Sign reversal of the Hall resistivity in amorphous Mo₃Si

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We observe a sign reversal in the Hall resistivity ρ_{xy} of the conventional superconductor amorphous (a) Mo₃Si. In the Ohmic regime, ρ_{xy} is qualitatively similar to that observed in the high- T_c superconductors. It changes sign near T_c , and the sign change persists until both ρ_{xy} and ρ_{xx} become immeasurably small at $T \sim 0.8T_c(H)$. At current densities above the depinning current density, the Hall anomaly persists at low temperatures $T \sim 0.2T_c(H)$. This is contrary to a theory by Ferrell which attributes the anomaly to the backflow of thermally excited quasiparticles. In addition a model proposed by Harris, Ong, and Yan explains the anomaly as an effect arising from the layered nature of the high- T_c cuprates. This model, however, does not explain the anomaly in a-Mo₃Si which is an isotropic unlayered material.

The Hall resistivity in nearly all high-temperature superconductors changes sign when temperature is lowered below T_c into the mixed state.¹ By contrast, a sign change has not been reported for most conventional superconductors,²⁻⁷ and uncertainty regarding sample quality in older work has caused questions to be raised regarding the validity of reported sign changes⁸⁻¹¹ in conventional superconductors.

In the mixed state of a type-II superconductor, vortex motion generates an electric field which results in a flux-flow resistivity. This electric field is given by the Josephson relation¹²

$$\mathbf{E} = -\mathbf{v}_{v} \times \mathbf{B} , \qquad (1)$$

(where **E** is the measured electric field, \mathbf{v}_v is the vortex velocity, and **B** is the magnetic field). From Eq. (1) it follows that vortex motion parallel to the transport current corresponds to a Hall electric field E_y while perpendicular motion corresponds to a dissipative longitudinal electric field E_x . When the vortices in a superconductor have a component of velocity upstream to the transport superfluid velocity \mathbf{v}_s , as determined by the normal-state Hall resistivity, the resulting Hall electric field has a sign opposite to that of the normal state.

Currently there is no generally accepted explanation for the Hall anomaly, although a variety of explanations¹³⁻¹⁸ do exist. Many explanations take a phenomenological approach; they consider forces which act on a vortex and attribute the Hall anomaly to the resulting vortex motion upstream to v_s . Early models of vortex motion^{19,20} predicted a Hall resistivity with the same sign as in the normal state. Since the discovery of high- T_c superconductors, several new models of vortex motion have been proposed to account for the sign anomaly. These models, however, remain controversial.

We have recently performed measurements of the Hall effect on amorphous (a) Mo₃Si, an isotropic lowtemperature superconductor with $T_c = 7.5$ K. This material shows unambiguous and reproducible Hall-effect sign reversal. In a recent paper Harris, Ong, and Yan¹⁶ offer an explanation for the anomalous Hall effect in $YBa_2Cu_3O_7$ (YBCO) based on its layered structure. Their model provides an appealing explanation for the sign change in YBCO, but it predicts no sign change in conventional isotropic unlayered superconductors. Therefore, such a model cannot be a general explanation for the effect. In addition to being an unlayered material, a-Mo₃Si has very weak flux pinning and is a dirty superconductor $(l \ll \xi_0$ where l is the mean free path and ξ_0 is the BCS coherence length) and thus adds further experimental evidence against the theory of Wang and Ting,¹⁷ which says that strong pinning in a moderately clean material $(l \sim \xi_0)$ causes the sign reversal. The sign reversal also persists to very low temperatures in this material, contrary to the theory of Ferrell.¹⁸ Ferrell attributes the Hall anomaly to a force due to thermally excited quasiparticles which drives vortices upstream to v_s . However, the number of thermally excited quasiparticles and therefore this force should be small at low temperatures.

We have measured the Hall and longitudinal resistivities of thin-film samples of a-Mo₃Si in the mixed state. The magnetic field is applied perpendicular to the films with a superconducting solenoid which has a maximum field of 9.0 T. The magnetic field is ramped from high to low field at both polarities while the temperature is held constant; the Hall resistivity is taken as the component of the transverse resistivity odd in the applied field. The samples range in thickness from 250 to 500 Å and have critical temperatures $T_c = 7.5$ K. The widths of the transitions are ≈ 100 mK, indicating that these samples are more microscopically disordered than thicker samples which have slightly higher critical temperatures (7.8-8.0 K) and narrower transition widths. Sample preparation and characterization details are given elsewhere.²¹

Figure 1 shows ρ_{xy} and ρ_{xx} plotted as a function of the applied magnetic field for several temperatures ranging from 3.3 to 7.0 K. In the normal state the Hall resistivity is positive, and, as the applied field nears H_{c_2} , ρ_{xy} changes sign reaching a negative peak when ρ_{xx} has fallen to half its normal-state value. Finally, at a value of magnetic field a few tesla below H_{c_2} , both ρ_{xy} and ρ_{xx} become immeasurably small. The magnitude of the negative peak increases steadily with decreasing temperature. This behavior is qualitatively similar to what is observed in YBCO¹⁵ except that in YBCO the magnitude of the negative peak in ρ_{xy} eventually reaches a maximum as the temperature is lowered and then disappears with a further decrease in temperature.

To reduce the effect of pinning we measure at currents in excess of the depinning current density (see Figs. 2 and 3). In these measurements the non-Ohmic resistance $R \equiv V/I$ is observable to magnetic fields as low as 1 T. The depinning current density in a-Mo₃Si is very small compared with that of most superconductors $(j_c \approx 500)$ A/cm² at T = 4.2 K and 1 T < $\mu_0 H < 4$ T). This current density is easily exceeded while sample heating (≤ 15 mW/cm²) is kept to a minimum.] In Fig. 2 both R_{xy} and R_{xx} are plotted as a function of magnetic field for various current densities at a temperature T = 4.2 K. The Ohmic behavior shown in Fig. 1, measured at j = 15 A/cm², is replotted for comparison with the non-Ohmic data. Over the range of magnetic field measured, R_{xy} is negative and reaches a value comparable to the negative peak observed in the Ohmic data. In low magnetic fields R_{xx} increases approximately linearly with increasing field, while at



FIG. 1. Ohmic Hall and longitudinal resistivities ρ_{xy} and ρ_{xx} (measured at $j = 15 \text{ A/cm}^2$) as a function of magnetic field for several temperatures.



FIG. 2. Ohmic and non-Ohmic Hall and longitudinal resistances R_{xy} and R_{xx} as a function of magnetic field at T = 4.2 K. Curves a, b, c, and d were measured at 15 A/cm² (Ohmic regime), 4.5, 6.0, and 9.0 kA/cm², respectively.

higher magnetic fields ($\mu_0 H \approx 5$ T) R_{xx} eventually decreases and passes through a minimum²² before it reaches the normal-state resistance at H_{c_2} .

Figure 3 shows R_{xy} and R_{xx} plotted as a function of magnetic field but at a lower temperature T=1.4 K and at higher current densities. R_{xy} again shows the sign anomaly, and $R_{xx}(B)$ is linear at low magnetic fields with a slope that is an increasing function of the current densi-



FIG. 3. Non-Ohmic Hall and longitudinal resistances R_{xy} and R_{xx} as a function of magnetic field at T = 1.4 K. Curves a, b, c, and d were measured at 5.3, 6.7, 11, and 15 kA/cm², respectively. The dashed line in the lower graph (R_{xx} vs B) is the calculated Bardeen-Stephen flux-flow resistance.

ty. As the current density is increased, $R_{xx}(B)$ approaches the Bardeen-Stephen¹⁹ free flux-flow resistance which is shown as a dashed line in Fig. 3. (To keep the power heating the sample from exceeding 15 mW/cm² the range of magnetic field was limited to a few tesla at the highest current densities.) R_{xy} , like R_{xx} , is linear at low magnetic fields. At higher fields there is a decrease in the magnitude of R_{xy} corresponding in field to the minimum observed in R_{xx} . The sign reversal of R_{xy} is clearly observable to magnetic fields less than 1 T.

Two classic models of vortex motion are those of Bardeen-Stephen (BS)¹⁹ and Nozières-Vinen (NV).²⁰ Each model makes assumptions about the forces acting on a vortex to determine its velocity and therefore the flux-flow resistivity through the Josephson relation, Eq. (1). Both models predict the same longitudinal resistivity

$$\rho_{xx} = \rho_n H / H_{c_2} \tag{2}$$

(where ρ_n is the normal-state resistivity, H is the applied magnetic field, and H_{c_2} is the upper critical field), but neither predicts that ρ_{xy} is opposite to that in the normal state, which is the case in many superconductors. The prediction for ρ_{xx} is generally accepted but is difficult to observe because of pinning effects. Recently Kunchur, Christen, and Phillips²³ have verified Eq. (2) near T_c in YBCO at high current densities $(j \gg j_c)$ using a pulsed current technique. Our data at low temperature are at least consistent with the result for ρ_{xx} .

In several recent models, new forces acting on a vortex have been proposed to explain motion upstream to the superfluid velocity v_s . Ferrell¹⁸ has calculated a force due to the backflow of thermally excited quasiparticles which scatter off a vortex. This force acts opposite to the applied superfluid velocity, and it is expected to disappear at low temperatures. Our data show that this force cannot explain the sign reversal in a-Mo₃Si because the number of thermally excited quasiparticles is very small at the lowest temperatures in our experiments. From Fig. 3 it is clear that R_{xy} is negative at a temperature T=1.4 K and in a magnetic field $\mu_0 H=1.0$ T. At this value of field the critical temperature is suppressed to a temperature $T_c \approx 7.0$ K (see Fig. 1), and therefore a temperature of 1.4 K corresponds to $0.2T_c(H)$. Further suppression of T_c might be expected due to the large current densities used in the measurement. However, we estimate the depairing critical current density²⁴ in zero magnetic field and at zero temperature to be $\sim 10^7$ A/cm^2 , which is much greater than the current density used in our measurements. Assuming that the number of thermally excited quasiparticles has the temperature dependence $(T/T_c)^4$, their number at T = 1.4 K should be reduced from that near T_c by a factor $\sim 10^{-3}$. (By contrast, the sign anomaly only occurs close to T_c in many high- T_c superconductors. This has been used as evidence in support of Ferrell's model.)

In another model, Wang and Ting¹⁷ propose that a force due to pinning is responsible for the upstream vortex motion and therefore the sign reversal. They extend the approaches of BS and NV to the moderately clean

limit $(l/\xi_0 \sim 1)$ and argue that in this limit strong pinning can cause the sign reversal. Recently, Budhani, Liou, and Cai²⁵ have tested this theory in Tl-2:2:1:2. They observed the Hall anomaly to disappear in a Tl-2:2:1:2 film after the pinning was enhanced with heavy-ion bombardment. This is the opposite of what one would expect based on Wang and Ting's theory.

Our observation of the Hall anomaly in a-Mo₃Si where $l/\xi_0 \sim 0.01$ adds further evidence that pinning effects, as described by the theory of Wang and Ting, are not the origin of the sign change of ρ_{xy} . The role pinning plays in vortex dynamics, however, remains unclear. Pinning in a-Mo₃Si is weak compared with that of other superconductors, but the Hall angle $[\Theta_H = \tan^{-1}(E_v/E_x)]$ is also very small ($\sim 10^{-4}$) and as a result may be particularly sensitive to pinning effects. We find that the Hall angle in the non-Ohmic regime, as calculated from data in Fig. 3 does not scale with increasing current density. In Fig. 4 the tangent of the Hall angle is plotted as a function of magnetic field and becomes more negative with increasing current density. Assuming that pinning affects vortex motion less at larger current densities, our data suggest that pinning actually suppresses the anomaly.

Harris, Ong, and Yan¹⁶ attribute the anomaly in YBCO to the interaction of vortex segments moving parallel to the Cu-O planes with those moving perpendicular. They argue that vortices, whether parallel or perpendicular to the Cu-O planes, intrinsically generate a Hall electric field of the same sign as that in the normal state. Because ρ_{xz} has a sign opposite to ρ_{xy} in the normal state in YBCO, they argue that parallel and perpendicular vortex segments experience forces which tend to move them in opposite directions. When these segments are coupled, the resulting vortex line may move either upstream or downstream relative to \mathbf{v}_s . This may explain the enhancement of the negative peak in ρ_{xy} that they observe when the magnetic field is rotated to increase the population of parallel segments. For magnetic fields perpendicular to the Cu-O planes they argue that parallel vortex segments exist due to fluctuation effects. Their model may explain the sign change in the layered cuprates, but it incorrectly predicts no sign change in a-Mo₃Si which is an isotropic unlayered material.

In an earlier work, Hagen et al.¹ collected data which



FIG. 4. Tangent of the Hall angle $\tan(\Theta_H) \equiv E_y / E_x$ as a function of magnetic field at T=1.4 K. The curves a, b, c, and d were measured at 5.3, 6.7, 11, and 15 kA/cm², respectively.

existed in the literature and tentatively concluded that very clean and very dirty materials do not have a sign anomaly, while moderately clean $(0.4 \le l/\xi_0 \le 5.0)$ materials do. One exception to this observation was provided by *a*-Mo-Ge. Our work on *a*-Mo₃Si shows that a sign anomaly does occur in at least one other dirty material. We speculate that all dirty materials may have a sign anomaly. Since many of the materials in the table in Ref. 1 also have strong pinning, the anomaly was perhaps difficult to observe because the vortices are pinned except in a narrow region near H_{c_2} . Further experimental work is clearly required.

The Hall-effect sign reversal has now been seen in a wide variety of materials, with many different atomic structures and a wide range of superconducting parame-

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ters. This strongly suggests that the sign reversal is a general property of vortex dynamics as discussed in Refs. 1, 15, and 26, and is not a consequence of special properties of individual materials. Theoretical work is clearly needed to resolve the problem.

We learned recently that Graybeal, Luo, and White²⁷ were investigating the sign anomaly in *a*-Mo-Ge. Their results are consistent with the results reported above.

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