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# Anomalous Zn- and Ni-substitution effects on superconductivity in the superconducting weak ferromagnets $RuSr_2RCu_2O_8$ (R = Gd, Eu)

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#### ABSTRACT

The effect of magnetic Ni and non-magnetic Zn impurities on superconducting transition temperature  $T_c$  in RuSr<sub>2</sub>R(Cu<sub>1-x</sub>(Ni, Zn)<sub>x</sub>)<sub>2</sub>O<sub>8</sub> with R = Gd or Eu (Ni- and Zn-substituted Ru1212Gd(Eu)) was extensively studied. It is found that the suppression rate  $dT_c/dx$  of RuSr<sub>2</sub>R(Cu<sub>1-x</sub>(Ni, Zn)<sub>x</sub>)<sub>2</sub>O<sub>8</sub> is comparable to that of underdoped YBa<sub>2</sub>(Cu<sub>1-x</sub>(Ni, Zn)<sub>x</sub>)<sub>3</sub>O<sub>7- $\delta$ </sub>. The suppression of superconductivity in Ni-substituted Ru1212Eu samples is more significant than that in Zn-substituted ones, indicative of Ni being a more effective pair-breaker than Zn. In strong contrast, the magnetic Ni impurity atoms have a weaker effect on superconductivity than non-magnetic Zn atoms in Ru1212Gd, similar to what was observed in the high- $T_c$  cuprates. These intriguing findings strongly suggest that the impurity-induced local disturbance of the 3*d*-spin correlation at Cu sites around Ni/Zn is distinctly different between Ru1212Gd and Ru1212Eu.

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#### 1. Introduction

Coexistence of superconductivity and weak ferromagnetism (WFM) has been reported in ruthenocuprates RuSr<sub>2</sub>ReCu<sub>2</sub>O<sub>8</sub> (Ru-1212) with Re = Gd, Eu, Y, and Sm [1,2]. The structure of Ru-1212 is similar to that of YBa<sub>2</sub>CuO<sub>3</sub> with replacement of the Cu-O chains by the RuO<sub>2</sub> planes. The RuO<sub>6</sub> octahedra share apical oxygens ( $O_{api-1}$ ) <sub>cal</sub>) with two layers of CuO<sub>5</sub> square pyramids. It is generally believed that superconductivity originates from the CuO<sub>2</sub> planes and magnetism is associated with Ru moments in the RuO<sub>2</sub> planes. The Ru-1212 provides an unprecedented opportunity to study how the superconducting CuO<sub>2</sub> bilayer coupling propagates through the magnetic RuO<sub>2</sub> layers. Neutron experiments have shown that the magnetic transition involving the Ru sites causes a structural response in the CuO<sub>2</sub> planes, suggesting a substantial hybridization between the Ru- $t_{2g}$  and the Cu- $3d_{x-y}^2$  orbitals [3]. The RuO<sub>2</sub> planes act as a charge reservoir which dopes holes into the  $CuO_2$ planes via charge transfer arising from overlap of the  $Ru-t_{2g}$  and the Cu-3 $d_x^2 q_y^2$  orbitals through O<sub>apical</sub>. Extended X-ray absorption fine structure reveals that the temperature dependence of the Debye-Waller factor of the Cu-O<sub>apical</sub> exhibits a peak feature, different from what was expected for thermal disorder, near magnetic ordering temperature of 135 K, indicative of a magnetoelastic coupling [4]. In spite of numerous investigations, the effect of in-plane impurity substitution on superconductivity in Ru-1212 to date is still lacking. In this work, we will show that Ni(Zn) is a more effective pair-breaker than Zn(Ni) for  $RuSr_2EuCu_2O_8$  and  $RuSr_2GdCu_2O_8$ , respectively. This finding strongly suggests that the impurity-induced local disturbance of the 3*d*-spin correlation at Cu sites around Ni/Zn is distinctly different between Ru1212Gd and Ru1212Eu.

#### 2. Experimental

The polycrystalline samples of  $\text{RuSr}_2\text{R}(\text{Cu}_{1-x}M_x)_2\text{O}_8$  (R = Gd, Eu and M = Zn, Ni) were prepared by the solid-state reaction method. Detailed synthesis processes can be found elsewhere [5]. X-ray powder diffraction investigations indicate that the samples studied are single-phase compounds with tetragonal *P4/mbm* structure. The resistivity measurements were performed in a quantum design physical properties measurement system (PPMS).

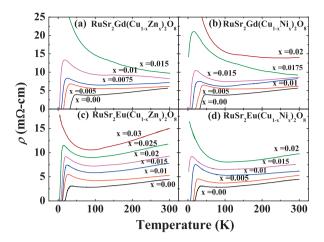
#### 3. Results and discussion

The temperature dependence of the resistivity for RuSr<sub>2</sub>Gd-(Cu<sub>1-x</sub>M<sub>x</sub>)<sub>2</sub>O<sub>8</sub> and RuSr<sub>2</sub>Eu(Cu<sub>1-x</sub>M<sub>x</sub>)<sub>2</sub>O<sub>8</sub> with M = Zn or Ni is displayed in Fig. 1a–d. It is found that  $T_c$  decreases and normal-state resistivity increases with increasing impurity content. An upturn in  $\rho(T)$  is observed at low temperatures for lower- $T_c$  samples. This behavior is likely due to a combination of single impurity scattering and localization effects [6]. More interestingly, the superconducting transition width,  $\delta T_c$ , is insensitive to impurity content, indicating that the broad  $\delta T_c$  (~15 K) for Ru1212Gd and Ru1212Eu

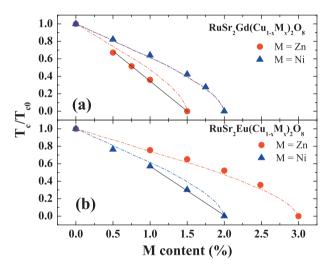




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**Fig. 1.**  $\rho$  versus *T* for (a and b) RuSr<sub>2</sub>Gd(Cu<sub>1-x</sub>M<sub>x</sub>)<sub>2</sub>O<sub>8</sub> and (c and d) RuSr<sub>2</sub>Eu-(Cu<sub>1-x</sub>M<sub>x</sub>)<sub>2</sub>O<sub>8</sub> with M = Zn or Ni.



**Fig. 2.**  $T_c/T_{c0}$  versus impurity content for (a) RuSr<sub>2</sub>Gd(Cu<sub>1-x</sub>M<sub>x</sub>)<sub>2</sub>O<sub>8</sub> and (b) RuSr<sub>2</sub>Eu(Cu<sub>1-x</sub>M<sub>x</sub>)<sub>2</sub>O<sub>8</sub> with M = Zn or Ni. The dashed curves are obtained by fitting data to A–G formula.

is intrinsically governed by a spontaneous vortex state associated with a net magnetic moment of  ${\sim}0.1~\mu_B$  per Ru [7].

To have a better understanding on the impurity-induced  $T_c$  suppression in samples studied, the normalized superconducting transition temperature  $T_c/T_{c0}$  as a function of impurity content for Ru1212Gd and Ru1212Eu is plotted in Fig. 2a and b, respectively.  $T_c$  is determined from the point on the  $\rho(T)$  curve where resistivity starts decreasing dramatically. It appears that the suppression rate  $dT_c/dx$  of RuSr<sub>2</sub>*R*(Cu<sub>1-x</sub>(Ni, Zn)<sub>x</sub>)<sub>2</sub>O<sub>8</sub> is comparable to that of underdoped  $YBa_2(Cu_{1-x}(Ni, Zn)_x)_3O_{7-\delta}$ . This provides a supportive evidence for  $RuSr_2RCu_2O_8$  (R = Gd, Eu) being underdoped superconductors determined from transport measurements [8]. Furthermore, a linear variation of  $T_c$  with impurity content observed in the higher doping regime for  $RuSr_2Gd(Cu_{1-x}Zn_x)_2O_8$  and RuSr<sub>2</sub>Eu(Cu<sub>1-x</sub>Ni<sub>x</sub>)<sub>2</sub>O<sub>8</sub> slightly deviates from the theoretical prediction from AG pair-breaking theory [9] as displayed by dashed curves in Fig. 2. It has been argued that  $T_c$  could be dominated by phase fluctuations of the order parameter for low superfluid density (underdoped) superconductors [10]. A reasonable explanation of the striking feature would then be a crossover from the pairbreaking to the phase fluctuation regime in the presence of higher disorder as impurity content increases.

In strong contrast to what was observed in Ru1212Gd and other high- $T_c$  cuprates,  $T_c$  suppression in Ni-substituted Ru1212Eu is more significant than that in Zn-substituted Ru1212Eu, indicative of Ni being a more effective pair-breaker than Zn. A possible scenario is that the electronic structure of quasi-two-dimensional CuO<sub>2</sub> layer for Ni- and Zn-substituted Ru1212Eu, in addition to the depairing effect by the potential scattering, is also modulated in a subtle way through the magnetoelastic coupling. More investigations are needed to clarify the intriguing impurity-induced  $T_c$ suppression in RuSr<sub>2</sub>RCu<sub>2</sub>O<sub>8</sub> (R = Gd, Eu).

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