Magnetotransport in NdB_6 single crystals

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We have measured the electrical resistivity, Hall effect, and magnetization of a NdB_6 single crystal, in a temperature range from 2 to 300 K, and in magnetic fields of up to 7 T. We find that the low-field Hall resistivity varies strongly with temperature. This variation arises entirely from the large anomalous Hall effect we find in NdB_6. In the paramagnetic region, the anomalous Hall coefficient is much larger than the ordinary one and is independent of temperature. As the antiferromagnetic order sets in, below ~8 K, it decreases sharply with decreasing temperature. We do not find any significant variations of the anomalous Hall coefficient in the neighborhood of the critical point. Both magnetization and Hall resistivity show an anomaly in low magnetic fields, which may arise from domain rotations.

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I. INTRODUCTION

Electronic transport studies can reveal interesting features in magnetic materials. In particular, the Hall effect contributes an anomalous term, proportional to the magnetization of the material, in addition to the ordinary contribution which arises from the Lorentz force. This spontaneous (or anomalous) contribution can be brought about by asymmetric scattering of the current carriers which are subject to spin-orbit interactions. In addition, there is a spontaneous contribution, independent of the scattering rate, arising from a spin-dependent "anomalous velocity" that current carriers acquire in magnetic systems.[1] The anomalous Hall effect (AHE) coefficient R_s is usually much larger than the ordinary Hall effect coefficient R_o in metallic magnetic materials.[2] Moreover, its magnitude has been shown to be proportional to the third moment of the deviation of the magnetization from its mean value.[3,4] Consequently, large variations of the anomalous Hall coefficient can be expected near critical points. Indeed, they have been observed in intermetallic $R_2Fe_{14}B$ single-crystalline compounds in the vicinity of both spin reorientation and paramagnetic-ferromagnetic transitions.[5] Here, we extend our studies of the Hall effect to antiferromagnetic metallic compounds such as NdB_6.

The rare-earth hexaboride compound NdB_6 crystallizes in the CsCl-type cubic lattice. It can be viewed as two interpenetrating simple cubic lattices of B_6 octahedra and of Nd ions, set apart by the vector $\langle 1/2,1/2,1/2\rangle$. Its magnetic and electronic properties have been the subject of rather intensive research, both experimental and theoretical, for the last few decades. NdB_6 orders in an *A*-type collinear antiferromagnetic (AF) structure below $T_N \approx 8$ K.[6] The ground state of the Nd^{3+} ions ($J=9/2$) in a crystalline electric field (CEF) is the $\Gamma_8^{(2)}$ quartet.[7] The first excited energy is 133 K above the ground state. A CEF-induced weak magnetic anisotropy would align magnetic ion spins along the easy [111] direction.[8] However, the experimentally found ordering is along the [001] direction.[6] Calculations of magnetic excitations in NdB_6 show that ferroquadrupolar (FQ) interactions, whose importance has been pointed out in several previous studies,[9–11] favor fourfold easy axes.[12] Therefore, the low-field magnetic anisotropy in NdB_6 most likely arises from competition between CEF and FQ effects. The anisotropy energy is much weaker than isotropic magnetic exchange interaction. Band structure calculations indicate that $4f$ levels are rather deep in NdB_6.[13] A simple folding procedure can be used to obtain the AF bands from their paramagnetic counterparts.[14] The experimentally found frequency branches of the de Haas–van Alphen effect[15] can be well reproduced by calculated Fermi surfaces.[16]

Both experiments and calculations show that $4f$ electrons hardly affect the AF band structure. Measurements of the Hall coefficient, which has been found to be temperature independent in the 0.7 to 300 K range, seem to support this.[15] It would imply that magnetic energy gaps, induced by magnetic Brillouin zone boundaries in the AF state,[17] do not affect electronic transport. However, it was later observed that, below T_N, the Hall coefficient in a single crystal of NdB_6 increases as temperature decreases.[18] These measurements were performed in magnetic fields of 8 and 15 T, applied along the [001] direction, with the current in the [110] direction. Superzone gaps and their effect on effective carrier concentration were then invoked in order to explain the observed variation of the Hall effect. Nevertheless, it was assumed in both Refs. 15 and 18 that the anomalous Hall contribution is negligible in NdB_6. We aim to cast additional light on these issues by studying the Hall effect of NdB_6 in low magnetic fields. We thus expect to avoid smearing out anomalies near the critical points.

In this paper we report results for the electrical resistivity, Hall effect, and magnetization measurements in NdB_6 single crystals in a temperature range from 2 to 300 K and in magnetic fields of up to 7 T. We find a large, unprecedented variation of the low-field Hall resistivity which arises entirely from the anomalous Hall effect. To the best of our knowledge, such behavior has not been reported for NdB_6. The experimental procedure is described in Sec. II. Results of magnetization, resistivity, and Hall effect measurements are reported and discussed in Sec. III. Conclusions are drawn in Sec. IV.

II. EXPERIMENT

Single crystals of NdB_6 were prepared by solution growth from Al flux. We performed all measurements on one specimen with dimensions of $0.35\times1.5\times5.0$ mm^3. In this way, we expect to avoid domain- and sample-shape-related effects when comparing results of different experiments. We measured the electrical resistivity and Hall effect with a six-probe method. Contact leads (25 μm gold wire) were soldered to the sample using pure indium. In our experiments, we used a dc (100 mA) or low-frequency ac current. An external magnetic field H, between 0 and 7 T, was oriented along the [001] direction, perpendicular to the sample, while the electrical current direction was along [110], which is the longest dimension of the sample. The Hall resistivity ρ_H was measured as a function of magnetic field, from −1 up to 1 T, for all experimental points. In addition, the variation of ρ_H with magnetic field, up to 7 T, was checked at 5, 10, 100, and 300 K. The magnetization measurements in the temperature range from 2 to 300 K and in magnetic fields up to 5 T were made in the same geometry with a commercial superconducting quantum interference device magnetometer.

III. RESULTS AND DISCUSSION

We first report magnetization measurements results. Figure 1 shows how the low-temperature dc magnetic susceptibility, obtained for several external magnetic fields, depends on temperature T. The inset of Fig. 1 exhibits how the inverse susceptibility $\chi^{-1}(T)$, obtained for $H=0.1$ T, varies with T. Above about 100 K, $\chi^{-1}(T)$ obeys the Curie-Weiss law. We obtain a value of $3.15\mu_B$ for the paramagnetic effective magnetic moment of Nd. A pronounced maximum in the M/H curve at low magnetic fields corresponds to the antiferromagnetic phase transition. As the applied field in the [001] direction becomes larger, the maximum in M/H broadens and the magnetization does not drop any longer below T_N. The field dependence of the magnetization at $T=2$ and 10 K is shown in Fig. 2. A clear change in the slope of the $M(H)$ curve is observed at $H\sim0.25$ T for $T<T_N$. Such behavior may come from domain rotations. Because of the cubic anisotropy of NdB_6, the equilibrium orientation of the magnetic sublattices in the AF state is not unique. Therefore, formation of antiferromagnetic domains may occur.[19] At zero field, the magnetization within a given domain can be aligned along one of the three easy axes, which are the cubic axes. The distribution of domains among the easy axes is most likely random. When a field H is applied, domains where spins are nearly perpendicular to H grow in size, at the expense of the other domains. This process generally depends on temperature and on the magnitude of the applied field.[20] When $H\gtrsim0.6$ T, all spins are antiferromagnetically ordered in the [001] plane, slightly canted toward the z axis (field direction). We observe some hysteresis (see dashed curve in Fig. 2) in the low-temperature $M(H)$ curves which may well arise from domain-wall motions. Hall effect and magnetoresistance data also show anomalies in approximately the same magnetic field range. We discuss them below.

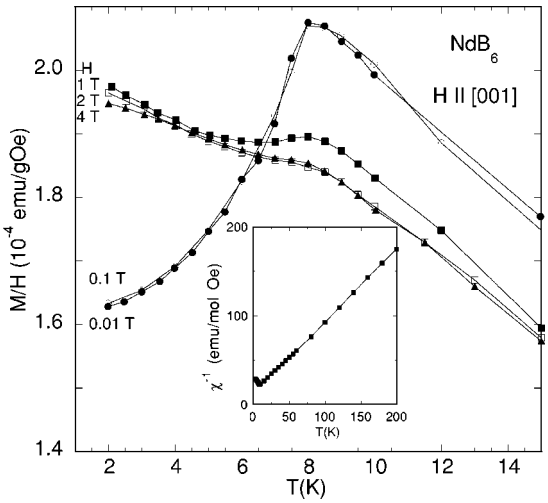

FIG. 1. Temperature dependence of the dc magnetic susceptibility at several magnetic fields. The inset shows the inverse magnetic susceptibility as a function of temperature. The solid lines are guides to the eye.

How the resistivity ρ of a NdB_6 single crystal varies with temperature in the range from 2 to 300 K is shown in Fig. 3. The resistivity drops sharply below T_N as the magnetic order sets in. CEF effects give rise to a broad shoulder, observed

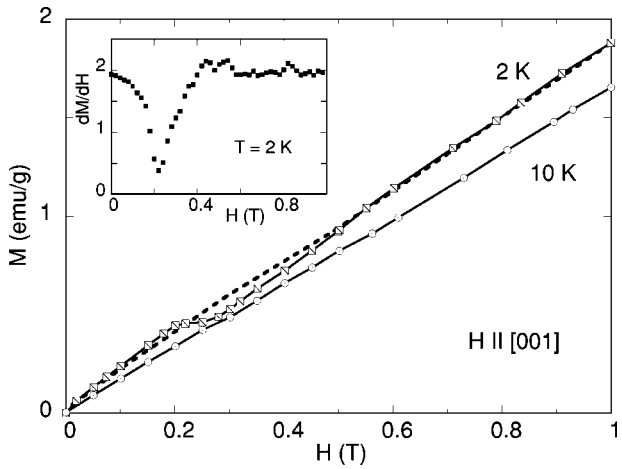

FIG. 2. Field dependence of the magnetization in NdB_6 at $T=2$ and 10 K. The solid lines are to guide the eye. The dashed line shows magnetization behavior for a field-down sweep at $T=2$ K. The inset shows dM/dH versus H at $T=2$ K.

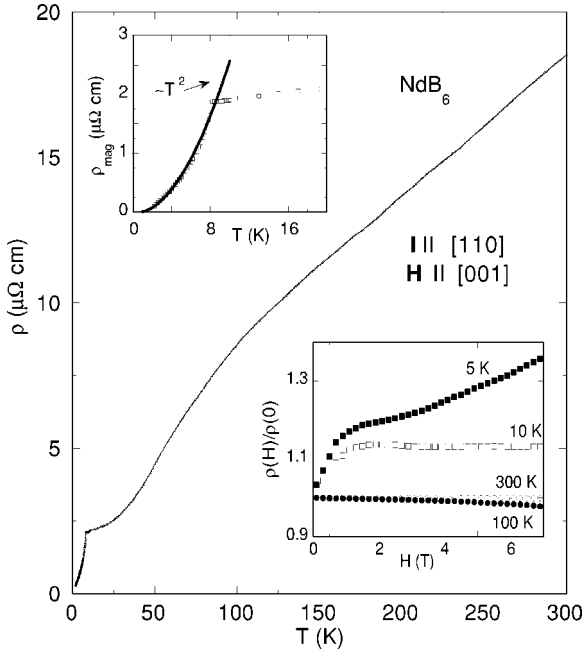

FIG. 3. Resistivity versus temperature in NdB_6 single crystals. The upper inset exhibits the magnetic contribution to the resistivity versus T in the temperature range $2 \leqslant T \leqslant 20$ K. The solid line shows a T^2 dependence. Variation of the electrical resistivity with the external magnetic field at $T=5$, 10, 100, and 300 K for the same sample is shown in the lower inset.

around 70 K.[21] For temperatures below approximately 7 K, the magnetic resistivity ρ_{mag}, obtained by subtracting from the total resistivity the lattice term [given by the lattice term of LaB_6 (Ref. 21)] and some constant residual resistivity, is proportional to T^2 ($\rho_{\mathrm{mag}}=AT^2$). This variation, shown in the upper inset of Fig. 3, may come from electron scattering by spin fluctuations. A least-squares fit yields $A=2.4\times10^{-8}\ \Omega\,\mathrm{cm\,K^{-2}}$. We find that A is strongly enhanced with respect to the usual values of electron-magnon scattering in magnetic metals.[22] Such a strong enhancement of the electrical resistivity is expected for antiferromagnetic materials when electron correlations are taken into account.[23] We note that the residual resistivity ($2.3\times10^{-7}\ \Omega\,\mathrm{cm}$) of the sample we have studied is quite small.

The magnetic field variation of the transverse resistivity in NdB_6 is shown in the lower inset of Fig. 3 for several temperatures. The resistivity decreases slightly when a magnetic field is applied at temperatures higher than 20 K. This decrease can be attributed to the suppression of spin fluctuations in the paramagnetic (PM) state by the external magnetic field. At low temperatures, the resistivity increases rapidly with magnetic field up to ~1 T and stays constant or increases much more slowly for larger values of H. The initial rise of ρ with field in the AF region may be related to the domain-wall motions. The behavior of the resistivity at higher fields can be explained by classical mechanisms. In particular, $\rho(H)/\rho(0) \propto H^2$ for $H>1.5$ T at $T=5$ K. Using known values for the effective electron mass[15] and the mobility values we have measured, we find that the cyclotron classical motion of electrons gives a good account of the measured magnetoresistance. Within this approximation,

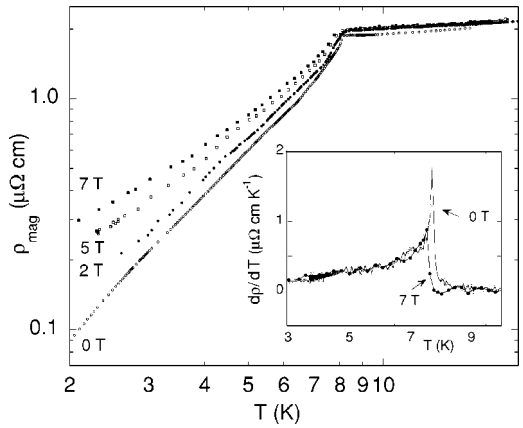

FIG. 4. Temperature dependence of the resistivity in NdB_6 for various magnetic fields. The inset shows $d\rho/dT$ versus T at $H=0$ and 7 T.

$\rho(H)/\rho(0) \approx (\mu H)^2$, where μ is the average mobility in the plane perpendicular to the applied magnetic field. We obtain values of 6×10^2 and $2.3\times10^2\ \mathrm{cm^2\,V\,s^{-1}}$ for μ at 5 and 10 K, respectively. This yields $\rho(H)/\rho(0)=3.6\times10^{-3}H$ and $5\times10^{-4}H$ (H is in units of tesla) at $T=5$ and 10 K, respectively, in excellent agreement with experimental data.

Figure 4 exhibits the low-temperature behavior of $\rho_{\mathrm{mag}}(T)$ for several magnetic fields. The T^2 variation found for the magnetic resistivity at zero magnetic field is not followed at fields larger than 2 T. The inset shows the temperature derivative of the resistivity in the critical region, determined numerically for $H=0$ and 7 T. $d\rho/dT$ falls off nearly vertically above the transition, far below the specific heat of NdB_6.[18] $d\rho/dT$ resembles the specific heat behavior just below T_N. In terms of the Fisher and Langer theory,[24] this points to negligible short-range order above T_N. The critical temperature decreases slightly with applied magnetic field, from 7.7 K at $H=0$ T to 7.55 K at $H=7$ T.

We now turn to the Hall effect results. Hall resistivity ρ_H data, for a field applied along the [001] direction, are plotted as a function of temperature in Fig. 5. At low fields, ρ_H increases as T decreases down to T_N; it drops sharply below the critical temperature. The inset in Fig. 5 exhibits how the Hall voltage and the magnetization vary with magnetic field at $T=3$ K. The Hall resistivity follows quite closely the magnetization of the sample. The Hall voltage and the magnetization show similar anomalies at low fields. As discussed above, this anomaly may be produced by domain rotations. It is clear that the measured ρ_H is very sensitive to the magnetic state of the sample.

To interpret these results, we use for the Hall resistivity the phenomenological expression $\rho_H=R_oB+R_s4\pi M_s$, where R_o is the normal Hall coefficient, R_s is the AHE coefficient, B is the applied magnetic induction, and M_s is the spontaneous magnetization. For temperatures above T_N, there is no spontaneous moment contribution to ρ_H, but the paramagnetic moment induced by the applied field is important. In the paramagnetic region $M_s=\chi H$, where χ is the magnetic susceptibility. Using this relation we obtain $\rho_H/H=R_o+4\pi\chi^*[R_s+R_o(1-N)]$, where $\chi^*=\chi/(1+4\pi N\chi)$ is the effective susceptibility which includes the effects of the demag-

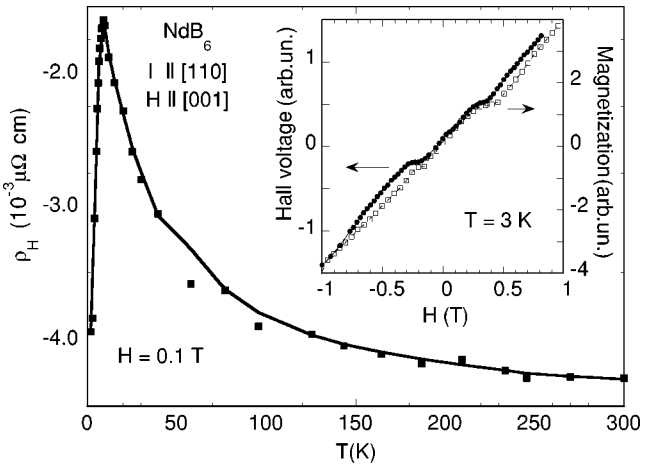

FIG. 5. Hall resistivity at $H=0.1$ T as a function of temperature for NdB_6 single crystal. The solid line is a guide to the eye. The insets show the magnetic field variation of Hall voltage and M at $T=3$ K.

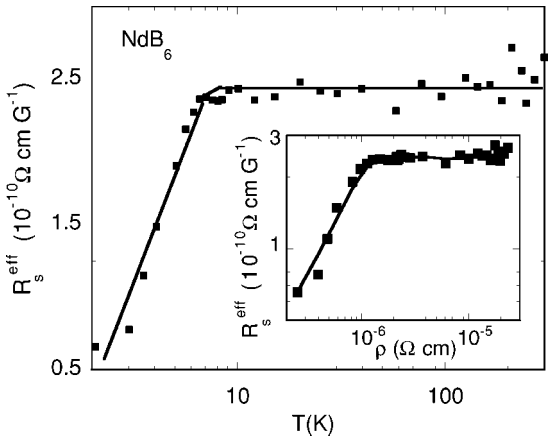

FIG. 7. Anomalous Hall coefficient R_s as a function of temperature in the NdB_6 single crystal. The solid line is a guide to the eye. The inset exhibits R_s as a function of the total resistivity for the same crystal.

netization field (N is the demagnetization factor). The total Hall resistivity is linear in H; however, it has a normal part and a part that depends on the magnetic susceptibility. The magnetic field variation of ρ_H in the PM state is exhibited in the inset of Fig. 6 for several temperatures. Indeed, we find a linear $\rho_H(H)$ dependence. Plotting ρ_H/H versus χ^* we obtain the linear behavior shown in Fig. 6. This implies that both Hall coefficients are independent of T. The ordinate intercept yields $R_o=-4.59\times10^{-12}$ Ω cm/Oe while the slope of the straight line gives $R_s=2.4\times10^{-10}$ Ω cm/G. The value of the ordinary Hall coefficient agrees very well with those reported previously.[15,18] However, no temperature variation of the Hall effect above T_N has been observed in Refs. 15 and 18. Here, we find experimentally a strong temperature dependence of the Hall resistivity in the paramagnetic state which comes entirely from the magnetic susceptibility. A large anomalous contribution to the Hall effect in the PM phase has also been observed in the parent compound EuB_6.[25]

In order to estimate R_s below T_N, we assume that the ordinary Hall coefficient does not vary with temperature.

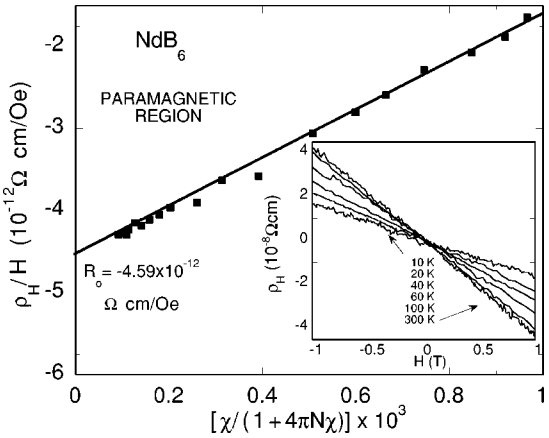

FIG. 6. Slope of ρ_H versus H in the paramagnetic region plotted against the effective susceptibility for the NdB_6 single crystal. The solid line is the linear fit to experimental points. The normal Hall effect coefficient R_o has the constant value indicated.

This is contrary to what has been argued in Ref. 18. A rise of the Hall resistivity below T_N for fields of 8 and 15 T, reported in this reference, was interpreted in terms of the ordinary Hall effect and ascribed to a modification of the Fermi surface induced by an antiferromagnetic modulation. Our data do not support such a picture. In the first place, we observe a large AHE in the PM region. Therefore, we expect an important contribution from the AHE to the Hall resistivity in the ordered phase. In addition, the Fermi surface reconstruction upon AF ordering in NdB_6 is not a drastic one. In the AF phase, the cyclotron mass of the nearly spherical Fermi surface is only slightly heavier than the one observed in the PM region.[14,15] Therefore, the density of states and, consequently, the effective carrier concentration is not expected to be significantly affected by magnetic ordering. In addition, superzone gaps, if their effect were important, would affect the resistivity as well. However, this is not observed. Consequently, we put $R_o=$const in the expression for ρ_H. To obtain R_s, we use values of the longitudinal magnetization, measured below T_N, for M_s. M_s in the equation for ρ_H is the spontaneous magnetic moment. However, what M_s stands for in AF materials is not so obvious. With our choice, we calculate some "effective" AHE coefficient from the relation $R_s^{\text{eff}}=(\rho_H-R_oH)/4\pi M$.

How the effective AHE coefficient varies with temperature in the whole temperature range we have studied is shown in Fig. 7. Up to T_N, R_s^{eff} rises quite sharply with increasing temperature and becomes independent of T beyond T_N. R_s^{eff} does not show any large variations through the magnetic phase transition, contrary to what is observed in uniaxial ferromagnets.[5] Such variations may come from critical fluctuations of the magnetization. However, we do not expect large magnetization fluctuations along the direction of the applied magnetic field ([001]) in NdB_6, since magnetic moments in the AF phase are perpendicular to H for small magnetic fields. On the other hand, critical scattering at T_N, clearly evident at zero field, can be completely washed out by high magnetic fields.[20] Therefore, we also expect no important effects in large magnetic fields when Nd spins are along the field direction.

The variation of R_s^{eff} with the total resistivity for NdB_6 is exhibited in the inset of Fig. 7. The AHE coefficient increases fairly linearly with ρ in the AF region, in agreement with predictions for skew scattering.[26] In this mechanism, spin-polarized electrons are displaced preferentially to one side of the scattering center because of spin-orbit coupling, and the AHE resistivity is proportional to the longitudinal resistivity. However, since our data points cover only a small temperature interval below T_N, we cannot rule out $R_s^{\text{eff}} \propto \rho^2$. In the paramagnetic region, R_s^{eff} is independent of ρ, in accordance with predictions for scattering of *s* electrons by localized magnetic moments.[3]

Very large Hall constants with anomalous positive signs are often found in mixed-valence and Kondo-lattice compounds, particularly in heavy-fermion systems. A model invoking skew scattering and anomalous velocity contributions to the Hall effect gives $\rho_H(T)/H = R_o + \gamma\chi'(T)\rho_{\text{mag}}(T)$ for heavy-fermion systems, where $\chi'(T) = \chi(T)/C$ is the reduced susceptibility and C is the Curie constant.[27,28] The prefactor γ is related to the phase shifts of the conduction electrons in scattering processes. Our data for $\rho_H(T)$ at high temperatures ($T \gtrsim 100$ K) follow this relation quite well. Fortuitously, the value of R_o, obtained by plotting ρ_H versus $\chi\rho_{\text{mag}}$ and extrapolating to $\chi\rho_{\text{mag}} = 0$, is -4.9×10^{-12} Ω cm/Oe, which is very close to the value we estimate above. Therefore, high-temperature results for the Hall resistivity in NdB_6, which is not a heavy-fermion material, agree with predictions of calculations for incoherent skew scattering in heavy-fermion compounds. This may imply an important role for $4f$ levels in scattering processes in NdB_6.

IV. CONCLUDING REMARKS

Our measurements of the magnetization, resistivity, and Hall effect in a NdB_6 single crystal reveal some interesting features. Below T_N, both magnetization and Hall resistivity show anomalies at low magnetic fields. They may arise from magnetic domain-wall motion. This follows from the cubic anisotropy of NdB_6 which permits equilibrium orientations of the staggered magnetization along the three crystalline axes. When a weak magnetic field H is applied, domains with spins perpendicular to H grow in size. This phenomenon may lead as well to the large initial increase of resistivity we observe in magnetic fields.

The temperature variation of the electrical resistivity at low temperatures is unusually high in NdB_6. We attribute this to spin fluctuations of correlated electrons. We find that the low-field Hall resistivity varies strongly with temperature. Such a variation has not been previously reported for NdB_6. Taking into account that (i) in the paramagnetic phase, the Hall effect depends on temperature only through the magnetic susceptibility, and (ii) Fermi surface reconstruction cannot account for the Hall effect changes in the antiferromagnetic phase, we conclude that the observed behavior arises from the large anomalous contribution to the Hall effect. Below T_N, skew scattering is likely responsible for the observed temperature variation of the Hall effect, as expected for pure magnetic materials. The anomalous Hall coefficient rises quite sharply as the critical temperature is approached, and becomes independent of temperature above T_N, in agreement with *s-f* interaction-based models. We do not find any large variation of the Hall effect through the magnetic phase transition.

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