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TWO-DIMENSIONAL MAGNETISM IN LANGMUIR-BLODGETT FILMS OF MANGANESE STEARATE MEASURED IN A He₃-SQUID MAGNETOMETER

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Abstract. — The magnetisation of manganese stearate layers on aluminium foil has been measured between 20 K and 0.4 K. With the applied magnetic field parallel to the film we deduced an equivalent susceptibility of $6.4 \times 10^{-3}$ at 4.2 K. This is close to our paramagnetic calculation based on the measured manganese spacing. Our results would be consistent with a transition to antiferromagnetism below 0.4 K.

Introduction

The Langmuir-Blodgett technique [1] provides a means of creating two-dimensional arrays of magnetic ions which can then be used in the characterization of low dimensional magnetism. Of particular interest is the experimental observation of magnetic ordering in two dimensions. Pomerantz [2] has reported indications of magnetic ordering near 2 K from ESR measurements on Langmuir Blodgett films of manganese stearate (octadecanoate). His samples varied in thickness from one to several hundred layers. The only other report has been a brief reference [3] to a susceptibility maximum in a monolayer sample near 0.3 K. In the research work reported here we proposed to detect any phase transition directly using a SQUID magnetometer.

Film preparation

The Langmuir Blodgett (LB) process can be used to produce samples of well defined dimensions where the planes of magnetic ions are well separated from each other, thus forming a good approximation to a two-dimensional magnet. The successful preparation of such films requires pure materials and a very clean environment. The films are produced by moving a substrate through a monomolecular layer spread on a liquid surface while maintaining a fixed surface pressure. On each up or down stroke a new layer is added. The monolayer material is usually a long chain hydrocarbon with a hydrophilic head group which in our case was stearic acid doped with Mn²⁺ ions.

For substrates we used 350 mm by 150 mm sheets of aluminium foil on which LB films are known to deposit well. Using foil allowed a large surface area with a small substrate volume thereby minimising the signal from the substrate. A 20 mm wide strip of LB film was deposited along the length of one side of the foil which was then rolled into a tube. This provided strength and gave the sample an axial symmetry, corresponding to that of the magnet and pickup coil, so that the sample would be magnetised parallel to the LB layers.

The magnetometer

The samples were measured with a SQUID magnetometer in a top loading He₃ cryostat. The foil was suspended at one end from a moveable cold platform, which was connected to the He₃ pot via flexible heat links of copper braid. The temperature was monitored with a calibrated germanium thermometer mounted on the He₃ pot.

Measurements of magnetization were made by moving the sample in and out of a second order gradiometer coil maintained at 4.2 K. The length of the substrate was chosen to be several times the length of the pickup coil so that the background signal remained essentially constant. The stability of field provided by the high homogeneity superconducting magnet was augmented by trapping the flux inside a cylindrical superconducting niobium screen surrounding the sample and pickup coil.

The instrument function for the magnetometer was simulated on a microcomputer, including the effect of the superconducting screen [4], and checked against the flux linked with a test solenoid as sample. Another calibration was obtained using a "semi-infinite" rod of pure aluminium of volume susceptibility $2.4 \times 10^{-8}$.

Results

Four LB films were measured with thicknesses of 1, 2, 11 and 101 layers of MnSt₂. The results from the first two films differed from the predicted shape and also exhibited an axial shift of the peak position. The observed signal profiles could not be reconciled with the simulations or the other results, and furthermore the signals were not temperature dependent. It
Fig. 1. — Variation of flux linked with the second order gradiometer pickup coil from a sample of 101 layers of manganese stearate. The \textit{z} axis lies parallel to the plane of the film which is in the form of a coaxial cylindrical spiral.

Fig. 2. — Inverse susceptibility per layer plotted against temperature for manganese stearate films. The symbols are for 11 layers (●) 2 mT, (▼) 4 mT, for 101 layers (○) 2 mT, (▲) 4 mT, (■) 6 mT. The solid line is a least squares fit to all the data including measurements at 20 K (not shown).

was concluded that the manganese stearate signal had been swamped by some contamination and these results were therefore discarded.

The 101 layer film was measured at temperatures of 20, 4.2, 1.2 and 0.4 K and the results are shown in figure 1. The peak to peak difference in these curves is directly proportional to the magnetization of the sample. The data for two film thicknesses (11 and 101 layers) and for several values of applied field (2 mT, 4 mT, 6 mT) are plotted in figure 2 as the inverse of susceptibility against temperature. The solid line shows the least squares fit to all the data in the range 0.4 K to 20 K. The data for each field setting show consistent deviations from the average which may be attributable to systematic errors (up to 0.5 mT) in the field calibration: the random uncertainties in the field setting were estimated to be ± 0.1 mT. Nevertheless the individual fits for each field value all show negative temperature intercepts which fall in the range 0.6 ± 0.4 K. Recordings of the peak signal response during a steady decrease in the sample temperature were smooth with no indication of inflections in the temperature range from 20 K to 0.4 K.

Discussion

Our results indicate no evidence of a transition from the paramagnetic phase at least down to 0.4 K. This differs from the previous report by Pomerantz [2] of a transition to weak ferromagnetism at 2 K. However the temperature intercept of − 0.6 ± 0.4 K, shown in figure 2, would be consistent with a transition to antiferromagnetism below 0.4 K, or perhaps to weak ferromagnetism.

There are two possible factors which may account for the differences between our results and those of Pomerantz. Firstly most of the results reported by Pomerantz were obtained with the applied field in the direction of the film normal whereas in our case the field lay parallel to the film, which would yield a different if these “two-dimensional” films were magnetically anisotropic. Secondly when Pomerantz changed the preparation route for his powder samples he observed a weak ferromagnetic to antiferromagnetic transition at a higher temperature [5].