Magnetovolume Effect of SrRuO₃

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The magnetic properties of an itinerant ferromagnet SrRuO₃ have been determined in the hydrostatic pressure, temperature and magnetic field range 0-13.2 kbar, 4.2-300 K, and 0-90 kOe, respectively. The pressure derivatives of the Curie temperature and saturation magnetization at 4.2 K are found to be -0.67 K/kbar and -0.47 emu/g kbar, respectively.

Key words: ferromagnet, magnetovolume effect, SrRuO₃, pressure effect

1. Introduction

SrRuO₃ is known as a ferromagnetic oxide showing a metallic conductivity with a perovskite structure. The Curie temperature T_c and the saturation magnetic moment of SrRuO₃ are ~160 K and 1.1 μ_B per formula unit at 4.2 K, respectively¹⁾⁻³⁾. The crystallographic structure belongs to an orthorhombic system (Pbnm-D_{2h}¹⁶), where Ru ions form nearly cubic cells. SrRuO₃ intrigues as the magnetic system that exhibits magnetic order at low temperature resulting from 4d electrons based primary on the Ru site. Only very recently have the results of band-structure calculations on SrRuO₃ been published by Singh⁴⁾ and Santi and Jarlborg⁵⁾. They find that the main characteristics of the band structure in SrRuO₃ arise from strong Ru 4d- O 2p hybridization. The electrons that originate from the Ru 4d orbitals are responsible for the itinerant magnetism. Furthermore, they emphasized the high sensitivity of the magnetic behavior to parameters such as the unit cell size or the amount of distortion. The aim of this paper is to get an information on the electronic state of the itinerant ferromagnet SrRuO₃ by measuring the pressure change of the magnetization σ at 4.2 K and the Curie temperature.

2. Experimental

Polycrystalline sample of SrRuO₃ was prepared by conventional solid-state reaction from stoichiometric mixture of Ru powder (99.9%) and SrCO₃ (99.9%). The mixture was calcined for 5 hr in air at 1000 °C. The reaction product was then reground, pressed into pellets and sintered in air at 1050 °C for 5 hr.

The crystal structure at room temperature of SrRuO₃ was confirmed with X-ray diffraction to be orthorhombic structure. No additional reflections were observed. The lattice parameters were found to be $a=5.577$ Å, $b=7.848$ Å and $c=5.542$ Å. This result is in good agreement with that recently observed by Shepard et al.⁶⁾.

The temperature dependence of the magnetization at atmospheric pressure was measured using a commercial superconducting quantum interference device (SQUID) magnetometer. The magnetization under hydrostatic pressure up to 10.1 kbar was studied with a subtraction magnetometer by employing a magnetically compensated high-purity binary Cu-Ti pressure cell. The pressure was calibrated from the superconducting critical temperature of Pb. The Curie temperature was determined by an a.c. transformer method. The primary and secondary coils were wound on a sample rod with radius of about 0.5 mm and length of about 10 mm. An a.c. current of a constant amplitude flowed in the primary coil and second voltage, which is directly proportional to initial permeability μ , was recorded as a function of temperature at various pressures up to 13.2 kbar. Hydrostatic pressure was applied to the sample in a Teflon pressure cell filled with a Fluorinert by using a piston-cylinder type device. The temperature was measured using alumel-chromel thermocouple located in

the Teflon pressure cell. The pressure was calibrated from the solid-liquid transition temperature of Hg.

3. Experimental Results and Discussion

Magnetization versus temperature (σ - T) curves under atmospheric pressure, which were measured after the sample was cooled down to 4.2 K without, and in applied fields, are shown in Fig. 1. For zero field cooling (ZFC), the σ - T curve shows a maximum around 140 K, and shows a decrease in magnetization with further decreasing temperature. The magnetization shows an usual curve against temperature for field cooling (FC). The magnetization is not saturated at the field of 1 kOe. Thus, the behavior for the σ - T curve is very different each other for FC and ZFC. This may be attributed to magnetic inhomogeneities present in the material. The Curie temperature was found to be 162.9 K. This was determined from the linear relation between σ^2 and T just below T_c . The inverse susceptibility χ^{-1} follows the Curie-Weiss law above about 200 K. The paramagnetic Curie temperature θ_p and the effective Bohr magneton μ_{eff} were determined to be 163 K and $2.84 \mu_B$, respectively. The value of μ_{eff} determined in the present experiment agrees well with that reported by Callaghan et al.⁷⁾

Figure 2 shows initial permeability μ versus temperature

Fig. 1 Temperature dependence of magnetization σ and inverse susceptibility χ^{-1} for SrRuO_3 . ZFC and FC mean the zero field cooling and the field cooling.

Fig. 2 Temperature dependence of the initial permeability μ for SrRuO_3 at different pressures.

curves for SrRuO_3 at different pressures. As seen in the figure, μ increases rapidly with decreasing temperature. This corresponds to the appearance of the ferromagnetic phase below T_c . With further decrease of temperature, μ decreases rapidly due to the appearance of the strong magnetocrystalline anisotropy energy. The Curie temperature was defined as the inflection point on μ versus T curve as seen in the figure. The Curie temperature at various pressures are plotted against applied pressure in Fig. 3. The Curie temperature was found to decrease linearly with applied pressure in the pressure range investigated. The value of dT_c/dp determined from the result in Fig. 3 is found to be -0.67 K/kbar . This is in good agreement with the values given by Menyuk et al.⁸⁾ and Neumeier et al.⁹⁾ and, however, somewhat smaller than the value reported by Shikano et al.¹⁰⁾

The result of magnetization measurement for SrRuO_3 at various pressures between 2.7 kbar and 10.1 kbar is shown in Fig. 4. The spontaneous magnetization σ_s at 4.2 K was determined by extrapolation to $H/\sigma=0$ of the linear part of the σ^2 versus H/σ plot (Arrott plot) at high fields. At atmospheric pressure the magnetic moment at 4.2 K of SrRuO_3 is found to be $1.1 \mu_B$ per formula unit. As seen in the inset of Fig. 4, σ_s at 4.2 K decreases linearly with pressure. The value of $d\sigma_s/dp$ determined from the result in this measurement is found to be -0.47 emu/g kbar . The

relative change of σ_0 under pressure is given by $d \ln \sigma_0 / dp = -1.7 \times 10^{-2} / \text{kbar}$. It is possible to estimate that within the experimental error $d \ln \sigma_0 / dp \approx d \ln \sigma / dp$ because T_c of SrRuO_3 is high, where σ_0 means the spontaneous magnetization at 0 K.

Takahashi discussed the origin of the itinerant electron ferromagnetism using the spin fluctuation theory¹¹⁾. From this theory, we can derive the following relation for the pressure dependence of T_c and σ_0 :

$$[3d \ln \sigma / dp] / 2 = d \ln T_c / dp - [5d \ln T_0 / dp] / 8, \quad (1)$$

where T_0^{-1} is the longitudinal stiffness constant of spin fluctuation. In Eq. (1), the terms of $d \ln \sigma / dp$ and $d \ln T_c / dp$ for SrRuO_3 were found to be $-17.0 \times 10^{-3} \text{ kbar}^{-1}$ and $-4.2 \times 10^{-3} \text{ kbar}^{-1}$ in this study, respectively. Therefore, it seems that the pressure effect of T_c and σ_0 for SrRuO_3 may be explained by taking account of the pressure effect of T_0 .

Fig. 3 The pressure change of the Curie temperature T_c for SrRuO_3

Fig. 4 Magnetization curves at 4.2 K for SrRuO_3 under various pressures. The inset shows the pressure change of the spontaneous magnetization at 4.2 K for SrRuO_3

References

- 1) A. Kanbayasi: *J. Phys. Soc. Jpn.*, **41**, 1876 (1976).
- 2) F. Fukunaga and N. Tsuda: *J. Phys. Soc. Jpn.*, **63**, 3798 (1994).
- 3) L. Klein, J. S. Dodge, C. H. Ahn, J. W. Reiner, L. Mieville, T. H. Geballe, M. R. Beasley and A. Kapitulnik: *J. Phys: Condens. Matter*, **8**, 10111 (1996).
- 4) D. J. Singh: *J. Appl. Phys.*, **79**, 4818 (1996).
- 5) G. Santi and T. Jarlborg: *J Phys: Condens. Matter*, **9**, 9563 (1997).
- 6) M. Shepard, G. Cao, S. McCall, F. Freibert and J. E. Crow: *J. Appl. Phys.*, **79**, 4821 (1996).
- 7) A. Callaghan, C. W. Moeller and R. Ward: *Inorg. Chem.*, **5**, 1572 (1966).
- 8) N. Menyuk, J. A. Kafalas, K. Dwight and J. B. Goodenough: *J. Appl. Phys.*, **40**, 1324 (1969).
- 9) J. J. Neumeier, A. L. Cornelius and J. S. Schilling: *Physica B*, **198**, 324 (1994).
- 10) M. Shikano, T. Huang, Y. Inaguma, M. Itoh and T. Nakamura: *Solid State Commun.*, **90**, 115 (1994).
- 11) Y. Takahashi: *J. Phys. Soc. Jpn.*, **55**, 3553 (1986).