

Enhanced Magnetic Properties and Spin-Phonon Coupling in Ni-Substituted α - $\text{Cu}_2\text{V}_2\text{O}_7$: The Role of Bond Length, Bond Angles, and Distorted Polyhedral Structure

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Table S1: Lattice parameters, bond lengths and bond angles for $\text{Cu}_{2-x}\text{Ni}_x\text{V}_2\text{O}_7$ ($x=0.0, 0.05$, and 0.1)

Samples	$\text{Cu}_2\text{V}_2\text{O}_7$	$\text{Cu}_{1.95}\text{Ni}_{0.05}\text{V}_2\text{O}_7$	$\text{Cu}_{1.90}\text{Ni}_{0.1}\text{V}_2\text{O}_7$
a(Å)	20.666(0)	20.663(1)	20.661(3)
b(Å)	8.398(9)	8.394(3)	8.389(2)
c(Å)	6.443(4)	6.446(1)	6.450(1)
V(Å ³)	1118.39(7)	1118.10(8)	1117.97(7)
χ^2	1.41	2.90	1.93
R _p	5.71%	3.96%	4.36%
WR _p	6.74%	5.43%	5.92%
Cu-O ₁ (Å)	1.884(4)	1.880(1)	1.948(8)
Cu-O ₂ (Å)	1.963(8)	1.950(7)	1.948(8)
Cu-O ₃ (Å)	1.963(8)	1.950(7)	1.948(8)
Cu-O ₄ (Å)	2.015(2)	1.981(5)	1.987(6)
Cu-O ₅ (Å)	2.465(1)	2.500(2)	2.501(1)
<Cu-O ₁ -Cu> [degree]	91.15	90.15	89.10

<Cu-O ₃ -Cu> [degree]	104.22	107.23	106.15
<V-O ₄ -V> [degree]	142.34	139.33	138.22
V-O ₁ (Å)	1.607(1)	1.621(2)	1.642(3)
V-O ₂ (Å)	1.721(2)	1.704(1)	1.717(2)
V-O ₃ (Å)	1.769(1)	1.787(0)	1.717(1)
V-O ₄ (Å)	1.769(0)	1.787(0)	1.799(2)

Table S2. Bond length, coordination number, disorder factor and R-factor obtained by EXAFS fitting for Ni-doped Cu₂V₂O₇ for Cu, Ni K-edges at 15K and 300K

Cu K-edge at 15K:

Path	Parameter	Cu ₂ V ₂ O ₇	Cu _{1.95} Ni _{0.05} V ₂ O ₇	Cu _{1.90} Ni _{0.1} V ₂ O ₇
Cu-O1	R(Å)	1.80 ±0.02	1.79±0.04	1.87±0.03
	N	1	1	3
	σ ² (Å ²)	0.0026±0.001	0.0046±0.001	0.0015±0.001
	R-factor	0.0037	0.0043	0.0038
Cu-O2	R(Å)	1.92 ±0.03	1.94±0.04	1.98±0.04
	N	2	2	1
	σ ² (Å ²)	0.0024±0.001	0.0026±0.001	0.0023±0.001
	R-factor	0.0037	0.0043	0.0038
Cu-O3	R(Å)	2.02 ±0.03	2.01±0.04	
	N	1	1	
	σ ² (Å ²)	0.0022±0.001	0.0030±0.001	
	R-factor	0.0037	0.0043	

Cu-O5	R(Å)	2.61 ±0.04	2.50±0.05	2.55±0.04
	N	1	1	1
	$\sigma^2(\text{\AA}^2)$	0.0011±0.001	0.0012±0.001	0.0013±0.001
	R-factor	0.0037	0.0043	0.0038
Cu-Cu	R(Å)	3.16 ±0.02	3.16±0.04	3.15±0.01
	N	2	2	2
	$\sigma^2(\text{\AA}^2)$	0.0109±0.001	0.0148±0.001	0.0169±0.001
	R-factor	0.0037	0.0043	0.0038

Ni K-edge at 15K:

Path	Parameter	$\text{Cu}_{1.95}\text{Ni}_{0.05}\text{V}_2\text{O}_7$	$\text{Cu}_{1.90}\text{Ni}_{0.1}\text{V}_2\text{O}_7$
Cu/Ni-O1	R(Å)	1.82±0.04	1.91±0.01
	N	1	3
	$\sigma^2(\text{\AA}^2)$	0.0015±0.001	0.0010±0.001
	R-factor	0.0020	0.0034
Cu/Ni-O2	R(Å)	1.96±0.04	1.99±0.04
	N	2	1
	$\sigma^2(\text{\AA}^2)$	0.0010±0.001	0.0023±0.001
	R-factor	0.0020	0.0034
Cu/Ni-O3	R(Å)	2.24±0.04	
	N	1	
	$\sigma^2(\text{\AA}^2)$	0.0046±0.001	
	R-factor	0.0020	
Cu/Ni-O5	R(Å)	2.50±0.05	2.52±0.04
	N	1	1
	$\sigma^2(\text{\AA}^2)$	0.0066±0.001	0.0010±0.001

R-factor	0.0020	0.0034
Cu/Ni-Cu/Ni R($\text{\AA}$)	3.16 $\pm$ 0.04	3.15 $\pm$ 0.01
N	2	2
$\sigma^2(\text{\AA}^2)$	0.0243 $\pm$ 0.001	0.0252 $\pm$ 0.001
R-factor	0.0020	0.0034

Cu K-edge at 300K:

Path	Parameter	$\text{Cu}_2\text{V}_2\text{O}_7$	$\text{Cu}_{1.95}\text{Ni}_{0.05}\text{V}_2\text{O}_7$	$\text{Cu}_{1.90}\text{Ni}_{0.1}\text{V}_2\text{O}_7$
Cu-O1	R($\text{\AA}$)	1.87 $\pm$ 0.01	1.88 $\pm$ 0.02	1.93 $\pm$ 0.01
	N	1	1	3
	$\sigma^2(\text{\AA}^2)$	0.0053 $\pm$ 0.001	0.0011 $\pm$ 0.001	0.0013 $\pm$ 0.001
	R-factor	0.0031	0.0021	0.0084
Cu-O2	R($\text{\AA}$)	1.97 $\pm$ 0.01	1.95 $\pm$ 0.04	1.98 $\pm$ 0.03
	N	2	2	1
	$\sigma^2(\text{\AA}^2)$	0.0014 $\pm$ 0.001	0.0092 $\pm$ 0.001	0.0018 $\pm$ 0.001
	R-factor	0.0031	0.0021	0.0084
Cu-O3	R($\text{\AA}$)	2.02 $\pm$ 0.01	2.01 $\pm$ 0.04	
	N	1	1	
	$\sigma^2(\text{\AA}^2)$	0.0023 $\pm$ 0.001	0.0031 $\pm$ 0.001	
	R-factor	0.0031	0.0021	

Cu-O5	R(Å)	2.60 ±0.03	2.59±0.05	2.52±0.05
	N	1	1	1
	$\sigma^2(\text{Å}^2)$	0.0024±0.001	0.0023±0.001	0.0076±0.001
	R-factor	0.0031	0.0021	0.0084
Cu-Cu	R(Å)	3.17 ±0.03	3.18±0.05	3.09±0.04
	N	2	2	2
	$\sigma^2(\text{Å}^2)$	0.0122±0.001	0.0064±0.001	0.0021±0.001
	R-factor	0.0031	0.0021	0.0084

Ni K-edge *from $\text{Cu}_{2-x}\text{Ni}_x\text{V}_2\text{O}_7$ ($x=0.05, 0.1$)* at 300K:

Path	Parameter	$\text{Cu}_{1.95}\text{Ni}_{0.05}\text{V}_2\text{O}_7$	$\text{Cu}_{1.90}\text{Ni}_{0.1}\text{V}_2\text{O}_7$
Cu/Ni-O1	R(Å)	1.89±0.04	1.93±0.01
	N	1	3
	$\sigma^2(\text{Å}^2)$	0.0022±0.001	0.0015±0.001
	R-factor	0.0020	0.0034
Cu/Ni-O2	R(Å)	1.98±0.04	1.98±0.04
	N	2	1

	$\sigma^2(\text{\AA}^2)$	0.0010±0.001	0.0023±0.001
	R-factor	0.0020	0.0034
Cu/Ni-O3	R($\text{\AA}$)	2.01±0.04	
	N	1	
	$\sigma^2(\text{\AA}^2)$	0.0027±0.001	
	R-factor	0.0020	
Cu/Ni-O5	R($\text{\AA}$)	2.52±0.05	2.56±0.04
	N	1	1
	$\sigma^2(\text{\AA}^2)$	0.0066±0.001	0.0010±0.001
	R-factor	0.0020	0.0034
Cu/Ni-Cu/Ni	R($\text{\AA}$)	3.17±0.04	3.11±0.01
	N	2	2
	$\sigma^2(\text{\AA}^2)$	0.0243±0.001	0.0252±0.001
	R-factor	0.0020	0.0034

Ni K-edge *from standard sample NiO* at 300K:

Path	Parameter	NiO
Ni1-O1	R($\text{\AA}$)	2.06±0.02
	N	6
	$\sigma^2(\text{\AA}^2)$	0.0021±0.001
	R-factor	0.0022
Ni1-Ni2	R($\text{\AA}$)	2.93±0.02

	N	12
	$\sigma^2(\text{\AA}^2)$	0.0030±0.001
	R-factor	0.0022
Ni1-O3	R($\text{\AA}$)	3.58±0.03
	N	8
	$\sigma^2(\text{\AA}^2)$	0.0034±0.001
	R-factor	0.0022

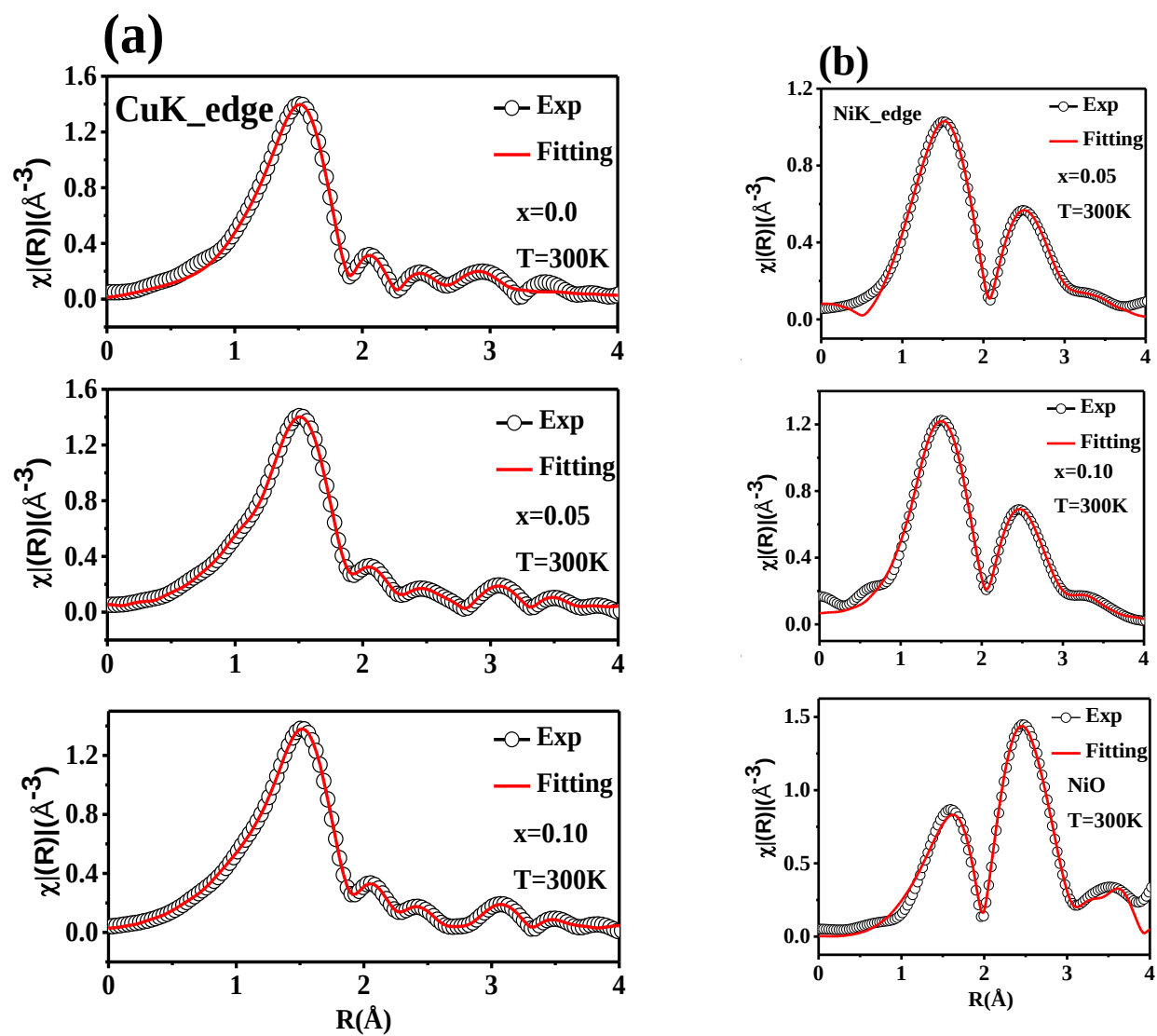


Figure S1: (a) Describes the modulus of Fourier-transformed EXAFS against radial distance at the Cu-k edge of $\text{Cu}_{2-x}\text{Ni}_x\text{V}_2\text{O}_7$ samples and (b) describes the same plot at Ni K-edge of respective samples. The solid red line is the theoretical fitting of $\chi(R)$ vs. R pattern.

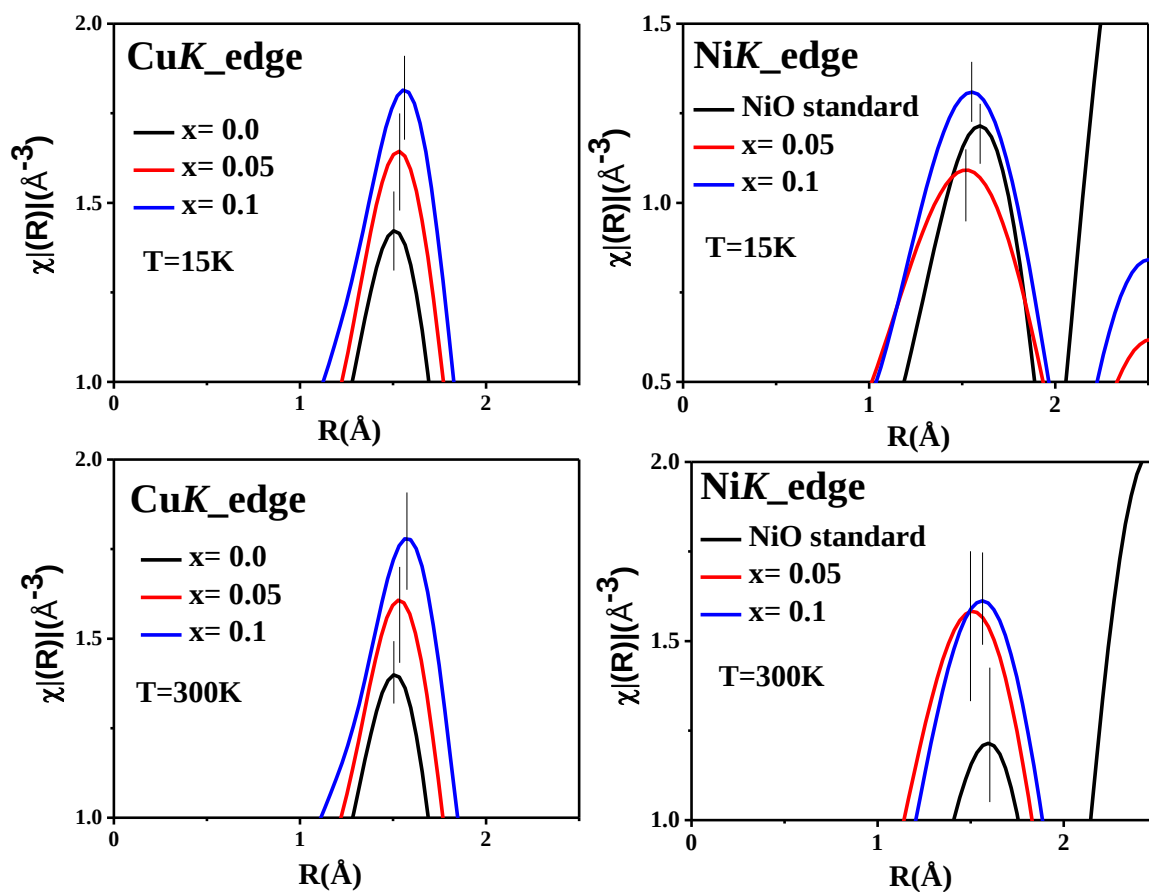


Figure S2: Describes the modulus of Fourier-transformed EXAFS against radial distance for the first shell at the Cu *K*-edge and Ni *K*-edge of $\text{Cu}_{2-x}\text{Ni}_x\text{V}_2\text{O}_7$ samples at 15 and 300 K.