Study of Magnetic Phase Transitions in YVO_3 by Non-linear Susceptibility

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The linear and non-linear ac-susceptibilities of the Mott insulator YVO_3 were measured using a Hartshorn mutual inductance bridge. It is found by the temperature dependence of ac-susceptibilities that there are two magnetic phase transitions at 72.1 and 112.0 K. Above 112 K, the values of linear and non-linear susceptibilities qualitatively consist with the result of the mean-field theory. It is verified that there is a power-law dependence between linear and non-linear susceptibilities, which is predicted by the mean-field theory.

Key words: linear and non-linear ac-susceptibilities, YVO_3 , Mott insulator, perovskite, antiferromagnet

1. Introduction

Non-linear susceptibility is a good probe to study the phase transition related to a spin correlation. For example, the non-linear susceptibility only shows a divergent behavior at a spin freezing temperature. In a quadrupolar ordering in rare earth compounds,¹⁾ it was proposed that the study of non-linear susceptibility is very complementary to that of linear susceptibility and is the observation of well defined anomalies in the temperature dependence. The mean-field theory expected for the expansion of a Brillouin function that the value of non-linear susceptibility is negative and there is a power-law relation between the linear and non-linear susceptibilities.²⁾

Since the measurement of non-linear susceptibility sees only non-linear effects, it is not sensitive to the eddy currents produced in a metallic gasket of a diamond-anvil cell by an alternative magnetic field. Thus non-linear susceptibility can be used to determine the pressure dependence of phase transition temperature.³⁾

Transition-metal oxides which have a perovskite-like structure have been extensively studied since the discovery of high- T_c superconductors. Solid solutions of the perovskite type compound $\text{Y}_{1-x}\text{Ca}_x\text{VO}_3$ exhibit an insulator-metal transition at the dopant calcium ion concentration around $x = 0.5$.^{4),5)} The parent compound YVO_3 is a Mott insulator with an orthorhombic GdFeO_3 -type structure.^{6),7)} Magnetic specific heat measurements showed two phase transitions, that is, one is the transition of second order at 115 K and the

other of first order at 78 K. The magnetic structure of intermediate phase is antiferromagnetic in both the inter- and intra-planes, a G-type antiferromagnetic structure, while that of low temperature is antiferromagnetic in the intra-plane but ferromagnetic in the inter-plane, a C-type antiferromagnetic structure.⁷⁾ However, the recent neutron diffraction measurement showed that YVO_3 orders with the C-type antiferromagnetic structure at 118 K and changes to the G-type antiferromagnetic structure at about 77 K.⁸⁾

In this study, we have carried out the measurement of the linear and non-linear susceptibilities on YVO_3 to study the magnetic phase transitions. We discuss the relation between the linear and non-linear susceptibilities around the magnetic phase transition.

2. Experimental procedures

Y_2O_3 , V_2O_3 and VO were mixed together in the correct molar proportion and heated at 1000 °C; the product was ground and heated at 1300 °C. The polycrystalline samples of YVO_3 were synthesized from the melt by a floating-zone method. The sample was confirmed to be a single phase by the powder X-ray diffraction.⁵⁾

A Hartshorn-type bridge incorporating two lock-in amplifiers is used to monitor temperature-induced changes in the mutual inductance of copper coil system consisting of a coaxial pair of two oppositely wound secondary coils and a primary coil. Each secondary coil is 43.5 mm long and contains 4900 turns in 14 layers of 0.12 mm diameter Cu-wire. The primary coil is 140 mm long and contains 700 turns in two layers of 0.34 mm diameter Cu-wire. This coil system remains at liquid-helium or liquid-nitrogen temperatures throughout given experiments.

The induced voltage in secondary coils is proportional to the time differential of magnetization. The magnetization, m , is expanded in powers of the applied field h around the magnetic phase transition as

$$m = \chi_0 h + \chi_2 h^3 + \chi_4 h^5 + \dots,$$

where χ_n ($n = 0, 2, 4, \dots$) is susceptibility and usually has a complex form. Therefore when an alternative magnetic field, $h_0 \sin \omega t$, is applied, the induced voltage, V , in the secondary coils, one contained the

sample and the other empty, is given as

$$\begin{aligned} V &= -Ah_0\omega[(\chi_0 + \frac{3}{4}\chi_2 h_0^2 + \dots)\cos\omega t \\ &\quad - \frac{3}{4}(\chi_2 + \frac{5}{4}\chi_4 h_0^2 + \dots)h_0^2\cos 3\omega t + \dots] \\ &= -Ah_0\omega[\chi_0^t\cos\omega t - \frac{3}{4}\chi_2^t h_0^2\cos 3\omega t + \dots], (1) \end{aligned}$$

where

$$\begin{aligned} \chi_0^t &= \chi_0 + \frac{3}{4}\chi_2 h_0^2 + \dots, \\ \chi_2^t &= \chi_2 + \frac{5}{4}\chi_4 h_0^2 + \dots, \end{aligned}$$

and A is constant related to the coil condition and the sample.

Balancing of the mutual inductance bridge is initially accomplished without a sample at liquid-helium or liquid-nitrogen temperature. Then any change in the mutual inductance with a sample located at the center of one of the secondary coils is related to the change in the susceptibility of the sample. The output signal from Hartshorn mutual inductance bridge was fed to two lock-in amplifiers. One was operate at the frequency of ω , and the other at 3ω . Hence we have observed the fundamental, linear, susceptibility and the third harmonics, non-linear, susceptibility simultaneously. We have also measured not only in-phase, real component, but also out-of-phase, imaginary component, of susceptibility using a two-phase lock-in amplifier. The analyzed output voltage was calibrated by measuring the susceptibility of Gd_2O_3 .⁹⁾ The phase scale was calibrated by measurements for the susceptibility of Nb and $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ in the superconducting state.

3. Results and Discussion

Figure 1 shows the real components of linear and non-linear ac-susceptibility, $\text{Re}\chi_0^t$ and $\text{Re}\chi_2^t$, as a function of temperature measured with $h_0 \sin \omega t$ at $h_0 = 34.2$ Oe and $\omega = 150$ Hz. The data were recorded with decreasing temperature in which the coil system remained at liquid-helium temperature. It is found that these temperature dependences exhibit two distinct anomalies at $T_N = (112.0 \pm 0.2)$ and $T'_N = (72.1 \pm 0.2)$ K which indicate magnetic phase transitions. Thus non-linear susceptibility shows a distinct anomaly at the first order phase transition temperature. The $\text{Re}\chi_2^t$ value is negative and is observed only between about 1 K above and below transition temperatures. These anomalies are observed not only in $\text{Re}\chi_0^t$ and $\text{Re}\chi_2^t$ but also in the imaginary components of linear and non-linear ac-susceptibilities. As shown in Fig. 1, we did not observed a significant difference between temperature dependences of χ_0^t and χ_2^t around T_N and T'_N although the transitions are of the second order at T_N and of the first order at T'_N .

Fig. 1 Real components of linear and non-linear ac-susceptibilities, $\text{Re}\chi_0^t$ (a) and $\text{Re}\chi_2^t$ (b), as a function of temperature measured with $h_0 \sin \omega t$ at $h_0 = 34.2$ Oe and $\omega = 150$ Hz. The coil system remained at liquid-helium temperature throughout this experiment.

These two transition temperatures are not in good agreement with previous studies.^{7),8)} One of the reasons for this disagreement is different oxygen content in samples. Moreover, the transition at T'_N is of the first order with lattice distortion. The hysteresis in the temperature dependence of magnetic Bragg reflections is observed by the neutron diffraction measurement.⁸⁾ Thus the lower T'_N value determined by the present work seems to be caused by the supercooling effect in the first order phase transition.

We measured χ_0^t and χ_2^t using an alternating field with finite amplitude which include the higher harmonic terms as shown in Eq. (1). Then we have measured the h_0 dependence of χ_0^t and χ_2^t in the range from 19 to 35 Oe at 157 Hz around T_N . The data were recorded with decreasing temperature in which the coil system remained at liquid-nitrogen temperature. Figure 2 shows the result of h_0 dependence of $|\chi_0^t|$ and $|\chi_2^t|$ at 112.35 K just above T_N where $|\chi_n^t|$ means $\sqrt{\chi_n^t \times \chi_n^{t*}}$ ($n = 0, 2$). As shown in Fig. 2, there is no h_0 dependence of $|\chi_0^t|$ and $|\chi_2^t|$ within the experimental accuracy. This result suggests that the contribution of higher harmonics in χ_0^t and χ_2^t can be ignore around T_N below 35 Oe. Therefore we directly

Fig. 2 Variations of $|\chi_0^t|$ and $|\chi_2^t|$ as a function of h_0^2 with $\omega = 150$ Hz at 112.35 K just above T_N where $|\chi_n^t|$ means $\sqrt{\chi_n^t \times \chi_n^{t*}}$. The open and closed circles are for $|\chi_0^t|$ and $|\chi_2^t|$, respectively. The coil system remained at liquid-nitrogen temperature throughout these experiments. Broken lines are to guide the reader's eye.

observed χ_0 and χ_2 in these experimental conditions.

In an antiferromagnetic phase transition, the mean-field theory gives the result that the sign of χ_2 value changes at T_N , that is, χ_2 is negative above T_N and is positive below T_N .²⁾ Since the observed $\text{Re}\chi_2$ value is negative around T_N as shown in Fig. 1 and we directly observed χ_2 , the χ_2 value is negative around T_N . This result qualitatively consists with that of the mean-field theory above T_N but is inconsistent with that below T_N .

Furthermore, the mean-field theory predicts that there is a relation between χ_0 and $-\chi_2$ above the ferromagnetic and antiferromagnetic phase transition temperatures being given as

$$\chi_0 \propto -\chi_2^\eta$$

where $\eta = 4$.²⁾ Figure 3 shows the relation between $\text{Re}\chi_0$ and $-\text{Re}\chi_2$ just above T_N . As shown in Fig. 3 on a double logarithmic plot, there is a linear relation between $\text{Re}\chi_0$ and $-\text{Re}\chi_2$; the power-law dependence. Thus the power-law predication is verified experimentally by the present work. However, the η value is estimated to be (1.31 ± 0.04) which is different from the mean-field value. Since the non-linear susceptibility around T_N is observed only in temperature region from 113 to 111 K, the non-linear susceptibility can be the critical phenomena. The imaginary components of linear and non-linear susceptibilities, which represent losses are observed around T_N and T'_N . These two facts probably cause the quantitative inconsistent between the experimental result and the mean-field prediction.

In conclusion, we have measured linear and non-linear ac-susceptibilities, χ_0 and χ_2 , of YVO_3 . It is found by the temperature dependence of ac-susceptibilities that there are two magnetic phase tran-

Fig. 3 $\text{Re}\chi_0$ plotted against $-\text{Re}\chi_2$ in temperature range from 112.78 to 112.06 K on a double logarithmic plot. The solid line represents the fitting line obtained by the analysis. χ_0^t and χ_2^t were measured with $h_0 \sin \omega t$ at $h_0 = 34.2$ Oe and $\omega = 150$ Hz. The coil system remained at liquid-nitrogen temperature throughout this experiment.

sitions at 72.1 and 112.0 K. Above 112 K, the χ_0 and χ_2 values qualitatively consist with the result of the mean-field theory; the χ_0 value is positive and the χ_2 value negative. It is verified that there is a power-law dependence between χ_0 and χ_2 , above 112 K, which is predicted by the mean-field theory. The coefficient of this power is estimated to be 1.31 which is inconsistent with the prediction.

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