Quantized Hall Effect in Ultrathin Metallic Films

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The Hall coefficient R_H of ultrathin epitaxial Pb films is determined experimentally. A comparison with electrical conductivity data leads to the conclusion that the investigated Pb layers behave like a size-quantized metal. Pronounced variations of R_H with the film thickness were found. The observed reversal of the sign of R_H is discussed within the available theory of the quantum size effect describing the galvanomagnetic properties of metals. We find that the observed phenomenon cannot be explained by the free electron model of a quantized layer. [S0031-9007(96)00249-9]

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There is growing interest in quantum size effects (QSE) in ultrathin metallic films [1]. Recent studies have clearly shown the occurrence of the QSE in the electrical conductivity of ultrathin Pb films [2] but, to our knowledge, the influence of the size quantization on the Hall effect in ultrathin metallic film has never been investigated experimentally before. From the theoretical point of view the Hall effect in quantized metallic systems was discussed only by Calecki [3]. He studied the galvanomagnetic properties of thin quantized metallic films for the case in which electrons are scattered elastically by surface roughness and volume impurities. The conclusions that could be derived from this work are as follows. In the presence of bulk impurities only, represented by δ-function scattering potentials, the Hall constant is equal to $-1/ne$. Scattering by the surface roughness causes a variation of the Hall coefficient with the film thickness d. When the correlation length of the surface roughness is sufficiently small compared to the Fermi wavelength λ_F, then $R_H = -1/ne$ when only one subband is occupied by electrons ($\nu_F = 1$). When the number of occupied subbands is large ($\nu_F \gg 1$), $R_H = (-1/ne)\,(4/15)\nu_F \sim d$. Calecki [3] also states that in the low-correlation-length limit the Hall constant does not depend on the parameters describing the surface roughness and does not give directly the value of electron density.

In this Letter we present results of the experimental determination of the Hall coefficient in a weak magnetic field as a function of film thickness and compare the experimental data with predictions of existing theory of the Hall effect in quantized ultrathin metallic films. Ultrathin Pb films provide a promising starting point for obtaining a general understanding of the influence of the QSE on the Hall effect because we have previously observed pronounced QSE effects in such films [2]. In the present study we combine the UHV technology used previously to study the QSE in the specific conductivity [2] with a new experimental method used specifically to detect the weak signals produced by the Hall effect.

The sample preparation and the measurements were performed in an ultrahigh vacuum system with a base pressure $<7 \times 10^{-11}$ mbar. It was equipped with a He cryostat and a reflection high-energy electron diffraction (RHEED) system. Semi-insulating Si(111) oriented to within 0.05° was used as the substrate. The specific resistivity of Si at room temperature was about 7000 Ω cm. The size of the Si samples was $8 \times 5 \times 0.6$ mm^3. They were chemically cleaned by standard procedures and mounted in the cryostat. Two cooled Mo clamps were used as current contacts. The three potential contacts for resistivity and Hall voltage measurements were electrolytically sharpened tungsten wires pressed against the substrate. The final cleaning was performed by flashing for a few seconds to about 1500 K by direct resistive heating. This resulted in the appearance of a sharp (7×7) superstructure RHEED pattern with no visible traces of contamination. Before deposition of Pb the (7×7) surface was converted into a Si(111)-$(\sqrt{3} \times \sqrt{3})R30°$Ag superstructure. This was achieved by deposition of 1.1 ML (monolayer) of Ag [1 ML= atomic density of a Si(111) bilayer, e.g., 7.83×10^{14} atoms/cm^2] at about 80 K followed by annealing for about 1 min at about 700 K [4]. During Pb deposition the substrate was also at 80 K.

The crystal structure of the growing Pb film was monitored with RHEED. The film was crystalline from the very beginning. Already at about 1.5 ML of Pb faint streaks with Pb lattice periodicity were visible. The RHEED pattern from 3 ML of Pb showed a superposition of two (111) orientations. The orientation with Pb$[1\bar{1}0]\|$Si$[1\bar{1}0]$ strongly dominated the second orientation, which was rotated 30°. At a thickness of 10 ML the streaks from the weaker orientation were not visible any longer. The thickness of the ultrathin layer was monitored by a quartz crystal oscillator which in turn was calibrated using the RHEED specular beam intensity oscillation technique. Thus the mean thickness of the Pb films could be determined with an accuracy better than $\pm 3\%$ of 1 ML Pb$_{(111)}$.

The resistivity of the growing film was measured during the deposition process. The method of measurement here was the same as that used previously [5]. The characteristic oscillations of the resistivity with increasing film thickness [2] are the main criterion for the occurrence of size quantization. After the desired thickness was reduced, the temperature of the sample was decreased to 20 K and the cryostat was placed in a coil which was driven by a frequency generator operating at a frequency of $f_1 = 4.2$ Hz. This coil produced an effective magnetic induction of 0.05 T at the sample. The current through the sample was supplied by another generator which was operated at a frequency $f_2 = 7.3$ Hz. The resulting product signal with the frequency $f_1 + f_2$ was then detected by a lock-in technique and stored for further evaluation. The current passing through the sample was about 200 μA for the sample with 3 ML of Pb and about 2 mA for the sample with 10 ML of Pb. The typical Hall effect signals of these films were in the range of 200 nV. Special care had to be taken to avoid spurious signals generated by ac magnetic fields and ac currents. The resistivity of the bare substrate [with $(\sqrt{3} \times \sqrt{3})R30°$Ag superstructure] at 20 K was more than 200 GΩ. The system was first calibrated with a commercial Hall probe and with a thick layer of Ag deposited on the same substrate. After completion of measurement, the sample was flashed and a new Ag superstructure was prepared. Then a new Pb film with another thickness was grown.

Figure 1(a) shows the Hall coefficient calculated from the data obtained from a set of ultrathin Pb layers prepared at 80 K and measured at 20 K. Each circle is from a separate film. Below 2.5 ML the noise rapidly increased with decreasing thickness due to the large resistivity of films. Figure 1(b) shows the thickness dependence of the specific conductivity for a sample prepared in the same conditions at 80 K and measured continuously during deposition.

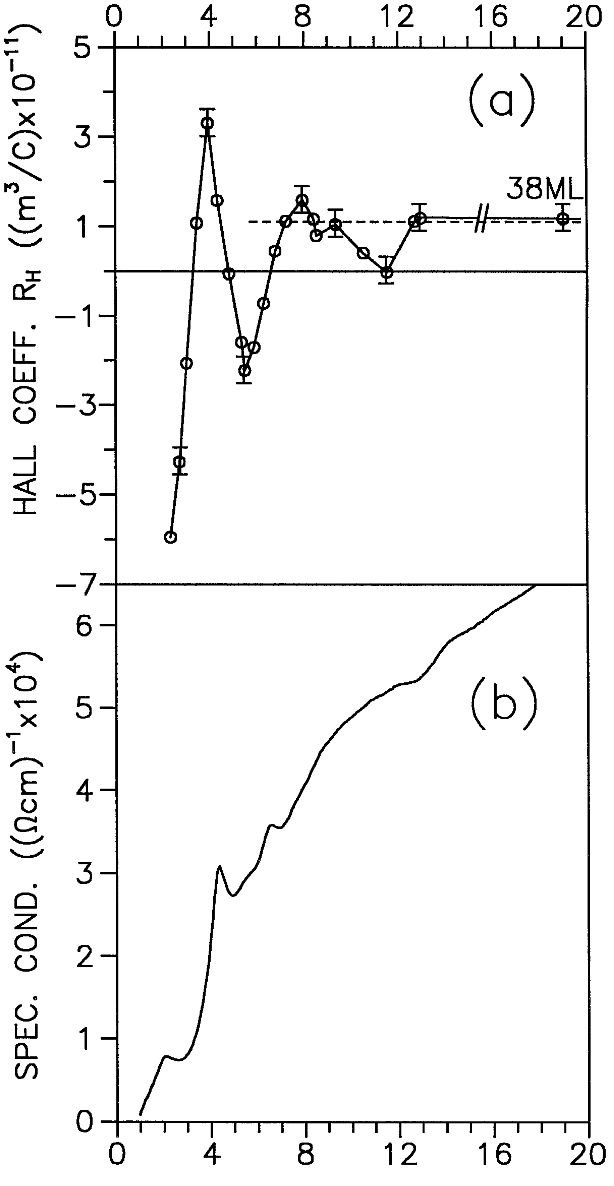

FIG. 1. Hall coefficient of ultrathin Pb(111) films deposited at 80 K on Si(111)-$(\sqrt{3} \times \sqrt{3})R30°$Ag, measured at 20 K (a). The dashed line is data for bulk Pb from [6] measured at 20 K. (b) shows the thickness dependence of the specific conductivity, measured during deposition in the same condition as in (a).

Surprisingly, the Hall coefficient possesses features which are not predicted by the available published theory. The most interesting phenomenon is the reversal of the sign of the Hall coefficient in the vicinity of 3.5, 5, and 7 ML of Pb. It should be noted that the Hall coefficient of bulk Pb is positive and equal to $+1.1 \times 10^{-11}$ m^3/C [6]. The data of Fig. 1(b) provide strong support for size quantization of the specific resistivity. Similar to our previous studies [2,5] in which data for Pb deposited on Si(111)-(6 × 6)Au were presented there is clear evidence that the main QSE conditions, e.g., sufficiently large mean free path of the carriers and thickness uniformity, are fulfilled also for the samples prepared on Si(111)-$(\sqrt{3} \times \sqrt{3})R30°$Ag. The theory of the Hall effect in quantized metallic systems [3] shows that the Hall coefficient is proportional to the reciprocal of the electron density and that its variations with thickness remain rather smooth, in contradiction to experiment. In the free electron QSE model (see, for example, Ref. [7]) the electron density increases continuously with a superimposed weak variation and almost saturates at a thickness of about $5\lambda_F$. The periodicity of this variation is $\lambda_F/2$. In real quantized Pb films it causes a modulation of the specific conductivity with a period close to about 2 ML [see Fig. 1(b)]. This phenomenon was discussed and explained previously [2]. It is difficult to explain the experimental Hall effect data of quantized Pb films within the framework of existing theories of the QSE in metallic layers. According to existing theory the QSE can influence the Hall coefficient

(i) via the variation of the electron density with thickness and (ii) via the anisotropy of the scattering time caused by the interaction of the current carriers with a rough surface [3]. Neither of two effects can reverse the sign of the Hall coefficient observed in experiment. Apparently there are other phenomena which can drastically influence the behavior of the Hall effect in such a system. Some indications come from the band structure of Pb. Anderson and Gold [8] have calculated the Fermi surface using the OPW method. A comparison with the Haas–van Alphen data shows that in bulk Pb the Fermi surface extends into the second and the third Brillouin zone (BZ). Electrons behave in the second BZ like holes and in the third BZ like electrons. It is possible that the size quantization influences the electron/hole ratio and causes dramatic changes of the Hall coefficient. Saalfrank [9] has calculated the band structure of Pb(111) slabs consisting of 1 to 7 ML of Pb. He found that in samples consisting of more than 1 ML of Pb there are always hole and electron pockets around Γ and P symmetry points of the BZ. This clearly indicates that the theoretical models developed to describe the QSE in the electrical resistivity [3,7] break down if they are used to explain galvanomagnetic phenomena such as the Hall effect. Apparently, a model is needed which takes the detailed band structure of Pb under size quantization into account.

In conclusion, we have experimentally determined the Hall coefficient of ultrathin Pb films. We have found that the strong influence of the QSE cannot be explained within the existing theory of the Hall effect in quantized metallic films. A simple comparison of the band structure of Pb with the results presented here indicates that the free electron model is insufficient.

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