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Curie Temperature Shift in Gadolinium Films due to Finite-Size Effects
(유한 효과에 의한 Gadolinium 박막의 Curie온도 이동)

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초 록 We determine the Curie temperature of thin gadolinium films by the measurement of magnetization as a function of temperature. From these data, we observe the Curie temperature shift in films from the bulk value, and also see the systematic behavior of this shift with thicknesses of films, that is, the larger the shift is, the thinner the film is. In this paper, we analyze and explain these results in light of finite-size scaling theory.

Abstract 자화도를 측정함으로써 gadolinium 박막의 강자성에서 상자성으로의 전이온도를 결정했다. 박막의 전이온도는 박막의 두께가 얇아짐에 따라 점점 더 bulk계의 전이온도보다 낮아지는 것이 관측되었으며 이 현상은 박막계의 유한성에 의해 일어나는 현상으로 알려져 있다. 우리는 이 결과를 유한효과 및 유한 축척이론에 근거하여 설명하였다.

1. Introduction

Ferromagnetic system, which undergoes the phase transition to the paramagnetic state at Curie temperature, shows the critical behaviors of second order phase transition, such as the anomaly of heat capacity, and the disappearance of spontaneous magnetization at Curie temperature. If at least one dimension of sample is greatly reduced comparable to the correlation length of near Curie temperature, then the physical quantities of this finite system are greatly deviated from those of bulk sample, mainly due to the change of boundary conditions in the reduced geometry. These deviations are systematic and can be formulated as a function of smallest dimension of finite system, which is known as finite-size scaling theory[1]

The simplest manifestation of finite-size effects is that the phase transition temperature of finite system is shifted from that of bulk system. The amount of this shift depends on the smallest dimension of finite system, for example, the film thickness in magnetic film. According to the finite-size scaling theory, the shift of phase transition temperature is expressed as

$$\frac{T_c(\infty)-T_c(d)}{T_c(\infty)} \sim d^{-\lambda} \qquad (1)$$

where $T_c(\infty)$ and $T_c(d)$ are the phase transition temperatures of bulk system and finite system of smallest dimension d, respectively. Also, the critical exponent λ is equal to $1/\nu$, where $\nu(=0.672)$ is the critical exponent of correlation length, $\xi(T)=\xi_0(1-T/T_c(\infty))^{-\nu}$.

Several experimental efforts to test this finite-size scaling theory have been done using liquid helium, which shows macroscopic quantum effects below superfluid transition temperature, T_λ. From the data of heat capacity[2] and superfluid onset temperature[3, 4] in helium film,

the exponent λ is found to be in between 1.0 and 1.8, clearly not equal to the theoretical prediction of 1.48.

The first systematic study on magnetic phase transition have been done by observing the resistivity behavior of nickel films[5] around Curie temperature. The Curie temperature is determined from the first derivative of resistivity (with respect to temperature) data, which show the anomaly at Curie temperature. The justification in determining the Curie temperature in this way is on the assumption[6] that the first derivative of resistivity with respect to temperature might be proportional to heat capacity, and also the fact that the heat capacity generally shows the anomaly at Curie temperature in the second order phase transition materials. In this experiment, they see the systematic Curie temperature shift in films. From the analysis of these shifts in light of finite-size scaling theory, they find that the critical exponent λ is 1.01 ± 0.1, which is again not equal to theoretical prediction of 1.48.

Recently, the magnetization measurements of ultra thin films[7] of Co, Ni and Ni-Co alloys on copper substrate have been done. These data show the abrupt change in the value of crtical exponent of β in magnetization $M(T) \sim (1-T/T_c)^{\beta}$. They identify this behavior as dimensionality cross-over from 3 to 2 dimension. From the Curie temperature shift in films of various thicknesses, they also find the value of exponent λ to be 1.02 and 1.25 for Co and Ni respectively, which also gives the descrepany between theory and experiment. However, the thickest film in this experiment is 16.7 monolayers. Therefore, in these ultra thin films, the two dimensional effects and finite-size effects are mixed up, so the value of λ might be misleading.

There have been experiments to study the phase transition in bulk gadolinium by the measurement of heat capacity[8] or resistivity[9]. However, the systematic measurement on the thickness dependence of any physical properties in this magnetic material has not been reported yet.

We have determined the Curie temperatures for gadolinium films of several thicknesses by the measurement of changes of magnetization with temperatures. In this paper, we will discuss our results in light of finite-size scaling theory. The experimental data show the contradictory result to that given by finite-size scaling theory, which is already shown in the previous works of others.

2. Experimental Details

(1) Preparation of Gadolinium Films

The gadolinium of 99.99% purity is evaporated on Corning 7059 Na free glass substrate by using a Leybold Univex 450 E-Beam Evaporator system. The working pressure is low 10^{-7} Torr by the vacuum system consisted of two turbomolecular pumps and a rotary vane mechanical pump. The film is grown at the rate of 10 Å/sec with less than 10% fluctuation of e-beam power. The thickness of film is monitored during evaporation with a quartz crystal thickness monitor within the error less than 5%. After evaporation, for better crystal formation, the sample is annealed in vacuum at mild temperature of 400℃ for about 4 hours.

(2) Magnetization Measurement

The magnetization of sample is measured by SQUID-based sample magnetometer (Model MPMS7 of Quantum Design). The schematic figure for essential detection part of system is shown in Fig. 1. The sample is magnetized by the field paralell to film sample, produced by superconducting magnet. The sample is then moved through the secondary detector coil. During this process, the induced current in detector coil due to the presence of magnetized object is measured by SQUID. This gives the absolute magnetization of the sample. The sensitivity of this system in determining the magnetization is 1×10^{-8}emu.

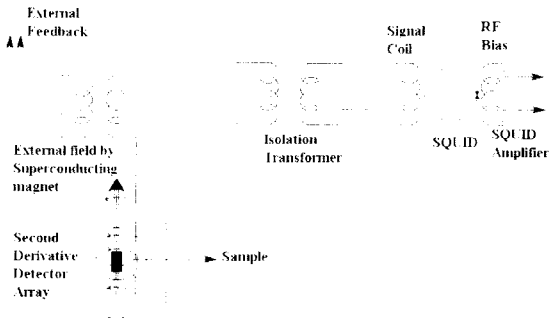

Fig. 1. The schematic figure for essential detection part of SQUID-based sample magnetometer (Model MPMS7 of Quantum Design) is shown in this figure.

The sample itself is located in a sealed sample chamber in which low pressure helium exchange gas provides thermal contact to the sample. System temperature control is achieved with helium gas flow and heater located in an annular region outside the sample chamber. The temperature stability at sample space is less than 0.05K at 300K and the temperature spatial variation in sample chamber is ±0.1K over 8cm.

The Curie temperature of a ferromagnetic sample does not depend on the constant applied field for magnetizing the sample. In high fields, magnetization falls continually with increasing temperature, in low fields it first rises and then falls. However, for both cases, the magnetization goes to zero at same temperature, that is, Curie temperature. We found that the field of 3000 Gauss was enough to get the smoothly decreasing magnetizations with increasing temperatures for all samples we used.

3. Data Analysis and Result

The Curie temperature T_c signifies zero magnetization and separates the ferromagnetic and paramagnetic states. Therefore, we can determine the Curie temperature from magnetization versus temperature data by identifying the temperature of zero magnetization as Curie temperature. The magnetization data near Curie temperature for films of thicknesses 1000Å, 3000Å, 7000Å, and bulk sample are shown in Fig. 2. In this figure, the magnetization axis is not drawn to scale. This is done so because we are only interested in the relative positions of zero magnetization, that is, Curie temperature shifts. In this figure, we can clearly see the systematic shifts of Curie temperature with thicknesses of films, that is, the larger the shift is, the thinner the film is.

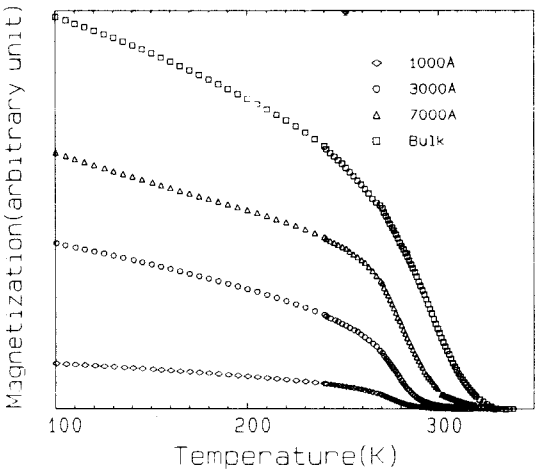

Fig 2. The magnetization data near Curie temperature for films of thicknesses 1000Å, 3000Å, 7000Å, and bulk sample are shown in this figure. We can clearly see the systematic shifts of Curie temperature with thicknesses of films, that is, the larger the shift is, the thinner the film is.

The magnetization versus temperature curves do not cut the temperature axis at large angle but bend over to form a small tail. This extension of magnetization above the estimated T_c is mainly due to the surface effect, that is, finite-size magnetic spin correlation effect. Therefore, the phase transition in the film is likely to be broader than the bulk phase transition[10, 11] In this case of a round phase transition, the inflection point is usually chosen as the Curie temperature[12]. This inflection point can be determined from dM/dT versus T curve. One example of this procedure(for 3000 Å) is shown in Fig. 3. From this curve, the

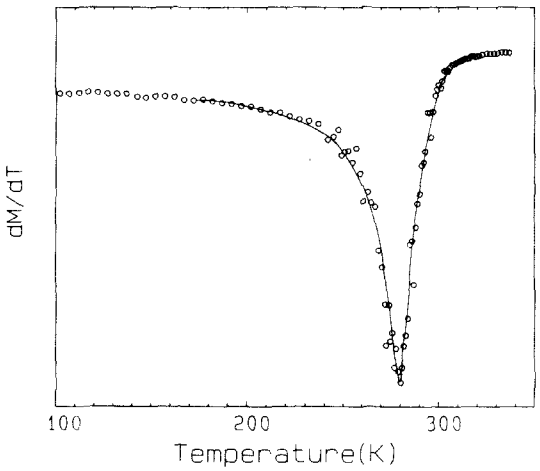

Fig. 3. The dM/dT versus T curve to determine the Curie temperature for 3000Å sample is shown in this figure. The solid line guides the eyes.

Curie temperature of 3000Å film is determined to be $280\pm3K$. Here, the error indicates the ambiguity in determining the minmum position of dM/dT in Fig. 3. Using the same procedure, we determine the Curie temperatures of 1000Å, 7000Å films and bulk sample to be 270 ± 2K, 285 ± 4K and 291 ± 2K, respectively.

To test finite-size scaling theory, equation (1) is casted into

$$\ln(T_c(\infty)-T_c(d))\sim\lambda\ \ln d+\text{const.} \quad (2)$$

by taking logarithms of both sides in equation (1). Then, if we plot $\ln(T_c(\infty)-T_c(d))$ versus $\ln d$, the slope of this line will be equal to $-\lambda$. These are shown in Fig. 4. From this figure, we can find the exponent λ to be 0.80 ± 0.15, not equal to the theoretical expectation of 1.48, which is already shown in the previous works of others both in liquid helium and magnetic systems. The discrepancy between theory and experiments might come from the assumption that for a finite system, the same divergent correlation length as for the bulk is used, i.e. no new critical length scale is manifest due to the finiteness of the system, which is central assumption of finite-size scaling theory (for more discussion, refer to ref. 3). Even though there are many experimental data against the finite-size scaling theory in both liquid helium and magnetic systems, we still need more collections of data for clearer answer for this. We believe that our experimental result is one of them.

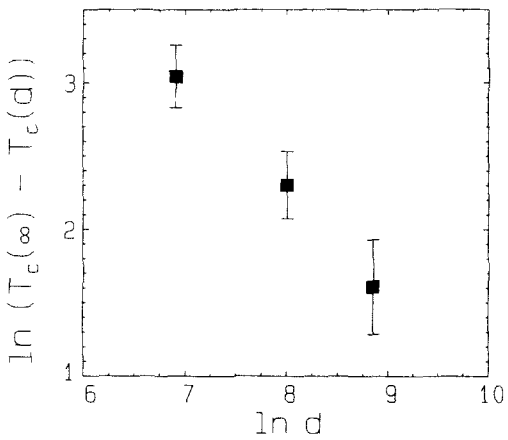

Fig. 4. The $\ln(T_c(\infty)-T_c(d))$ *versus* $\ln d$ curve to test finite-size scaling theory is shown in this figure. The slope of this line is determined to be 0.80 ± 0.15.

4. Conclusion

We determine the Curie temperatures of gadolinium films of several thicknesses by the measurement of changes of magnetization with temperatures. From these data, we can clearly see the systematic shift of Curie temperature with film thicknesses, which we believe, are due to the finiteness of sample. By analyzing the data in light of finite-size scaling theory, we determine the critical exponent λ to be 0.80 ± 0.15, clearly not equal to the theoretical prediction of 1.48, which is already seen in other experimetal works. However, for clearer justification of failure of finite-size scaling theory, we still need more collections of experimental data such as the heat capacity, resistivity and magnetization data in films of other ferromagnetic materials. Also,

we expect a new theory to resolve t.. ences between theory and experiments.

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