High-frequency dielectric anomaly with metal-insulator transition in $CuIr_2S_4$

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Abstract

Electromagnetic response of $CuIr_2S_4$ at 34 GHz has been measured by a cavity perturbation method. Clear anomalies appear in the temperature dependences of the resonant frequency and the full width at half-maximum (FWHM) of the Q-curves at the metal-insulator phase transition. These anomalies of the microwave responses are very suggestive of a simultaneously charge-ordering phase transition with the metal-insulator phase transition of $CuIr_2S_4$.

Key words: high-frequency; MI transition; charge-ordering; $CuIr_2S_4$

1. Introduction

Recently a spinel-type compound $CuIr_2S_4$ has attracted much attention for revealing the mechanism of the metal-insulator (MI) transition at 230 K [1,2]. High temperature paramagnetic phase is metallic while low temperature diamagnetic phase is insulating. The MI transition is accompanied by a first-order structural phase transition from the cubic phase (M) to the tetragonal phase (I) [1]. Since the valence of Cu is known to be +1 from the XPS measurements [3], the mean valence of Ir is +3.5. One expects that the MI transition comes up with a charge-ordering (CO) state of Ir^{3+} and Ir^{4+} [1,3,4].

Radaelli *et al.* [4] reported that a crystallographic structure in insulating phase is not tetragonal but triclinic by means of high-resolution X-ray diffraction. It is also interesting to note that they observed a recovery of the crystalline symmetry at temperatures below 50 K. We plan to search evidence for a possible CO transition of $CuIr_2S_4$ at 230 K and a crystalline transition at 50 K.

In this paper, we report the electromagnetic response of $CuIr_2S_4$ revealed by a cavity perturbation method.

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2. Experimental

Microwave experiments were done by a cavity perturbation method by using a vector network analyzer (ABmm, MVNA 8-350), where TE_{011} mode was employed at the resonant frequency of 34 GHz in the temperature range from 4.2 K to 300 K. Temperature was controlled by a variable temperature insert (Oxford). A cylindrical cavity was made of oxygen free copper. Automatic frequency control (AFC) system was used if necessary. We can monitor the resonant frequency F and the transmitted power P continuously as a function of temperature with the aid of AFC.

Polycrystalline samples of $CuIr_2S_4$ were prepared by solid state reaction. The sample was placed on a quartz rod at the electric-field maximum of microwave .

The Q-curve was measured at the various different temperatures. Resonant frequency F and the full width at half-maximum (FWHM) Γ were analyzed from the Q-curve. We also performed the blank mesurements where the resonant frequency is F_0 and FWHM is Γ_0. Actually, we focus on the differences ($\Delta F = F - F_0$, $\Delta\Gamma = \Gamma - \Gamma_0$) between the sample and the blank measurements.

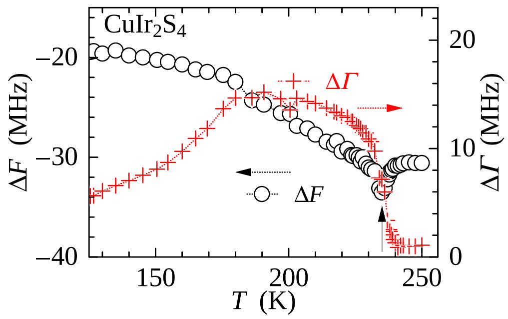

Fig. 1. ΔF and $\Delta\Gamma$ of $CuIr_2S_4$ as a function of increasing T (see text for notion).

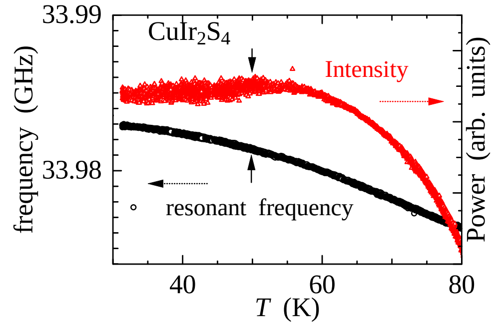

Fig. 2. Continuous monitoring of resonant frequency F and the microwave power P as a function of increasing T, using AFC.

3. Results and Discussion

In Fig. 1, we show ΔF and $\Delta\Gamma$ of $CuIr_2S_4$ as a function of increasing temperature (125 − 250 K). A remarkable feature of Fig. 1 is that ΔF increases at temperatures above 235 K. This is anomalous because the behaviour is different from what the depolarization peak predicts [5], i.e., ΔF should decrease in a metallic side with the conventional MI transition.

As ΔF is proportional to a real part of the complex dielectric constant $-\varepsilon_R$ [5], the ΔF increase at temperatures above 235 K may be caused by dielectric anomaly at the MI transition. The ΔF increase is also observed for the CO phase transition of $Pr_{0.5}Ca_{0.5}MnO_{3-\delta}$[6] where the CO phase transition is confirmed to occur simultaneously with the MI transition. We suggest that the CO transition is likely to appear with the MI transition of $CuIr_2S_4$.

The broad peak in $\Delta\Gamma$ and the ΔF increase with temperature, observed below 235 K (see Fig. 1), are reasonably explained in terms of depolarization peak. Generally speaking, the appearance of depolarization peak indicates at least a change by the two order of magnitude in resistivity. Actually, this is satisfied in the resistivity measurments of $CuIr_2S_4$ [2]. The curves of Fig. 1 have hysteresis with temperature, as was reported for the resitivity [2] as well as the susceptibility [1]. The anomalies appeared near 220 K in decrease temperature.

In Fig. 2, we show F and P as a function of increasing temperature (30 − 80 K) with the aid of AFC. There is no anomaly in F and P near 50 K where a crystallographic structural transition was reported [4]. We note that a resonant frequency has a monotonic decrease with temperature even for an empty cavity. This comes from a thermal expansion of the cavity. The P decreases above 60 K is also due to a change in resistivity of cavity material. Since Γ is proportional to $P^{-1/2}$, the P decrease means the increase in the Γ. We consider that there are no anomalies on the dielectric response at temperatures below 50 K within a limit of the measurement appratus.

We note that the irradiation of the intense X-ray may induce the crystallographic structural transition [7].

4. Conclusion

We observed the high-frequency anomaly with the metal-insulator phase transition of $CuIr_2S_4$ at 230K. Although this anomaly is partly explained by the effect of the depolarization peak, we observed the dielectric anomalies of $CuIr_2S_4$. There are similar to those of dielectric anomaly in the CO transition of $Pr_{0.5}Ca_{0.5}MnO_{3-\delta}$. This results suggest that the CO transition of $CuIr_2S_4$ occurs simultaneously with the MI transition. The frequency response did not manifest a clear anomaly at 50 K where a crystallographic structural transition was proposed.

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