Neutron Scattering Studies of Ce(Zn_{1-x}Cu_x)_2

T. OSAKABE, M. KOHGI, K. OHOYAMA and T. KITAI

Submitted to Physica B.
National Laboratory for High Energy Physics, 1992

KEK Reports are available from:

Technical Information & Library
National Laboratory for High Energy Physics
1-1 Oho, Tsukuba-shi
Ibaraki-ken, 305
JAPAN

Phone: 0298-64-1171
Telex: 3652-534 (Domestic)
(0)3652-534 (International)
Fax: 0298-64-4604
Cable: KEKOHO
NEUTRON SCATTERING STUDIES OF Ce(Zn_{1-x}Cu_x)₂

T. Osakabe, M. Kohgi, K. Ohoyama and T. Kitai*

Department of Physics, Tohoku University, Sendai 980, Japan
*Department of Electronics, Kyushu Institute of Technology, Kitakyushu 804, Japan

Abstract

Neutron quasi-elastic scattering studies and susceptibility measurements of Ce(Zn_{1-x}Cu_x)₂ with x = 0, 0.1, 0.15, 0.2 and 0.25 are reported. While the Néel temperature falls rapidly with increasing x and no ordering appears around x = 0.2, the linewidth of the quasi-elastic scattering remains constant.

KEYWORDS: Neutron scattering, Crystal field, Kondo effect, RKKY interaction, Ce(Zn_{1-x}Cu_x)₂

1. Introduction

CeZn₂, which has the same crystal structure as CeCu₂ (Imma space group), orders antiferromagnetically below 7.5 K with a metamagnetic magnetization process along the easy b-axis. It shows a Kondo behavior in the electrical resistivity[1]. The substitution of 25% of Zn atoms by Cu atoms suppresses the ordering temperature considerably (T_N < 1.5 K) [2] and yields more similar magnetic properties as those of CeCu₂ which shows typical properties of Kondo lattice. This indicates that there is a possibility that there exists a competition between the Kondo effect and the RKKY interaction in the mixed compound Ce(Zn_{1-x}Cu_x)₂. Of course there is another possibility that the phenomena are caused by a competition between the different kind exchange interactions or between different magnetic anisotropy since both side compounds show different types of magnetic ordering at low temperatures[3,4]. In order to clarify this point we have performed neutron scattering experiments as well as susceptibility measurements. As reported in a separate paper[5], we have observed well defined crystal field excitations for CeZn₂ at 16 meV and 39 meV. With increasing x, the excitation energy decreases linearly, while its linewidth increases with upward curvature. In this paper, we report the results of the neutron quasi-elastic scattering and magnetic susceptibility measurements.

2. Experimental details

Polycrystalline samples were prepared by melting together the required amount of Ce(3N), Zn(5N) and Cu(4N) in tantalum crucibles in 3 hours at
1150 °C. cooling at the rate of 1 °C/min. and holding for 24–48 hours at 450 °C. Single crystal samples were picked from the products. The susceptibilities were measured on the single crystal samples with x = 0.1, 0.15, 0.2 and 0.25 along a, b and c axis using a SQUID magnetometer in the temperature range from 1.9 K to 270 K under applied field of 5 kOe. For x = 0.2, we observed the susceptibility between 0.5 K and 5 K using Faraday method in a 3He cryostat. The Néel temperatures were determined from the cusps of the susceptibilities.

The neutron quasi-elastic scattering(QES) experiments were carried out on the crystal analyzer spectrometer LAM-D at the pulsed neutron source KENS in National Laboratory for High Energy Physics. The spectrometer is installed at a thermal beam hole and employs 4 pyrolytic-graphite energy-focusing-type analyzer, each with its own beryllium filter and detector. The analyzers are positioned at the scattering angles of 35° and 85° at both sides of the incident beam with fixed energy of 4.6 meV. Polycrystalline samples of the compounds with x = 0.0, 0.1, 0.15 and 0.25 were used in the experiments.

3. Results

Fig. 1 shows the results of the susceptibility measurements of the compounds with x = 0.1, 0.15, 0.2 and 0.25. The anisotropy of the magnetic susceptibilities of the compounds is large due to the low symmetry of the crystal structure. The easy axis changes systematically from b-axis to a-axis with increasing x. The solid lines in fig.1 are the results of least squares fit to the data with the theoretical magnetic susceptibility, $\chi^{-1} = \chi_{CEF}^{-1} + \lambda$.

Here, $\lambda$ is the molecular field parameter and $\chi_{CEF}$ is the susceptibility derived from the CEF Hamiltonian:

$$H_{CEF} = B_2^0 Q_2^0 + B_4^0 Q_4^0 + B_4^2 Q_4^2 + B_4^4 Q_4^4$$

where $B_{2n}^m$ and $Q_{2n}^m$ are the CEF parameters and Stevens' operators.

Table 1 lists the CEF excitation energies from the ground state to the first excited state obtained from the fit together with those determined by the neutron inelastic scattering experiments[5].

Fig. 2 shows the quasi-elastic spectra, $S(Q,\omega)$, of the system with x = 0, 0.1, 0.15 and 0.25 at scattering angle of 35° at 30 K, 23 K, 24 K and 24 K, respectively. The raw data measured by the LAM-D spectrometer were processed for background subtraction, incident flux normalization and absorption correction. The lines in fig. 2 show the results of least squares fit to the data with a Lorentzian as the spectral function of the quasi-elastic scattering, where the thick solid lines and the broken lines correspond to the quasi-elastic and incoherent elastic scattering, respectively. In the fitting procedures, the energy resolution, which is about 0.5 meV (HWHM) at $\hbar\omega=0$, was convoluted. The fitting range was from -4 meV to 4 meV. We also tried to fit with a Gaussian, but the agreement with the data was not satisfactory.

Fig. 3 summarizes the results of the neutron scattering experiments. The closed and open circles are the linewidths of the CEF scattering and quasi-elastic scattering, respectively. The Néel temperatures determined from the susceptibility measurements are also plotted by open squares. For x=0.2, no magnetic ordering was found down to 0.5 K.
4. Discussion

The result of the quasi-elastic scattering experiment shows that the spectra of Ce\(\text{Zn}_{1-x}\text{Cu}_x\) are reproduced well by a Lorentzian spectral function at around 20 K. However the linewidth of the spectra remains constant with increasing \(x\) (HWHM = 0.4 meV). This means that, though the spin fluctuations of the the ground state in this system are mainly caused by the single site spin relaxation process at about 20 K, the energy spread of the spin fluctuations does not change with increasing \(x\). It indicates that the Kondo temperature of this system is almost constant with increasing \(x\) in spite of the sudden decrease of the Néel temperature. The Kondo temperature is estimated to be less than 5 K from the linewidths of the quasi-elastic scattering.

As seen in table 1, the CEF excitation energies obtained from the susceptibilities by using the least squares fit procedure are qualitatively in agreement with those determined by the neutron inelastic scattering experiments. This indicates that 4f electrons in this system are well localized and the susceptibility is explained rather well by the crystal field model.

From the result of the present experiments, we suggest that the suppression of the ordering temperature in Ce\(\text{Zn}_{1-x}\text{Cu}_x\) is mainly caused by the competition between different kind exchange interactions or between different magnetic anisotropy due to the substitution of Zn atom by Cu atom rather than the competition between the Kondo effect and the RKKY interaction as expected earlier. Since there is no sign of the increasing of the Kondo temperature with increasing \(x\). Thus, the broadening of the linewidth of the CEF scattering is thought to be a result of disorder effect caused by the random substitution of Zn atom by Cu atom. However, it is still possible that the Kondo effect exists at a certain amount in this system. Since the total exchange energy becomes small around \(x = 0.2\), the competition between the Kondo effect and the RKKY interaction will be significant in the compound Ce\(\text{Zn}_{1-x}\text{Cu}_x\) with \(x\) around 0.2.

Acknowledgements

We would like to thank T. Kamiyama and K. Shibata for assistance with the experiments at the LAM-D spectrometer and N. Sato for helping the susceptibility measurements with the 3He cryostat.

References

Table 1. Concentration dependence of the CEF excitation energy from the ground state to the first excited state. \( E_{\text{Ol}}(\text{neutron}) \) is determined by the neutron inelastic scattering experiments. \( E_{\text{Ol}}(\text{sus.}) \) is obtained from the fit to the susceptibilities.

<table>
<thead>
<tr>
<th>( x )</th>
<th>( 0.0 )</th>
<th>( 0.10 )</th>
<th>( 0.15 )</th>
<th>( 0.20 )</th>
<th>( 0.25 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( E_{\text{Ol}}(\text{neutron}) )</td>
<td>( 16.1 \pm 0.4 )</td>
<td>( 12.5 \pm 0.5 )</td>
<td>( 10.7 \pm 0.7 )</td>
<td>( 9.1 \pm 0.8 )</td>
<td>( 7.0 \pm 1 )</td>
</tr>
<tr>
<td>( E_{\text{Ol}}(\text{sus.}) )</td>
<td>( 12.5 \dagger )</td>
<td>10.3</td>
<td>4.9</td>
<td>5.6</td>
<td>3.4</td>
</tr>
</tbody>
</table>

\( \dagger \) From ref. [2] (unit: meV)

Figure Captions

**Fig. 1** Temperature dependence of the reciprocal susceptibilities of the compounds Ce(Zn\(_{1-x}\)Cu\(_x\))\(_2\) with (a) \( x = 0.1 \), (b) \( x = 0.15 \), (c) \( x = 0.20 \) and (d) \( x = 0.25 \). The solid lines indicate the results of least squares fit to the data using the CEF model.

**Fig. 2** Neutron quasi-elastic spectra of the compound Ce(Zn\(_{1-x}\)Cu\(_x\))\(_2\) with (a) \( x = 0.0 \), (b) \( x = 0.1 \), (c) \( x = 0.15 \) and (d) \( x = 0.25 \) observed with the LAM-D spectrometer. The lines are the results of least squares fit.

**Fig. 3** Concentration dependence of the line widths of the CEF scattering and the quasi-elastic scattering, and the Néel temperature.

---

Fig. 1
Fig. 2

Fig. 3