Low Field Hall Effect in $FeCl_3$ Graphite Intercalation Compunds

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Abstract

We have measured the Hall effect in stages 2, 3, 4, 5, 8, and 9 $FeCl_3$ graphite intercalated compounds in fields up to 5 KG and at temperatures 10K, 77K, and 300K. The variations of the Hall voltages with temperatures are relatively small for the intermediate stages. The Hall voltages for all stages are positive.

Introduction

Eventhough extensive investigations have been carried out on various graphite intercalation compounds (GIC), the nature of the microscopic mechanisms in these compounds is ambiguous. To understand the drastic change of transport and electronic properties upon the intercalation of graphite [1], more data and theoretical investigation are needed. Specifically, the charge transfer, electronic bonding, and relaxation effects need to be clarified for these GIC. The low temperature magnetic anomaly which has been observed in $FeCl_3$-GIC [2,3] is still not fully understood and one has to provide an adequate explanation which can fit most of the data in this system.

As a continuation of our comprehensive studies of $FeCl_3$-GIC, we report in this work the results of the low field Hall effect measurement at different temperatures for various stages of this copound. The long term stability and uncomplicated process of preparation make $FeCl_3$ an ideal model of GIC system. In low field measurements, one can extract information about the transport coefficients of these GIC. The high field Hall effect and magnetoresistivity data [4] have exhibited Shubnikov-de Haas quantum oscillations (SdH) which were relatively large in the intermediate stages. In this work the Hall effect is measured in magnetic fields up to 5 KG, and at temperatures 10 K, 77 K, and 300 K for stages 2, 3, 4, 5, 8, and 9 $FeCl_3$-GIC.

Experimental

The $FeCl_3$-GIC samples were prepared using a standard two-zone furnace technique [1] where stage index was controlled by the temperature difference between the graphite host (HOPG) and the $FeCl_3$ powder. The samples were in the form of thin rectangular plates of dimensions 1.5x0.5x.1 cm^3. The samples measured were charcterized by means of x-ray diffraction and Mössbauer effect [5,6]. The x-ray diffractograms were also used to determine the c-axis repeat distance of each sample. Layers of about 1.5x0.5x0.001 cm^3 were peeled from the samples and then mounted on flat substrates. The five-probe dc technique was used to measure the Hall voltage and also the magnetoresistance, which will be published elsewhere.

Results and Analysis

Based on our previous measurements [2,4] and the measurements reported here, we have observed that there is a characteristic low stage and high stage behavior in the physical properties of $FeCl_3$-GIC and the distinction falls between stage 4 and 5. Therefore in the first part of this work the Hall data of one of the lower stages and one of the higher stages, as well as stage 5 will be discussed at three different temperatures. The data shown in Fig(1) are the Hall voltage versus the applied magnetic field for stage 3 at 10 K, 77 K, and room temperature. The magnetic field throughout this work was always parallel to the c axis. As shown in the figure, the Hall voltage is postive, indicating the dominance of hole carriers, and nearly linear with field at room temperature. As the temperature is decreased, it appears that there are some long period quantum oscillations. At this point, these oscillations have not been carefully examined and the details about the analysis of the different frequencies of these oscillations and the extracted physical quantities will be pesented elsewhere. Quantities plotted in Fig.(1) are repeated for stage 9, as shown in Fig (2). The data of stage 9, however do not clearly exhibit the low temperature oscillations shown for stage 3. The lines on all graphs of this work are a guide to the eye.

The Hall voltage of the lower and higher stages is relatively sensitive to temperature indicating that the density of states in these stages is temperature dependent. The Hall voltage of stage 5, shown in the plot of Fig. (3), has the least temperature dependence among all the stages. The Hall data at low field for stage 5 are consistent with our high field measurements [4]. These results indicate that in stage 5 the carriers are confined to a two dimensional plane and undergo a minimum scattering relative to all other stages. In higher stages, where one would expect an increase of the two dimensional nature of the system, as the number of graphite layers increased new conduction band channels open across the graphite layers [7] and an out-of-plane scattering arises.

In figures 4, 5, and 6, the Hall voltage is plotted versus the applied magnetic fields for various stages of $FeCl_3$-GIC at 10 K, 77 K, and room temperature, respectively. As shown in the figures, one common result can be observed on the three graphs that above 4 KG stage 5 has the largest Hall signal among all the stages. At room temperature the Hall voltage seems to be linear and increases with the stage index up to stage 5 then decrease again for the higher stages. As the temperature decreases, one can observe oscillations in the intermediate stages which become more dominant in stage 5. In this low field limit and at the three different temperatures the carriers of all the stages are holes, this is in contrast to the high field limit (above 10 KG) where the Hall Coefficients of the higher stages change sign from negative in low field to postive values at the high field limit [4].

If the simple one band model is used to calculate the carrier densities at the three different temperatures, the results will not be consistent with the previously reported data. In this low field both kinds of carriers contribute to the conduction and one can not approximate the standard two band model with a simple one carrier equation. Stage 5 is expected to exhibit the largest carrier density, and this is not observed in this case. This leads us to conclude that in the low field limit both electrons and holes contribute to the conduction, however the holes are the dominant carriers.

In conclusion, the Hall Coefficient is postive for all the stages which have been investigated in this work and stage 5 exhibit the largest Hall signal at all temperature. The Hall voltage is not senstive to the temperature for intermediate stages indicating constant density of carriers. A simple band model can not be used to determine the carrier density in this low field limit.

References

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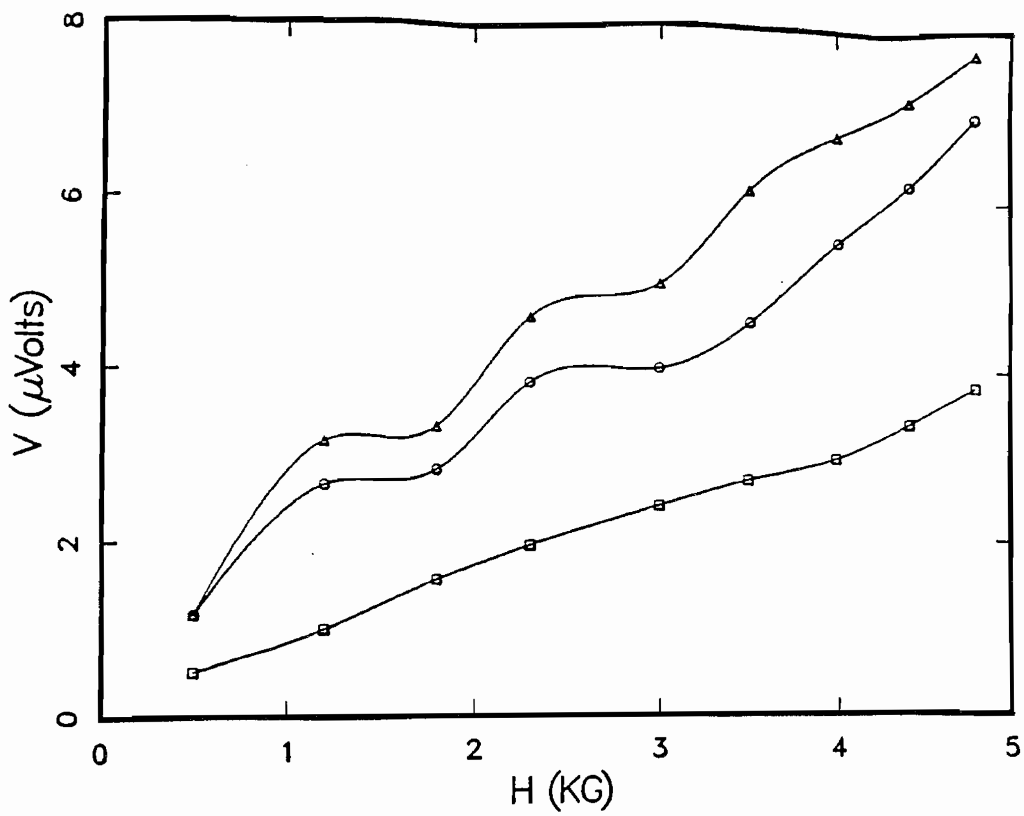

Fig. 1. Hall voltage vs. magnetic field for stage 3. The squares are room temperatue, the circles are 77 K, and the triangles are 10 K.

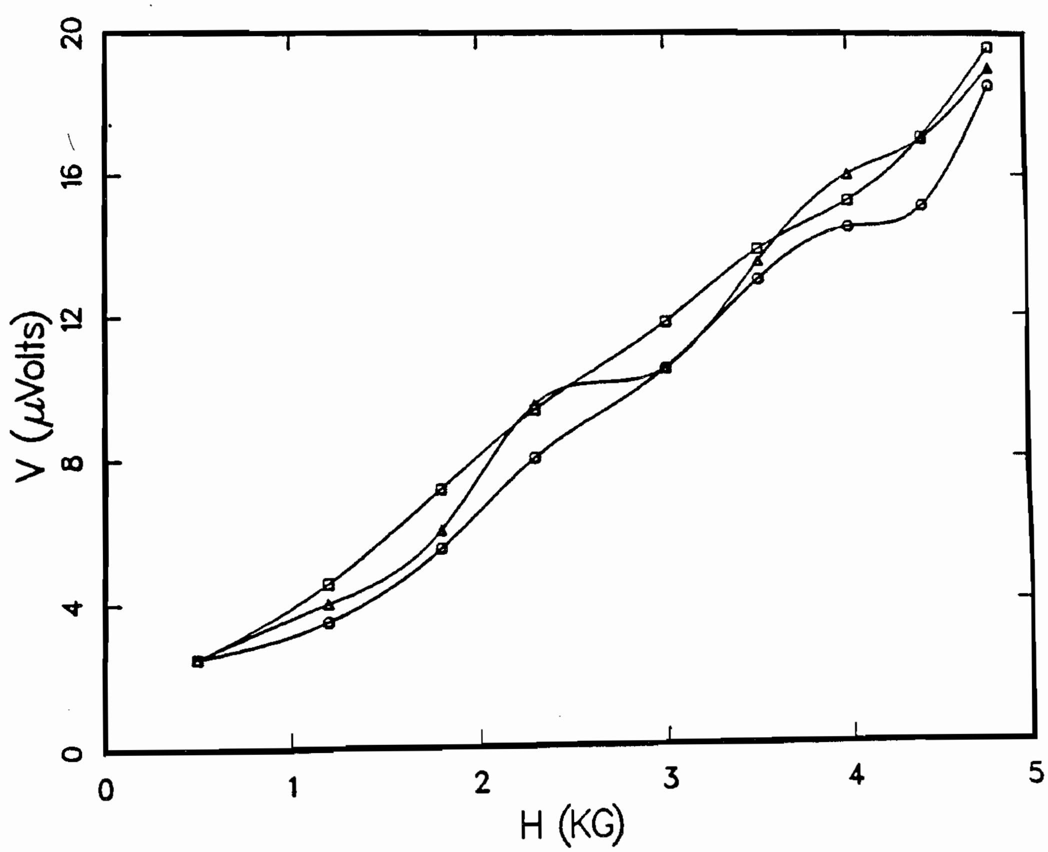

Fig. 2. Hall voltage vs. magnetic field for stage 5. The squares are room temperatue, the circles are 77 K, and the triangles are 10 K.

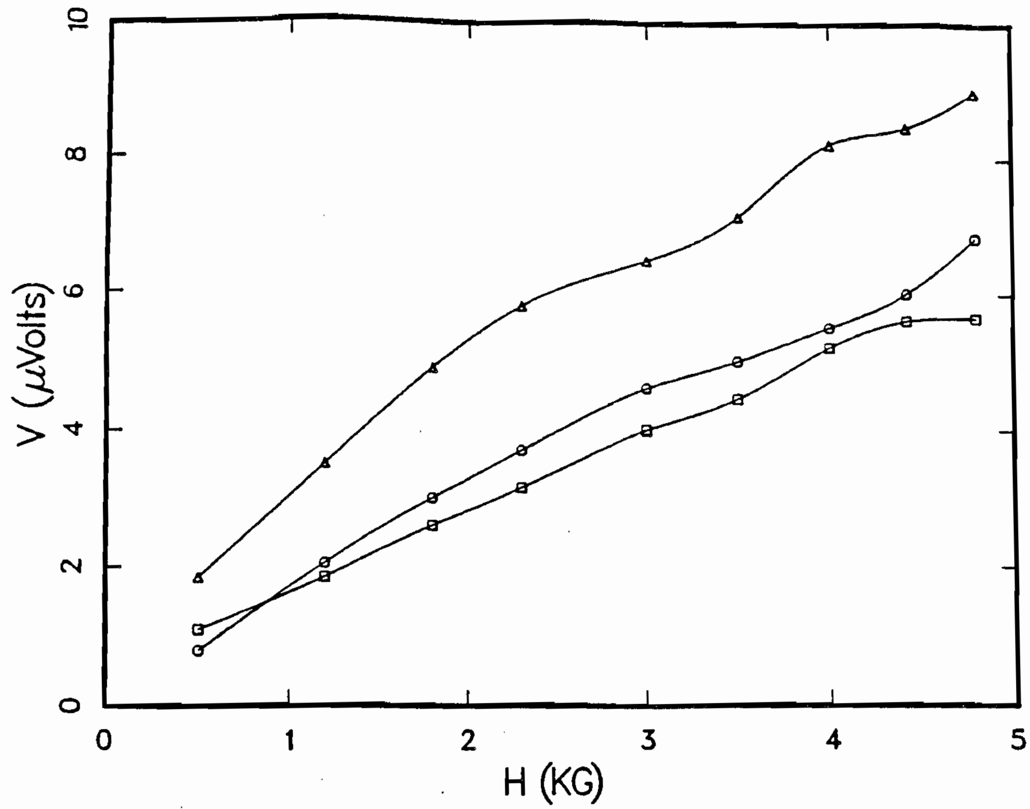

Fig. 3. Hall voltage vs. magnetic field for stage 9. The squares are room temperatue, the circles are 77 K, and the triangles are 10 K.

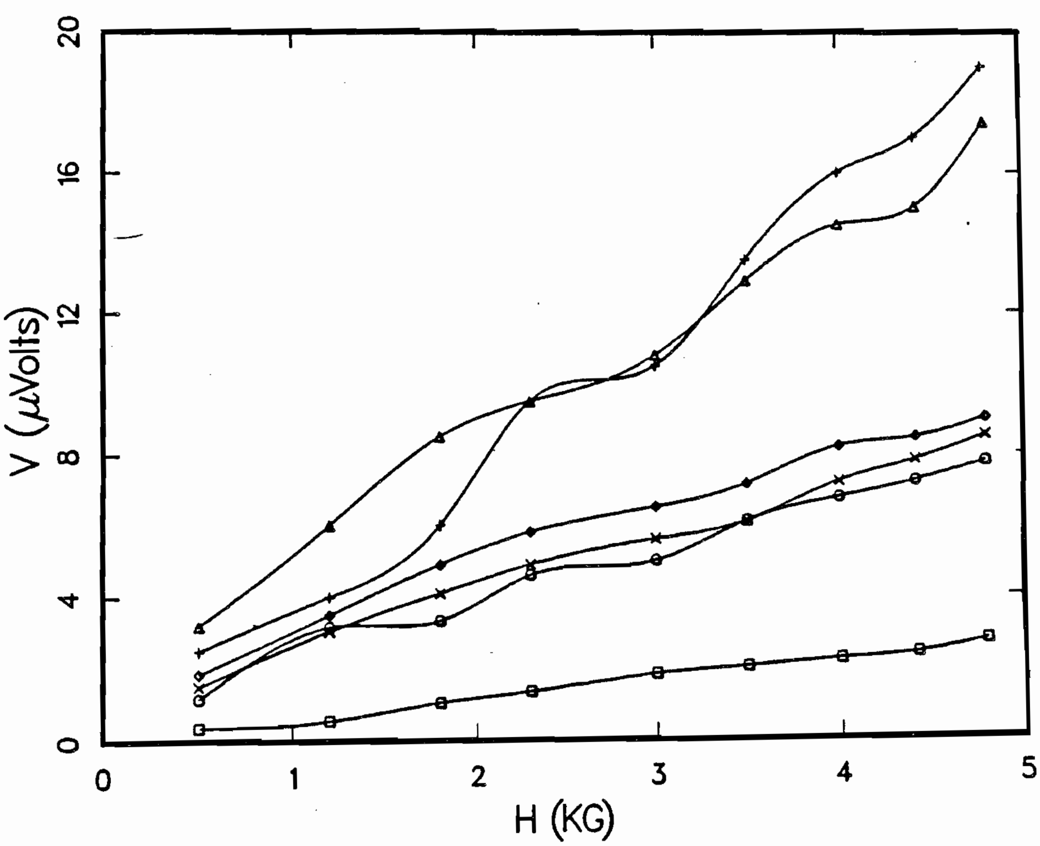

Fig. 4. Hall voltage vs. magnetic field at room temperature. Stage 2, 3, 4, 5, 8, and 9, are designated by squares, circles, triangles, crosses, x, and diamonds, respectively.

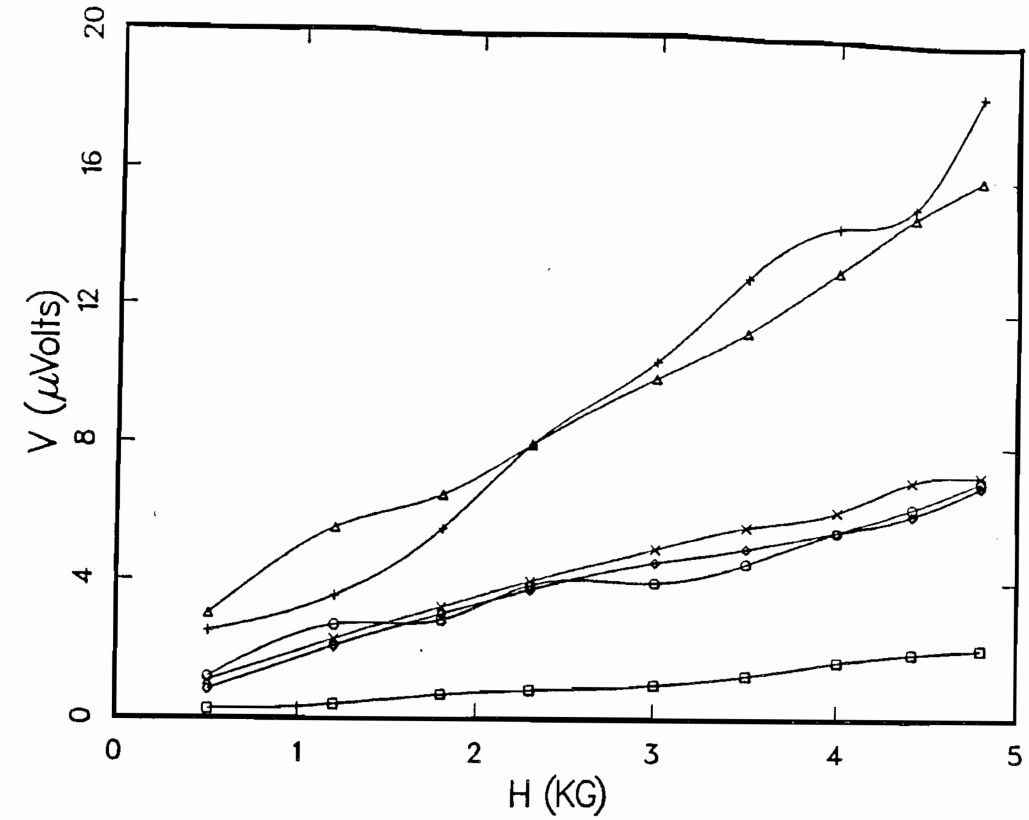

Fig. 5. Hall voltage vs. magnetic field at 77 K. Stage 2, 3, 4, 5, 8, and 9, are designated by squares, circles, triangles, crosses, x, and diamonds, respectively.

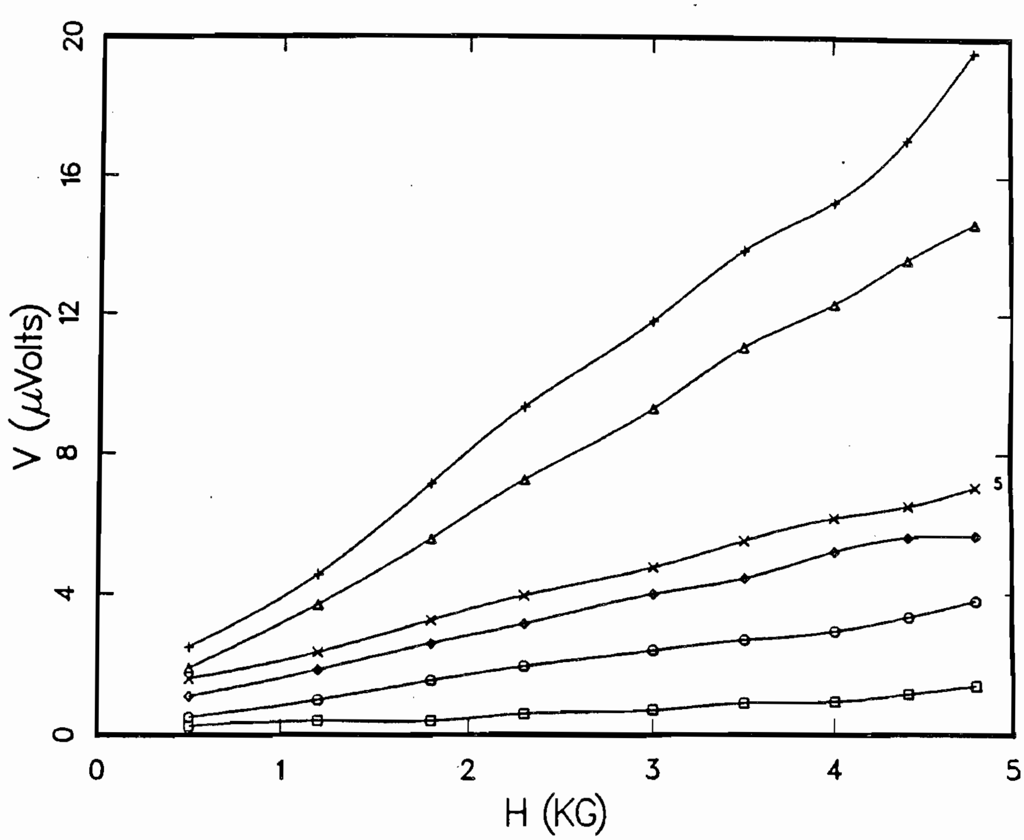

Fig. 6. Hall voltage vs. magnetic field at 10K. Stage 2, 3, 4, 5, 8, and 9, are designated by squares, circles, triangles, crosses, x, and diamonds, respectively.