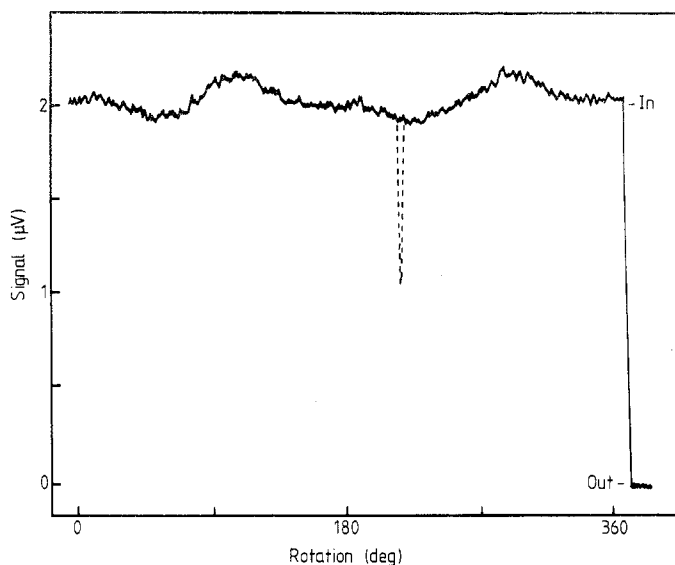
$$\rho_T = \rho_0(1 + \eta\omega_c^2\tau^2/2) \quad (3)$$

where  $\omega_c = eB/m^*$  is the cyclotron frequency. For small  $\omega_c\tau$  (low fields) or for directions in which there are no open orbits, the magnetoresistivity is  $\rho_0$ . For large  $\omega_c\tau$  (high fields) the magnetoresistivity is increased, with consequent decrease in the induced magnetic moment. Rotation of the sample will therefore reveal open-orbit directions by dips in the magnetic moment whenever the open-orbit is in the  $xy$  plane, perpendicular to the magnetic field. (Note that in the case of IT a peak is produced, rather than a dip.)

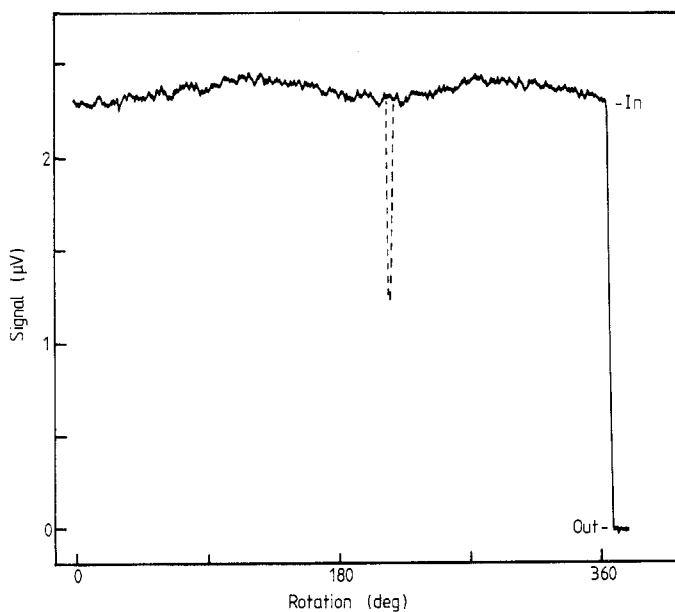
In addition to detecting the induced magnetisation of the sample, the above method had the advantage of being sensitive to the DHVA magnetisation. Indeed, essentially identical methods have been employed in DHVA-effect measurements in the past, using vertical-field magnets. (These experiments could not, however, measure magnetoresistance.) The two effects are easily separated by operating above about 2 K, where DHVA oscillations are negligible, or by detecting at the second harmonic of the modulation frequency, where only the non-linear DHVA magnetisation contributes. The DHVA effect could therefore also be used to make a simultaneous study of the Fermi surface parameters of our samples.

A large number of single-crystal potassium spheres was examined using the eddy-current method. Samples of about 3 mm diameter were grown by slow cooling of molten potassium spheres suspended in hot paraffin oil. The samples were held in sample holders by frozen oil during the experiments. The samples were thus similar to those used by CD in their measurements of oil-grown samples. The crystals' residual-resistance ratios (RRR) (measured *in situ* using the eddy-current method at room and helium temperature) were in the range 2500–5000.

Figures 2 and 3 show the induced magnetic moment of two potassium samples, K25N and K25M, measured with the induced eddy current method. In these, and all other samples examined to date (a total of fifteen samples), there is no evidence of the presence of open orbits. An open orbit would, in the simple sphere-and-cylinder model discussed above, give rise to a sharp drop in the signal at some particular angle, corresponding to the large transverse magnetoresistivity when the open orbit is excited. The large number of open-orbit directions apparent in CD's data and expected according to the CDW model could not escape detection in these measurements. Rotation of the crystal about the  $y$  axis must bring any open orbit into the  $xy$  plane at some point. (The angle between the open orbit and the  $x$  axis in the  $xy$  plane affects only the width of the

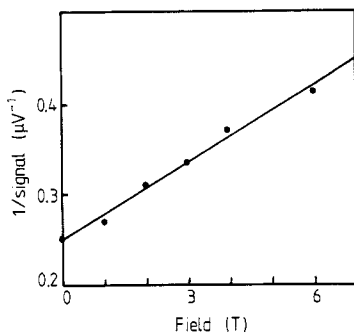


**Figure 2.** Pick-up signal (proportional to the induced magnetic moment) as a function of angle of rotation for potassium sample K25N. The modulation frequency was 2.40 Hz. The dip indicated (broken curve) is the expected open-orbit signal assuming an open-orbit fraction  $\eta = 10^{-4}$ . This sample had a RRR of 3450. ( $\omega_c\tau = 128$  at  $B = 6$  T.) The temperature was 4.2 K.



**Figure 3.** As figure 2 but for sample K25M. The RRR was 3480. ( $\omega_c\tau = 130$  at  $B = 6$  T.)

dip, not its magnitude.) To compare these results to the IT results of Coulter and Datars, we may use equation (3) to make an estimate of the size of signal expected in the presence of an open orbit. This is indicated by the dips marked on figures 2 and 3 for an open-



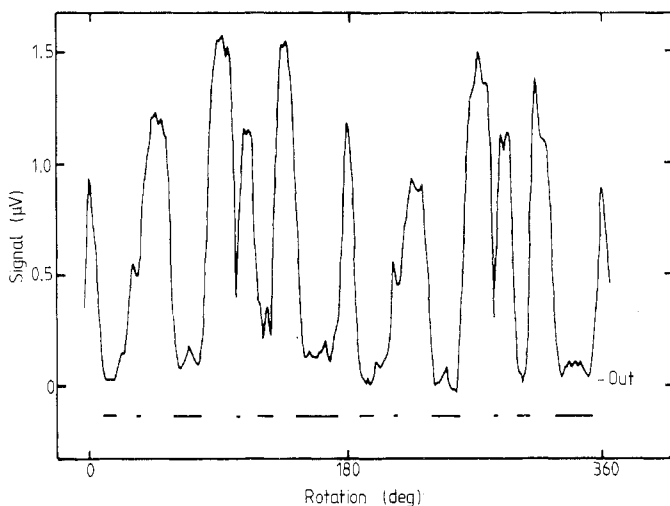
**Figure 4.** Field dependence of reciprocal of pick-up signal (proportional to magnetoresistance) for sample K25N at 4.2 K and an angle of rotation of  $79^\circ$ . A modulation frequency of 1.87 Hz was used. A least-squares fit to the data yields a Kohler slope of  $(5.3 \pm 0.4) \times 10^{-3}$ .

orbit fraction equal to that calculated by CD from their IT data. From the noise level of our data it can be estimated that  $\eta$ , the open-orbit fraction, is not greater than about  $5 \times 10^{-6}$  in our samples K25N and M. In other samples studied, the noise level restricted the maximum value of  $\eta$  to a few times  $10^{-5}$ .

It is interesting to note that there was some anisotropy in the angular dependence of the induced magnetic moment for all samples studied. The anisotropy was always twofold (that is, having two maxima in  $360^\circ$ ) and of the order of a few per cent (about 10% and 6% in samples K25N and K25M respectively at a field of 6 T). The most obvious explanation of this would be a slight departure of the samples from perfect sphericity. It is easy to show, by extending the results of Lass (1976) to our geometry, that for an ellipsoidal sample a shape anisotropy of  $\epsilon\%$  will result in two-fold variation in induced moment, also of  $\epsilon\%$  anisotropy. However, the anisotropy should be field-independent according to this picture and in those samples where the field dependence was examined the anisotropy was found to grow from a smaller zero-field value. Although we have no explanation for these results, experimentally this effect would lead to a directional dependence in the Kohler slope, an effect which is well documented in potassium using different experimental techniques (see e.g. Holyroyd and Datars 1975, O'Shea and Springford 1981). This point is to be examined in more detail.

The field dependence of the induced moment for sample K25N at fixed angle of rotation is shown in figure 4. In this case the inverse of the moment is plotted, as this is proportional to the transverse magnetoresistance. The magnetoresistance is linear in field with a Kohler slope of  $5.3 \times 10^{-3}$ . This value is typical of measurements on other samples and is consistent with reported values in potassium made using different methods.

Since in our experiments equation (1) is a low-frequency approximation for the induced magnetic moment, we ought strictly to make measurements at as high a frequency as possible in order to improve the signal-to-noise ratio, but at a sufficiently low frequency for the approximation (1) to be valid. For an isotropic resistivity  $\rho_0$ , equation (1) agrees with the exact expression for arbitrary frequency (Landau and Lifshitz 1960) to better than 4% as long as the skin depth  $\delta$ , defined by  $\delta = (\rho_0/\pi\mu_0f)^{1/2}$ , is greater than the sample radius  $a$ . Since potassium at helium temperatures is a very good conductor, this means restricting the maximum frequency used to about 3 Hz, depending on the size and conductivity of the sample. (The sample conductivity was calculated from the RRR.) The measurements presented here are well below this limit. Of all the samples studied, nine were examined in the low-frequency region while for the remainder a modulation frequency of up to 10 Hz was employed.



**Figure 5.** Pick-up signal versus angle for a copper crystal at 4.2 K in a field of 4.6 T. Also shown (solid curves and dots below the rotation diagram) are the directions of open-orbit dips expected for rotation of copper in a  $\langle 110 \rangle$  plane. The exact shapes of the open-orbit dips depend on the details of the Fermi surface parameters and sample shape (approximately a cube in this case).

DHVA measurements were originally intended to be used to relate any observed open-orbit directions with the crystallographic orientation. Although not necessary in view of null results of the magnetoresistance studies, DHVA measurements were made on some samples and enabled the crystal orientation to be determined. An extensive study of the DHVA effect was not made, but no unusual behaviour was noted in any of our samples.

In order to confirm that the induced-eddy-current method is sensitive to the presence of open orbits, similar measurements were made on a copper sample, shown in figure 5. The structure observed is the clear signature of open orbits and agrees well with the expected open-orbit directions (Klauder *et al* 1966) in this sample whose plane of rotation was known to be approximately  $\langle 110 \rangle$ . Measurements were made on copper in fields up to 6 T and over a range of modulation frequencies from 1.9 to 12 Hz. Open orbits were clearly evident at all modulation frequencies.

In summary, we have developed a straightforward method enabling high-field magnetoresistance to be measured as a function of crystalline orientation. A study of copper confirms that open-orbit magnetoresistance structure can easily be detected with this method. In the case of potassium, however, our data show no evidence for the presence of open orbits. This is in agreement with previous DHVA measurements of the Fermi surface of potassium and with helicon-resonance experiments, which have also failed to detect open orbits. However, our results clearly contradict the high-field IT studies by CD and their CDW interpretation by Overhauser. In view of the simplicity and directness of our experimental method, and the combined weight of evidence against open orbits in potassium, we believe that the high-field IT results should now be treated with some doubt. It is possible to explain many features of the IT data by hypothesising that friction in CD's IT apparatus is not negligible (Elliott *et al* 1988). We feel that this is the most likely explanation. Perhaps one should also bear in mind (despite the lack of quantitative theory) that since high-field IT experiments measure only a very small component of the

total induced magnetic moment, slight perturbation of the induced-current path by sample defects such as surface irregularities may be important (Fletcher 1982). In our eddy-current method, however, the total magnetic moment is detected. In any case, an intrinsic, open-orbit, explanation of the high-field IT data appears unlikely. Our present measurements provide, we would argue, strong evidence in support of the conventional view of a near-spherical Fermi surface for potassium.

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