RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 41, NUMBER 12 15 APRIL 1990-II

Hall effect in a single two-dimensional quasicrystal: $Al_{62}Si_3Cu_{20}Co_{15}$

Zhang Dian-lin, Lu Li, Wang Xue-mei, and Lin Shu-yuan
Institute of Physics, Chinese Academy of Sciences, 100080 Beijing, China

L. X. He
Laboratory of Atomic Imaging of Solids, Institute of Metal Research, Chinese Academy of Sciences, 110015 Shenyang, China

K. H. Kuo
Beijing Laboratory of Electron Microscopy, Chinese Academy of Sciences, 100080 Beijing, China

(Received 31 January 1990)

Measurements made on single $Al_{62}Si_3Cu_{20}Co_{15}$ two-dimensional decagonal quasicrystals show that when current flows along the periodic direction, the Hall coefficient is positive and temperature independent, while the Hall resistivity is negative and increases with temperature when current flows in the quasicrystalline plane. This is apparently the first observation of the anisotropic Hall effect in quasicrystals.

Since the first report of a rapidly quenched Al-Mn alloy showing quasicrystalline (QC) symmetry,[1] a great deal of knowledge has been accumulated about this kind of material.[2] Many experiments on transport properties showed that the behavior of a QC is very similar to that of metallic glasses. There were also indications that the transport properties of the structure are close to their crystalline rather than to their amorphous counterpart.[3] In fact, the existence of large numbers of defects in quenched QC's may shield the intrinsic properties unique to aperiodic crystals. Recently, a stable two-dimensional QC (2D QC) was found in Al-Si-Cu-Co and single quasicrystals of mm size have been obtained.[4] The high degree of perfection and the two dimensionality of the QC's provide an opportunity to compare the physical properties of a QC with its crystalline counterpart in the same sample, which may be an important step toward an understanding of the electronic properties of the structure. Here we report the measurements on the unusual anisotropic Hall effect of the 2D QC.

The $Al_{62}Si_3Cu_{20}Co_{15}$ QC was formed by slow solidification.[4] The samples were found to be well-formed prisms with a tenfold rotationary symmetry and to be a single quasicrystal over a whole sample which usually was a few tenths of a mm in size. The standard 5-probe ac method was used in preliminary measurements. However, the accuracy was not so satisfactory because the small sample width b (usually less than 0.1 mm) limited the measuring current, and when we used an external potentiometer to balance the zero-field signal, a small temperature drift or a change of contact resistance by the magnetic field could result in an off-balance signal, which sometimes could be comparable to the Hall voltage. To improve the measurements we have developed a method using current instead of voltage compensation. The arrangement is shown in Fig. 1. Two floated current sources (either ac or dc) that have high source impedance and are isolated from each other are applied to the sample in a configuration shown in this figure. Probes 5 and 6 are used to measure the Hall signal. By keeping I_1 constant and adjusting I_2, the voltage across probes 5 and 6 in zero field was balanced to lower than 10^{-8} V in the dc mode and 2×10^{-9} V in the ac mode. Probe 7 is grounded, which greatly stabilizes and decreases the common-mode signal. All the probes were made of Pt wires, 20 μm in diameter and spot-welded to the samples. The new method is proved to be effective in eliminating the zero drift and improving the accuracy of the Hall measurements. Detailed analysis[5] shows that in this configuration, the Hall voltage can be expressed by

$$V_H = \gamma R_H (I_1 + I_2) B/d\,,$$

where γ is a geometry factor weakly depending on b/a (a is the length in current direction), and is not much greater than unity ($\gamma < 1.2$ when $b < a$), d is the sample thickness.

The Hall voltage was measured by sweeping the field from -6 T to 6 T at constant temperature. Four samples have been measured. For samples 1 and 3 the current was

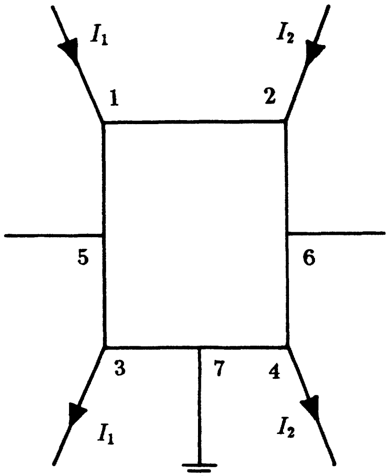

FIG. 1. The arrangement of the 7-probe method for the measurement of the Hall effect.

along the crystalline direction and the applied field was in the QC plane, while for samples 2 and 4 the current was in the QC plane and the field was along the tenfold axis. The dimensions of the four samples were, respectively, $1.1\times0.15\times0.1$, $0.15\times0.09\times0.06$, $0.14\times0.07\times0.02$, and $0.16\times0.09\times0.013$ mm^3. The total measuring current was ~10 mA in the 5-probe measurements and the 7-probe dc measurements. In the 7-probe ac (200 Hz) method I_1+I_2 was about 1 mA. All these measurements gave generally consistent results though with different accuracies.

For samples 1 and 3, where the current was parallel to the periodic direction, the results were surprisingly simple: the Hall coefficient $R_{H,C}$ is positive and temperature independent, which seems to imply a negligible temperature-dependent scattering effect from relaxation anisotropy and possibly rather simple morphology of the Fermi surface. The results are shown in Fig. 2. We see that the 7-probe ac method gives satisfactory temperature-independent $R_{H,C}$, while rather scattered data are obtained by other methods. However, the average value around which the data stagger is close to the value from the 7-probe ac measurement. Considering the least-off-balance signal and the very good stability of the latter method, it is reasonable to claim a temperature-independent $R_{H,C}$, which is about 2×10^{-4} cm^{3}C^{-1}. This yields a carrier concentration $n=3.2\times10^{22}$ cm^{-3} in the free-electron model neglecting the positive sign. Using the residual-resistivity value in the periodic direction of our best sample, $\rho_C\sim50$ $\mu\Omega$ cm,[6] we get a Hall mobility $\mu_H\sim4$ cm^2/V s. Again, assuming a free-electron density of states (DOS), the electron diffusivity D reaches about 10 cm^2/s, or a mean free path $l\sim10$ Å, taking the Fermi velocity to be 1×10^8 cm/s. The diffusivity should be even larger if the actual DOS is lower than its free-electron value.[3] It is interesting to note that the l value is close to the period in the crystalline direction (~8 Å).[4]

Many alloys containing transition metals exhibit a positive Hall coefficient, which is often attributed to the S-shaped anomaly in the $E(k)$ dispersion relation of the s electrons near the Fermi energy due to the effect of s-d hybridization.[7] The positive sign of the present material may originate from the same mechanism. The holelike behavior in the periodic direction was confirmed by thermopower measurements.[6] Both the positive $R_{H,C}$ and positive thermopower are likely to occur for conduction electrons hybridized with the d state of a transition element near the middle of B group.[8] To make the results of the Hall measurements consistent with the thermopower data,[6] the Fermi energy should be located at a position where a marked decrease in DOS takes place.

While satisfactory results were obtained when current flowed along the tenfold axis, the data became less certain for samples 2 and 4 where the current was in the QC plane even with our modified configuration. This may be caused by some inhomogeneity in the QC plane as indicated by the fact that for sample 4 we could not keep the balance across probes 5 and 6 unchanged when the temperature went down, in contrast to sample 3 for which no off-balance signal was observed when the temperature was decreased from 300 to 4.2 K. A further support to the anisotropic inhomogeneity was that, while for sample 3 the Hall signal was almost symmetrical with respect to the reverse of the magnetic field, the Hall signal of sample 4 was quite unsymmetrical, indicating a substantial contribution from uncompensated magnetoresistance (Fig. 3). In addition, it seemed that the noise in the QC plane was higher than in the periodic direction. It is not clear why this inhomogeneity did not manifest itself in the crystalline direction. Maybe some microcracks aligned along the axis were responsible for the apparent anisotropic inhomogeneity.

However, the measurements have shown unambiguously the negative sign of the Hall coefficient $R_{H,\mathrm{QC}}$ for samples 2 and 4, and the general trend that $|R_{H,\mathrm{QC}}|$ increases with temperature (Fig. 4). The temperature dependence of $R_{H,\mathrm{QC}}$ is certainly not caused by any second phase, because the random existence of other phases should smear out the anisotropy of the Hall effect, and the structural analyses also showed a single phase, as well as the very good single-quasicrystal nature of the samples.[4] If we separate $R_{H,\mathrm{QC}}$ into an ordinary term whose absolute value is not far from $R_{H,C}$ and an extraordinary term which rapidly inflates as temperature goes up,

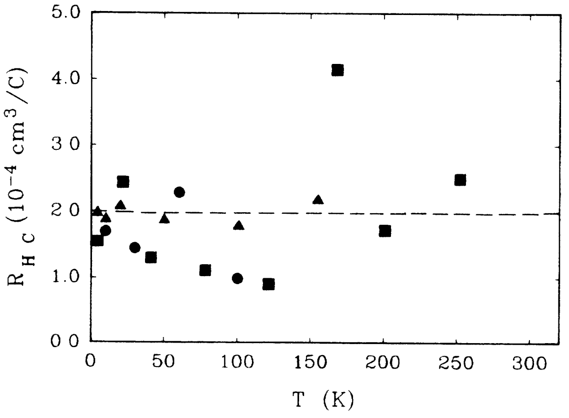

FIG. 2. The Hall coefficient $R_{H,C}$ of the $Al_{62}Si_3Cu_{20}Co_{15}$ quasicrystal, current along crystalline direction. ●, sample 1, 5-probe ac method; ■, sample 3, 7-probe dc method; ▲, sample 3, 7-probe ac method.

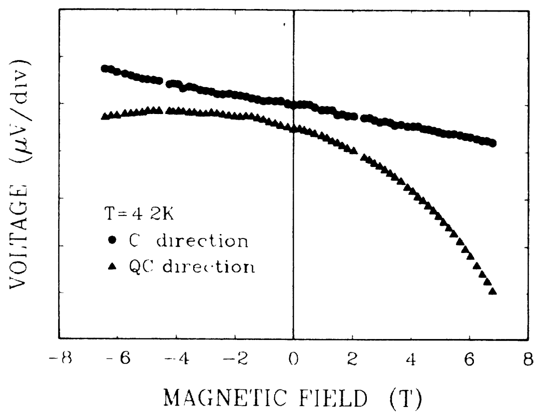

FIG. 3. The dependence of the "Hall voltage" against the magnetic field. We see that it is nearly symmetrical for sample 3, but quite unsymmetrical for sample 4.

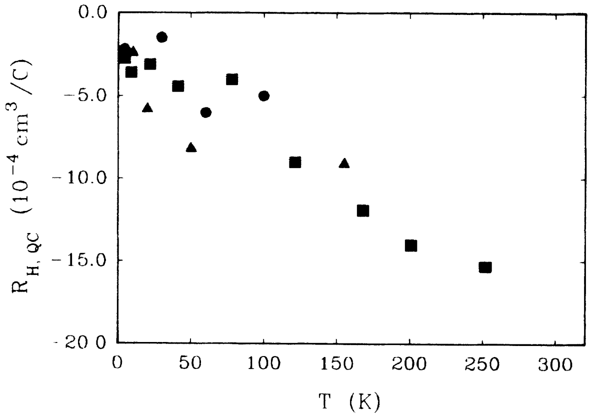

FIG. 4. The Hall coefficient $R_{H,QC}$ of the $Al_{62}Si_3Cu_{20}Co_{15}$ quasicrystal, current in the QC plane and magnetic field along the crystalline direction. ●, sample 2, 5-probe ac method; ■, sample 4, 7-probe dc method; ▲, sample 4, 7-probe ac method.

we see from Fig. 4 that with decreasing temperature $R_{H,QC}$ approximates to the ordinary term (alternatively, we may assume a very large ordinary term and an anomalous term with opposite sign, but in that case the ordinary Hall coefficient is not so consistent with $R_{H,C}$ as well as with the resistivity data).[6] The anomalous Hall effect is often observed in magnetic systems and is believed to be caused by spin-orbit coupling (skew scattering or side jump).[9] The skew scattering varies like the magnetization, that is, decreases when temperature increases, which does not agree with the present results. Since scattering plays a very important role in the side-jump mechanism, the itinerant *d* electrons or weakly localized conduction-band electrons which undergo frequent collisions may contribute little to conductivity, but much to the side jump.[9] To result in a substantial enhancement of $R_{H,QC}$ when the temperature goes up, either the contribution of collision frequency which increases with temperature should surpass the weakening of spin alignment, or the material is in a state of spontaneous magnetization which only weakly depends on temperature. However, the magnetizationlike nonlinearity of the Hall voltage against field was not observed up to 6 T for the present system. Furthermore, if the extraordinary term originates from spin-orbit coupling, we should admit that the spins are liable to be polarized along the crystalline direction, otherwise one would also find this anomalous term in $R_{H,C}$. Further research on magnetic properties is needed to clarify whether this magnetic anisotropy exists or not.

Ignoring the extraordinary term, it is safe to say that the ordinary Hall coefficient in the QC plane is negative, which was also confirmed by the negative sign of the thermopower in the plane.[6] The holelike behavior along the periodic axis and electronlike behavior in the QC plane is really unusual. Results for the Hall coefficient of transition metals in their amorphous, liquid, and crystalline phase, apart from those with a nearly full or empty *d* band, all have a positive Hall coefficient regardless of their state, which suggests that the positive Hall effect is not a structural effect but is something intrinsic to the electronic structure of transition metals.[7] Therefore, one should expect that the effect works equally well for both the QC and crystalline directions. However, if we admit that the holelike behavior of the present material originates from the hybridization of electron states, the immunity of electron dynamics in the QC plane to the hybridization implies that the structural anisotropy does influence the effect: either by *d*-*d* correlation (in consistence with the anisotropy of the anomalous Hall effect) which makes the *d* orbit orderly oriented and the hybridization energy largest along the periodic direction, or by the anisotropy of the energy dispersion relation of conduction electrons which makes the hybridization very ineffective in the QC plane. It would be interesting to determine if this striking contrast in carrier behavior along different directions is only a consequence of the layered structure of the material, or has something to do with the existence or lack of translational periodicity. The lack of knowledge about the atomic arrangement at present prevents us from further discussion.

In summary, we have shown for the first time that in a single 2D quasicrystal the dynamic properties of current carriers are quite different between the periodic and QC directions. Although the temperature coefficients of resistivity show different signs for crystalline and quasicrystalline directions, it is difficult to draw a conclusion that these opposite signs are intrinsic to the 2D QC because of the rather sample-dependent values.[6] At present we have obtained no definite results about magnetoresistance, which also showed strong sample dependence. It is interesting to note that both the resistivity and magnetoresistance are very sensitive to scattering. However, the thermopower[6] and the Hall coefficient are much less sample dependent. We have confidence that the anisotropy shown by thermopower and Hall effect is intrinsic to the present material. A detailed knowledge of the tiling pattern and atomic arrangement would be helpful to clarify if this difference in carrier properties is intrinsically related to the difference in periodic and aperiodic structures.

---

[1] D. Shechtman, I. Blech, D. Gratias, and J. W. Chen, Phys. Rev. Lett. **53**, 1951 (1984).

[2] See the review article by T. Janssen, Phys. Rep. **168**, 55 (1988), and reference therein.

[3] J. L. Wagner, K. M. Wong, and S. J. Poon, Phys. Rev. B **39**, 8091 (1989).

[4] L. X. He, Y. K. Wu, X. M. Meng, and K. H. Kuo, Philos. Mag. Lett. (to be published).

[5] Lu Li and Zhang Dian-lin (unpublished).

[6] Lin Shu-yuan, Wang Xue-mei, Lu Li, Zhang Dian-lin, L. X. He, and K. H. Kuo (unpublished).

[7] See the review article by M. A. Howson and B. L. Gallagher, Phys. Rep. **170**, 265 (1988).

[8] M. A. Howson and G. J. Morgan, Philos. Mag. **51**, 439 (1985).

[9] C. M. Hurd, in *The Hall Effect and its Applications*, edited by C. M. Chien and C. R. Westgate (Plenum, New York, 1980), p. 1.