Journal of Optoelectronics and Advanced Materials Vol. 7, No. 2, April 2005, p. 677 - 685

DYNAMICS OF SIMULTANEOUS MEASUREMENTS OF HALL AND MAGNETORESISTANCE EFFECTS IN LIQUID METALS AND SEMICONDUCTORS

M. Ogita*, T. Ito, M. Isai, I. Mogi[a], S. Awaji[a], K. Yokoo[b]

Faculty of Engineering, Shizuoka University, Hamamatsu 432-8561, Japan
[a]IMR, Tohoku University, Sendai 980-8577, Japan
[b]RIEC, Tohoku University, Sendai 980-8577, Japan

In the present paper the Hall measurement in the liquid state of metals, using two-frequency method, ac-dc method and simultaneous method will be described. The Hall effect has been measured in Hg and Ga, both solid and liquid state. We have also measured magnetoresistance and Hall effect in InSb single crystal in which magnetoresistance appears even in low magnetic field and Si in which magnetoresistance do not appear clearly. In order to investigate the magnetic field dependence of galvanomagnetic effects in metals (in solid and liquid state) and semiconductors, the measurement in high magnetic field up to ±9 [T] were also performed.

(Received August 18, 2004; accepted March 23, 2005)

Keywords: Hall effect, Magnetoresistance, Liquid, Solid, Two-frequency, Ac-dc method

1. Introduction

The Hall effect has been used to analyze the conduction mechanism in the liquid and solid states of materials. An ordinary dc-dc method is applied to measure the Hall voltage (where method consist of a dc sample current and a dc magnetic field) in the solid state physics. Hall signals generating in metal samples are very weak, less than μV, hence it's difficult to detect in metals, especially in the liquid state. There are several other problems due to electromotive force (EMF), polarization effect and other chemical reactions at the electrodes for measurements of Hall signals in the liquid states [1]**.** Therefore the two-frequency ac method discovered by Russell et al. [2] has been used to avoid the above problems in the liquid state for Hall measurement [1,3,4]. In this method, two different frequencies for sample current and magnetic field are used in order to remove the electrochemical reaction at the electrode interface. This two-frequency ac method has a very high sensitivity and a high frequency selectivity. However, the main drawback of this method is that if a nonlinear component appears in the measuring circuit system and/or the measurement sample itself, a false signal of the same frequency is generated as Hall voltage [5,6]. Furthermore it is not so easy to measure the Hall signal under a dynamic ac magnetic field. Therefore, we have proposed a new method, the ac-dc method in which the magnetic field is produced by a direct current [7]. The advantage of using this method is that it overcomes the nonlinearity effects arising at the interface between electrodes and solutions, and enables to show the sign of positive or negative Hall voltage corresponding to S or N direction of applied magnetic field. However when the signal obtained at the Hall electrode increases with the strength of an applied magnetic field, another false Hall signal was found even if the excellent ac-dc method has been used to detect Hall signal in the liquid state of metals. This false Hall signal does not appear in solid state. It has also been found that this false signal is

* Corresponding author: ogita@eng.shizuoka.ac.jp

generated due to the deformation effect arisen from the Lorentz force in the liquid state. Therefore, an ac current in the high frequency above 1 kHz was used to avoid making a false signal, where this type of false signal has diminished down to as low as white noise level [8]. After taking all precautions, a spurious signal still appears even at the high frequency region in the liquid state. It has also been found after analyzing the spectrum that the spurious signal has a component of B and B^2 for an applied magnetic field in the liquid state although this signal does not appear in the solid state. This fact seems to be an intrinsic magnetoresistance effect [6]. In high magnetic field (up to ±9 T), we have found a drastic change of the galvanomagnetic effect. To analyze and overcome this problem the Hall effect and the magnetoresistance effect were simultaneously measured adding four-point probe method and compared the Hall signal with the magnetoresistance one [10]. In case of solid state semiconductors like Si and InSb big differences on galvanomagnetic effect between low and high magnetic fields do appear.

In the present paper the Hall measurement in the liquid state of metals, using two-frequency method, ac-dc method and simultaneous method will be described. We have measured Hall effect in Hg and Ga, both solid and liquid state. Furthermore, we measured magnetoresistance and Hall effect in InSb single crystal in which magnetoresistance appears even at low magnetic field and Si in which magnetoresistance do not appear clearly. In order to investigate the magnetic field dependence of galvanomagnetic effects in metals (in solid and liquid state) and semiconductors, the measurement in high magnetic field up to ±9 [T] were also performed.

2. Experimental

The measurement circuit is shown in Fig. 1. A ceramic cell of Macor or Pyrex glass cell were used with filling Hg and Ga (99.9999% purity), and mounted at the middle point of magnet air gap of 0-0.5 T dc magnetic field or in a dynamic ac magnetic field of half the dc strength.

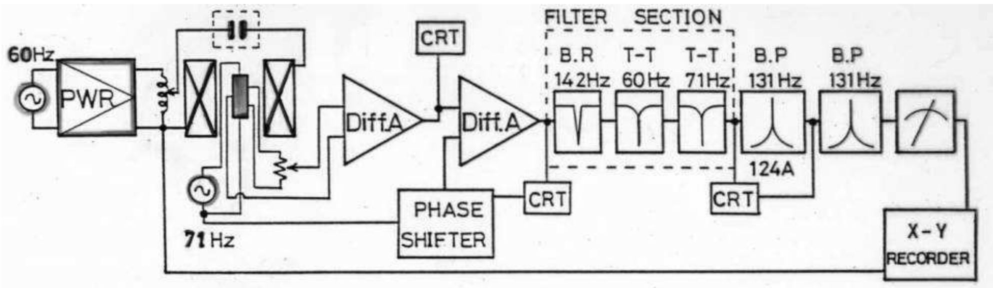

Fig. 1. Diagram of the two-frequency ac method.

The ac electromagnet was made of 0.35 mm laminated iron sheet core and operated in series resonance at 60 Hz with a capacitor bank. The temperature of the cell was monitored with thermocouple or thermistor element to keep a constant temperature. A thermocouple for measuring sample temperature is fixed on the reverse side of the Macor board. Movements of the sample metal surface were observed by detecting a reflected beam from a He-Ne laser using a position sensitive detector (PSD) or photodiode (PD). The variation of the resistance of metals is detected using Kelvin double bridge. A metal sample in liquid state is enclosed into a Macor plate with 200 and 400 μm ditch.

3. Results and discussion

A two-frequency ac method is an excellent method for measurement of a very weak Hall signal less than 1 μV order in noisy and fluctuated solution because this method has a very high sensitivity and a high frequency-selectivity. When an ac sample current of ω_I and an ac magnetic field of a different frequency ω_B are applied to the sample, a true Hall signal is a sum of signal frequency ($\omega_I + \omega_B$) or difference ($\omega_I - \omega_B$). As the result using the intermodulation of two different frequency signals, this sum or difference frequency is different from original frequency of current ω_I and magnetic field ω_B. The data obtained in Fig. 2 apparently seem to be a complete results but spectrum of the frequency shows that there are many harmonic trains in Fig. 3.

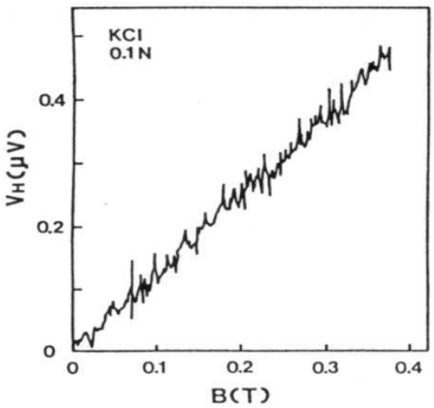

Fig. 2. Hall voltage vs. magnetic field measured by the two - frequency ac method.

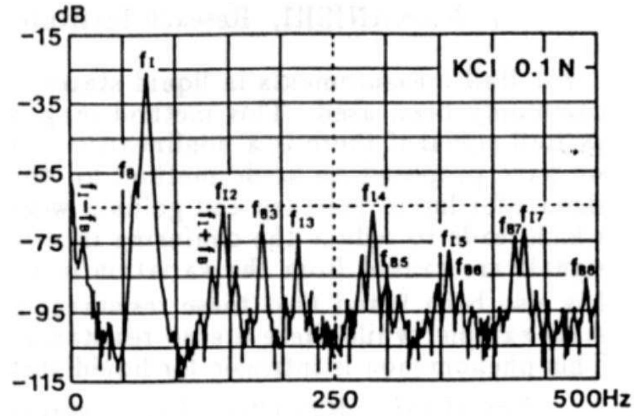

Fig. 3. Frequency spectrum at Hall electrode.

This fact shows the existence of nonlinearity in the measuring system [5]. The nonlinearity has been verified to emerge from the interface between solution and electrode [6]. The movements accompanying shift of the location and/or vibration in the liquid state are shown in Fig. 4. These movements in liquid state metals, e.g., Hg during Hall measurements were also observed at the surface by reflection of a He-Ne laser beam. The frequency spectrum of the surface movement caused by the two-frequency ac method contains the same frequency component as that of the expected true Hall signal in the liquid state, but no corresponding movements were observed in solid state as shown in Fig. 5. In order to obtain Hall signal data in liquid state we have proposed the ac-dc method, which is able to avoid producing a false signal due to nonlinearity at the interface between liquid solution and electrodes.

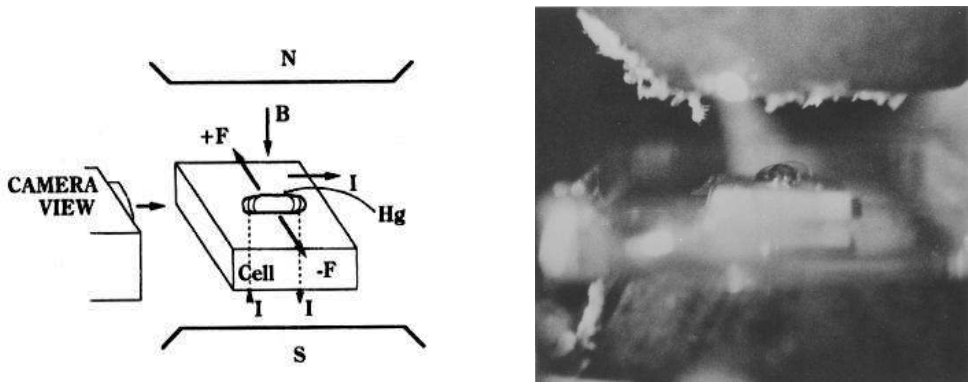

Fig. 4. (A) Schematic diagram as viewed from the side. (B) Movement due to the Lorentz force on the open space of a sample cell are demonstrated and taken by a double exposure at 0.5 T of dc-B and ±2 A dc-I.

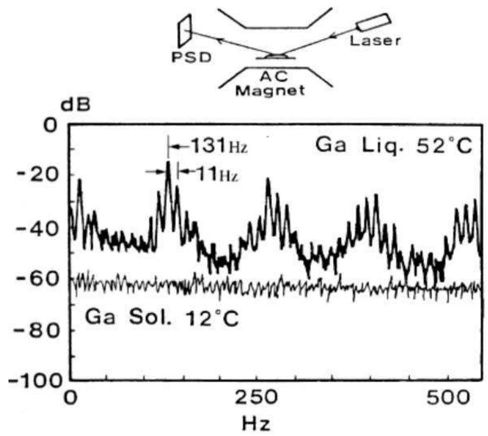

Fig. 5. The spectrum measured with the two-frequency ac method consisting of 60 Hz of ac-B and 71 Hz of ac-I shows surface movements of the liquid Ga sample at 52 ^{o}C accompanied by many higher harmonics including pseudo signals of 11 and 131 Hz. These features cannot be seen for the solid state Ga at 12 ^{o}C.

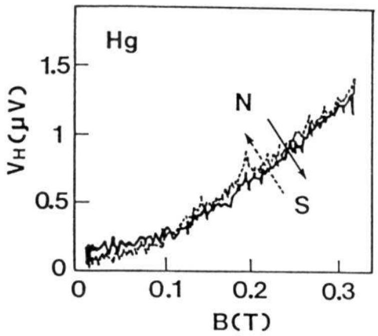

Fig. 6. Hall signal in liquid state of Hg through ac-dc method.

The signals detected at the Hall electrodes have the same frequency as sample current I_S and enables to show the sign of obtained signals corresponding to the polarity of applied magnetic field B. In the case of liquid Hg metal when ac-dc method was applied, the results are, however, shown in Fig. 6, and the Hall signal is reproduced with decreasing the temperature. It may be considered that this unnecessary effect results from distortion of Lorentz force and give rise to a faint variation of resistance in the liquid state samples. The change in resistance was 10^{-3} Ω/Tesla, but no change was observed in solidified metals. The movements of liquid samples during the Hall measurement were observed at the surface of liquid Hg using a reflected of a He-Ne laser beam. The movements were also confirmed by four-point probe method. It has been found that the variation voltage ΔV_R caused by resistance change due to Lorentz force can be reduced to white noise level with increasing the frequency of the sample current over 1 kHz [8]. The unnecessary variation voltages ΔV_R which is shown in Fig. 7 are negligible, increasing the frequency of sample current I_S over 1 kHz in the ac-dc method. The Hall signals with a carrier sign corresponding to the field polarity were obtained at 10 kHz for even the liquid metals using ac-dc method [Fig. 8]. The Hall coefficients, R_H obtained from the Hall measurement for Hg was calculated as a divalent metal and Ga as trivalent metal were 5~7 × 10^{-5} and 7~9 × 10^{-5} cm^3/Coulomb, respectively. The Hall coefficients R_H for Hg and Na showed almost the same values in both the liquid and solid states. Particularly the values for Na were in good agreement with the values expected from the nearly free electron model. The R_H values for Ga, however, showed a big discrepancy between the liquid and solid states. The R_H value in the solid state was nine times as much as in the liquid. The latter value is almost the same as the value calculated from the free electron model.

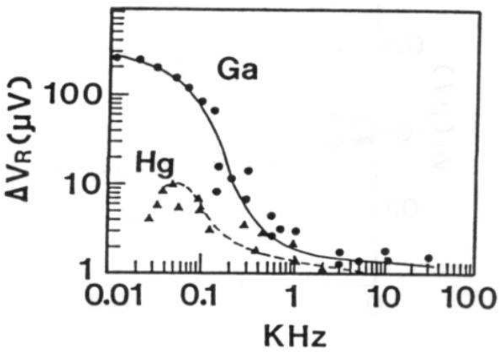

Fig. 7. Incorrect signal, V_R, vs. frequency for Hg and Ga.

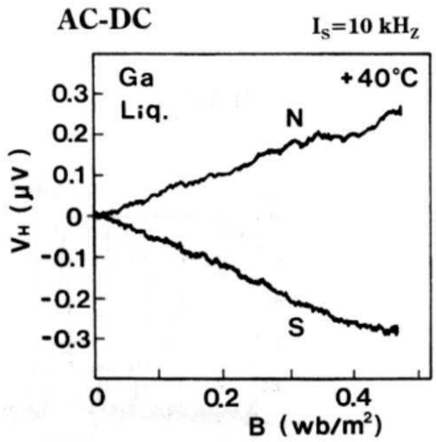

Fig. 8. Hall signals for liquid Ga at 40 °C through ac-dc method at 7kHz ac current.

The Hall coefficient for Ga shows also a big discrepancy at the melting point, but resistivity ρ increases slightly with temperature. The same trivalent metal, In, showed only ordinary metal characteristics of ρ and R_H in both liquid and solid state.

In the higher frequency region around 5 kHz, Hall and magnetoresistance effect were evaluated by two-frequency ac method and a spectrum analyzer (HP3585A) as shown in Fig. 9. Hall signals V_H are expected to reveal at the frequency of ($f_I \pm f_B$) as shown in Fig. 9(b) for solid state, the signals detected at the Hall electrode in liquid state appeared at the frequency of ($f_I \pm f_B$) and ($f_1 \pm 2f_B$) is shown in Fig. 9 (a). It is considered that ($f_1 \pm 2f_B$) components result from an applied magnetic field B. This effect is not a result of nonlinearity at the electrode or nonlinear vibration since there are no harmonic trains in this higher frequency spectrum of liquid state. When the sample was solidified with decreasing temperature, only Hall signal ($f_I \pm f_B$) was appeared [Fig. 9(b)]. Hence the magnetoresistance effect in the liquid state was verified using the four-point probe method as shown in Fig. 9(c). Frequency component ($f_1 \pm 2f_B$) has the same frequency positions as that in liquid state Hall measurement. Subsequently the sample was solidified with decreasing temperature. The spectrum shown in Fig. 9(c) was changed into that shown in Fig. 9(d). Two pairs of sideband disappeared in the solid state and only a fundamental peak appeared at the sample current frequency. This means that the magnetoresistance effect exists in liquid state, but not in solid state in weak magnetic field below approximately 0.3 T. Therefore all the signal obtained at the Hall electrodes in the liquid states are not always true Hall signals, due to magnetoresistance effect. It has been clarified that a magnetoresistance effect intrinsically exists in the sample solution. The Hall voltage in the liquid state measurement is mixed with some other signals.

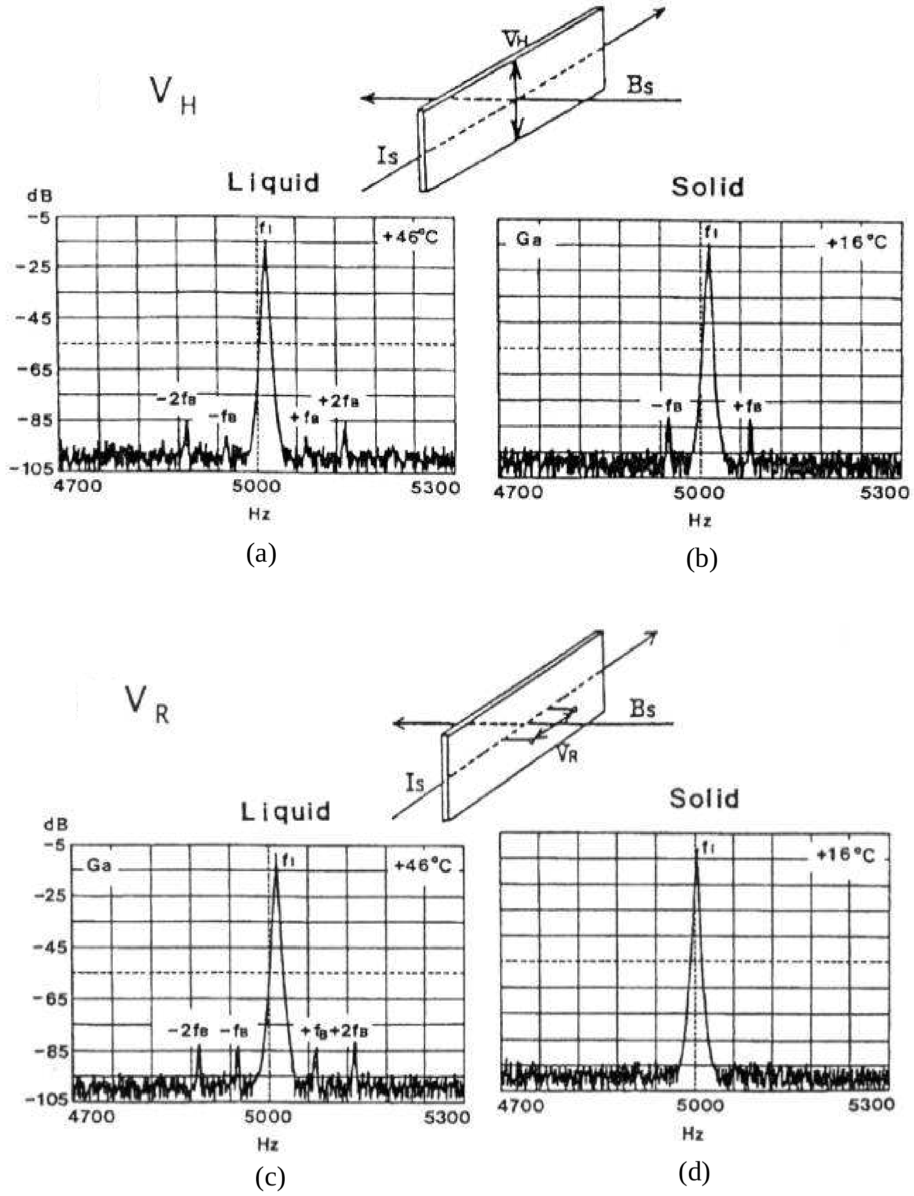

Fig. 9. Spectra of (a), (b) Hall voltage by means of the two-frequency ac method and (c), (d) variation of voltage by means of the four point probe method, for Ga at 46 ^{o}C and solid Ga a t 16 ^{o}C, in the high frequency region for a 5 kHz sample current and 0 kHz, 0.23 T magnetic field, respectively.

To overcome these problems the Hall effect and the magnetoresistance effect were simultaneously measured (Fig. 10). These signals were examined and compared each other in high magnetic field up to ±9 T at IMR, Tohoku University. Fig. 11 shows the results of simultaneous measurements of Hall and magnetoresistance effect in liquid and solid state. The Hg and Ga have basically same type of results. In solid state magnetoresistance effect is quite small, almost negligible, but in liquid state the effect has a big change of parabolic curve on the applied magnetic field B. The Hall voltage in liquid state receives a large affection due to magnetoresistance effect and shows almost clearer B^2 curve as far as magnetic field dependence. This result does not show simply Hall effect any more.

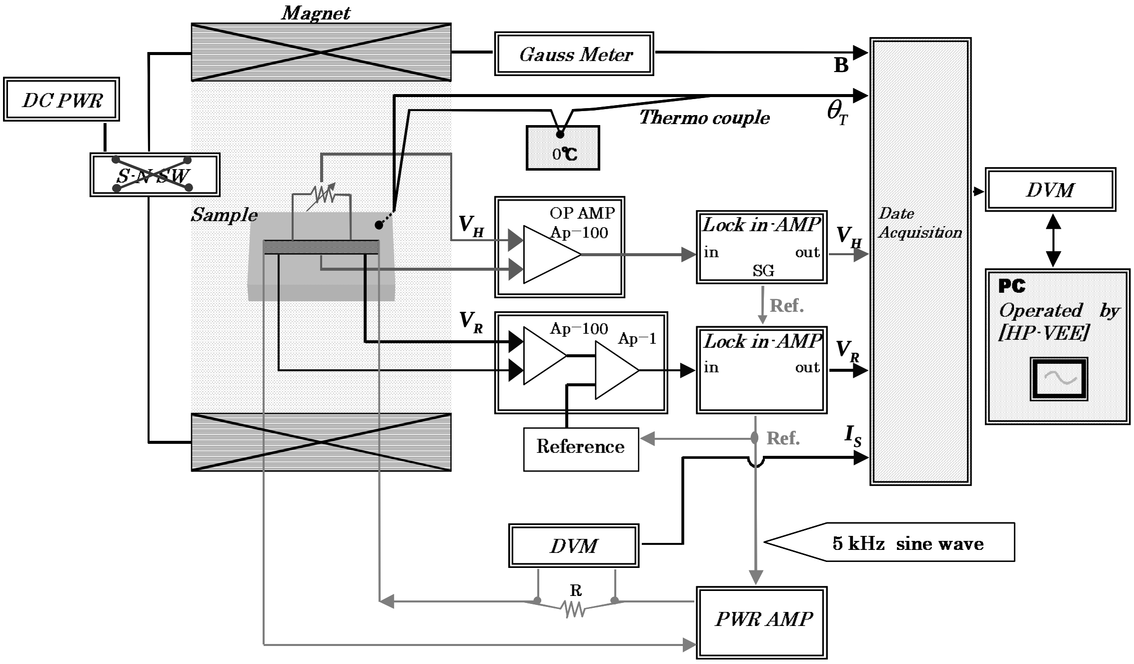

Fig. 10. Block diagram of measurement circuit.

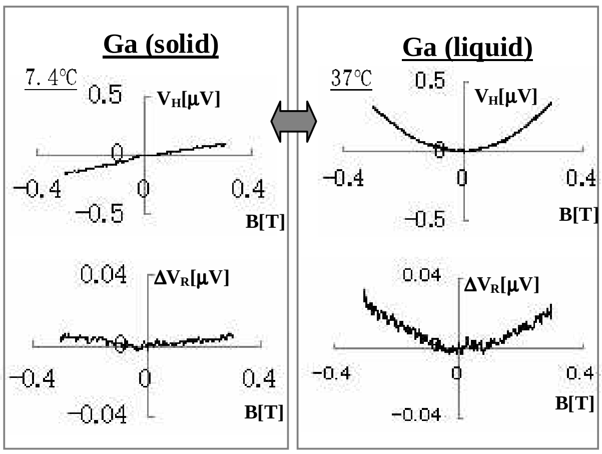

Fig. 11. Hall effect and magnetoresistance effect for Ga (in liquid and solid state).

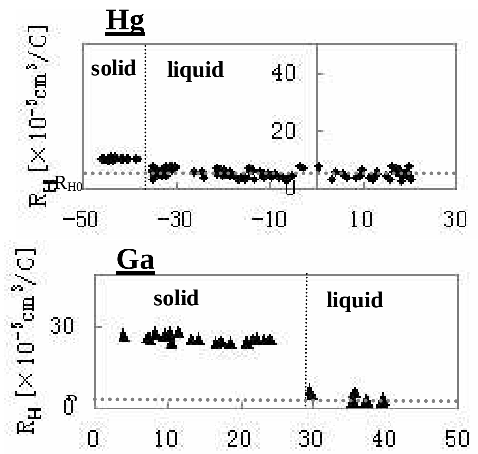

Fig. 12. Temperature dependence of Hall coefficient in Ga and Hg.

The temperature dependence of Hall coefficient in Ga and Hg is shown in Fig. 12. The temperature dependence data of Hall coefficient was obtained by computer data processing using simultaneous measurements in Hg and Ga. Many data affected by magnetoresistance effect are able to be saved by computer data processing using symmetric property of resistance. It has been found that the Hall coefficient is almost constant with temperature. In liquid state the results are in very good agreement with the values calculated from the nearly free electron model and also with other's results [4,9]. Thus, we found that it was possible to pick up the true Hall voltage from obtained Hall signals affected by magnetoresistance effect to a certain extent.

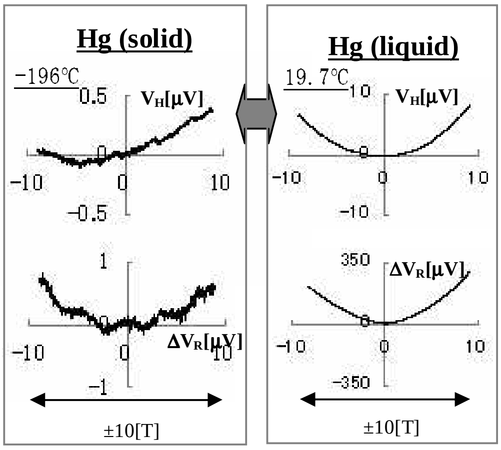

Fig. 13. Hall effect and magnetoresistance effect for Hg (in liquid and solid state) at high magnetic field vertical units are different.

The results on Hall effect and magnetoresistance effect in Hg for high magnetic field are shown in Fig. 13. In liquid state, Hall effect is affected by very big magnetoresistance effect comparing with solid state, where the vertical units are very different in Fig. 13. Therefore we have measured the Hall and magnetoresistance effects for semiconductors in which these effects have generally been well-known in low magnetic field. The result of n-type Si single crystal is shown in Fig. 14(a). It has been found that the magnetoresistance effect does not apper in low magnetic field up to ±0.3 T, but in high magnetic fields it can be observed clearly. The Hall voltage, even of Si has an affection of magnetoresistance effect in high magnetic field up to ±9 T. Contrarily, in case of InSb, which is very well-known to show both the galvanomagnetic effects, and magnetoresistance effect appears even in low magnetic field. On the other hand, in high magnetic field, a very big magnetoresistance clearly appeared and the curve becomes linear excepting near the origin, for an applied magnetic field (Fig. 14 b).

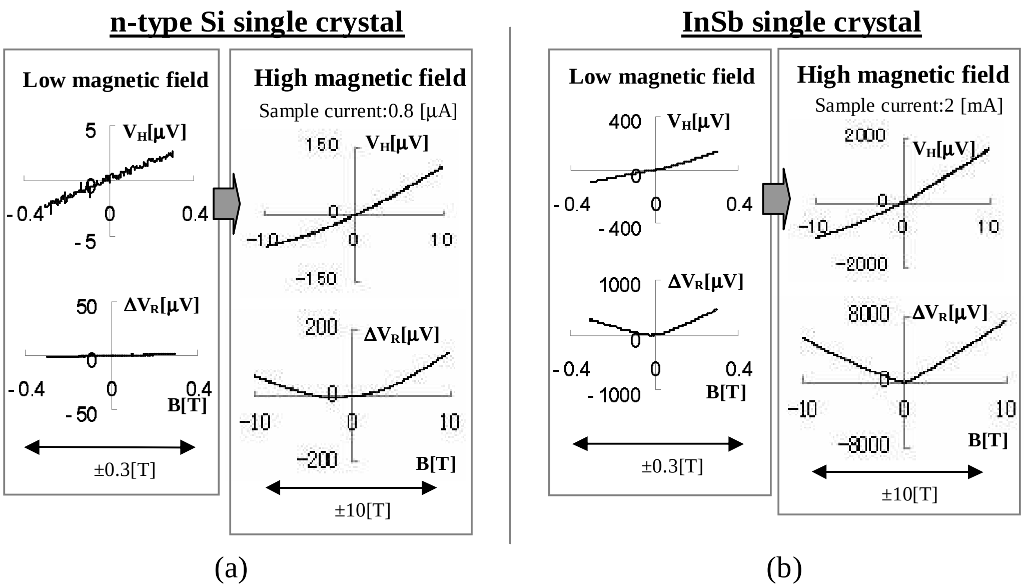

Fig. 14. Hall and magnetoresistance effect in InSb and n-type Si single crystal.

4. Conclusions

The following conclusions has been drawn from above experimental results and discussions:

In liquid state it has been found that during Hall measurements, nonlinearity exists at the electrodes which easily produce false signals similar to the Hall voltage.

Using ac-dc method, Hall signals of Hg and Ga in liquid state were obtained in the higher frequency region above 1 kHz. Although there are indistinguishable spurious signals in liquid state, but not in solid state.

Spurious signal due to change in resistance always exists and it is much larger in liquid state than in solid state. This signal is related to B and B^2 of applied magnetic field.

A discontinuity in Hall coefficient of Ga is observed at the melting point.

The Hall effect is affected in the liquid state by magnetoresistance effect much stronger than in solid state.

The Hall effect can be seen even in high magnetic field in spite of a strong reduction of magnetoresistance effect. Therefore the mechanism of Hall and magnetoresistance effects intrinsically differs from each other although both effects derive from Lorentz force.

References

[1] M.-C. Bellissent, P. Gerard, C. Longevialle, M. Meton, M. Pich, G. Morand, “Experimental Measurements of the Hall Effect in Very Dilute Solutions”, **118**(12), 1944-1950.

[2] B. R. Russell, C. Wahlig, “A New Method for the Measurement of Hall Coefficients,” Rev. Sci. Instrum. **2**, 1028-1029 (1950).

[3] N. E. Cusack, J. E. Enderby, P. W. Kendall, Y. Tièche, “The measurement of the Hall coefficient of liquid conductors,”J. Sci. Instrum. **42**, 256-259 (1965).

[4] A. J. Greenfield, “Hall Coefficients of Liquid Metals,” Phy. Rev. **135**(6A), A1589-A1595 (1964).

[5] Masami Ogita, Shun’ichi Yasuda, “Nonlinearities in the AC Measurement of Hall Effect in Liquids,” J. Electrochem. Soc. **135**(10), 2547-2549 (1988).

[6] M. Ogita, T. Fujinami, Y. Nakanishi, Y. Hatanaka, “Surface reflection and nonlinearities in ac Hall measurements of liquid metals,” Appl. Surf. Sci.. 113/114, 777-782 (1997).

[7] M. Ogita, S. Yasuda, “A Few Perplexities on Two-frequency AC Hall Effects in Solutions,” The Electrochemical Society Extended Abstracts **87-2**, Abstract No. 1478, 2017-2018 (1987).

[8] M. Ogita, S. Yasuda, M. Isai, Y. Nakanishi, I. Tanaka, “Effect of resistance variation for Hall measurements in liquid metals” Proc. Int. ISEM Symp. **5**, 495 – 498 (1994).

[9] J. E. Enderby, “The Hall Effect in Liquid Metals,” Proc. Phys. Soc. **81**, 772-779 (1963).

[10] M. Ogita, M. Nakao, C. D. Singh, I. Mogi, S. Awaji, “A novel approach for simultaneous measurements of Hall effect and magnetoresistance effect in solid and liquid state of gallium and mercury metals” Physica B 346-347, 306 – 309 (2004).