Observation of Cu spin fluctuations in over-doped regime $La_{2\text{-}x}Sr_xCuO_4$ nanoparticles

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The first observation of exchange anisotropy or exchange bias was reported for cobalt particles capped by cobalt oxide.[1)] The shifted hysteresis loop in field-cooled suggested the occurrence of exchange coupling between the ferromagnetic spin of cobalt with the antiferromagnetic spin of cobalt oxide. For gold nanoparticles encapsulated by butanethiol, clear evidence of magnetism was observed when the size of the gold cluster was 2.2 nm, as studied by muon-spin relaxation (μSR).[2)] The relaxation of muon spins decreased with increasing temperature in the longitudinal field, (LF-μSR), indicating the existence of a magnetic moment. Even though magnetism was clearly observed, this study could not distinguish whether the magnetic moment was located at the encapsulated molecules or inside the gold nanoparticles.

The existence of ferromagnetic spins at the surface was also suggested to lead to the observation of a hysteresis loop at 10 K in CuO nanoparticles.[3)] An anomalously enhanced Curie term at low temperatures in high-T_c superconductor cuprate (HTSC), $La_{2\text{-}x}Sr_xCuO_4$, with $x =$ 0.10–0.30, when the particle size was reduced to 113 nm was also linked to the role of the spins at the surface.[4)] This suggests that the surface effect plays an important role in both free-standing nanoparticles, like CuO and $La_{2\text{-}x}Sr_xCuO_4$, and nanoparticles capped by molecules, like oxide-coated particles of cobalt and nanogold capped by butanethiol. However, the mechanism of the role of the surface causing the emergence of ferromagnetism in nanoparticles is still an open question.

We aimed to investigate nano-sizing effects in $La_{2\text{-}x}Sr_xCuO_4$ because the observation of possible ferromagnetism is a new interesting phenomenon in HTSCs. In the bulk case, a ferromagnetic phase was predicted in the heavily overdoped regime where the superconductance was suppressed.[5,6)] This appearance of ferromagnetism in $La_{2\text{-}x}Sr_xCuO_4$ from the underdoped to the overdoped regime is a new question in HTSCs because superconductivity and magnetic ordering are believed to be interconnected.

To synthesize free-standing nanoparticles, the chemical reaction method is believed as one of the best methods because it is a bottom-up method.[7)] A bottom-up method implies spontaneous self-assembly from the atomic level to the nanoparticle level. The detailed synthesis route is reported in our former paper.[8)]

μSR measurements on $La_{2\text{-}x}Sr_xCuO_4$ with $x = 0.20$ and particle size of 46 nm were performed at the RIKEN-RAL Muon Facility, Rutherford-Appleton Laboratory, UK, using a pulsed positive surface muon beam. Figure 1 displays the temperature dependence of the relaxation rate of the muon spins. The relaxation rate starts rapidly increasing below 10 K. This indicates the slowing down of the Cu spin fluctuations below 10 K, which, in turn, suggests that magnetic correlations are developed at low temperatures. Compared to the bulk case, no magnetic correlation was observed even at 0.3 K.[9)]

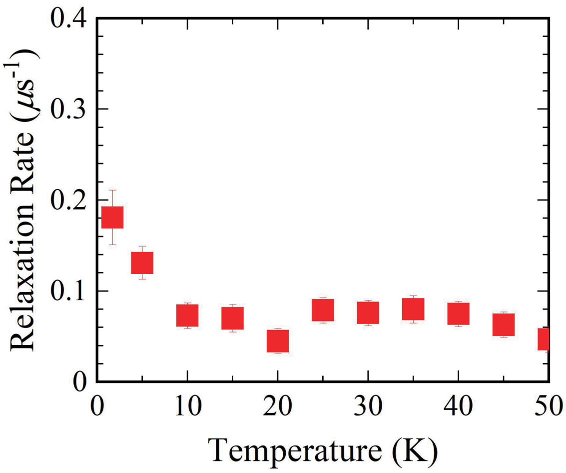

Fig. 1. Temperature dependence of relaxation rate of muon spins, λ, of $La_{2\text{-}x}Sr_xCuO_4$ with $x = 0.20$ and particle size of 46 nm.

Our present result reveals that nano-sizing induces weak magnetism owing to the development of Cu spin fluctuation. Moreover, our magnetization results show that the superconductivity is strongly suppressed in $La_{2\text{-}x}Sr_xCuO_4$ nanoparticles. These results suggest an anticorrelation between the magnetic order and the superconductivity and show that the superconductivity in an HTSC is strongly influenced by the magnetic order, which can be explained by the stripe theory. We provided a new insight of studying Cu spin dynamics by investigating the nano-sizing effects in an HTSC, which is typically examined based on the impurity effects on the Cu site.[9–11)]

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References

1) W. H. Meiklejohn, C. P. Bean, Phys. Rev. **102**, 1413 (1956).
2) M. H. Dehn *et al.*, Appl. Phys. Lett. **112**, 053105 (2018).
3) M. S. Seehra, A. Punnoose, Solid State Commun. **128**, 299 (2003).
4) Y. Yin *et al.*, J. Phys. Chem. C **117**, 3028 (2013).
5) A. Kopp *et al.*, Proc. Natl. Acad. Sci. USA **104**, 6123 (2007).
6) J. E. Sonier *et al.*, Proc. Natl. Acad. Sci. USA **107**, 17131 (2010).
7) A. G. Kolhatkar *et al.*, Int. J. Mol. Sci. **14**, 15977 (2013).
8) S. Winarsih *et al.*, Key. Eng. Mater. **966**, 357 (2019).
9) T. Adachi *et al.*, Phys. Rev. B **78**, 134515 (2008).
10) T. Adachi, *et al.*, Phys. Rev B **70**, 060504(R) (2004).
11) Risdiana, *et al.*, Phys. Rev. B **77**, 054516 (2008).