実験報告書様式（一般利用課題・成果公開利用）

（ ※本報告書は英語で記述してください。ただし、産業利用課題として採択されている方は日本語で記述していただいても結構です。 ）

| # MLF Experimental Report | 提出日 Date of Report |
|---|---|
| 課題番号 Project No.<br>2016A0035<br>実験課題名 Title of experiment<br>Magnetism and superconductivity in hyperexpanded fullerides close to the Mott transition<br>実験責任者名 Name of principal investigator<br>Kosmas Prassides<br>所属 Affiliation<br>WPI-Advanced Institute for Materials Research (WPI-AIMR), Tohoku University | 装置責任者 Name of responsible person<br>Yasuhiro Miyake<br>装置名 Name of Instrument/（BL No.）<br>D1 Instrument<br>実施日 Date of Experiment<br>2017//01/26 – 2017/01/28 |

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。（適宜、図表添付のこと）
Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

| 1. 試料 Name of sample(s) and chemical formula, or compositions including physical form. |
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| Rubidium caesium fulleride, $Rb_{0.75}Cs_{2.75}C_{60}$ |

| 2. 実験方法及び結果 （実験がうまくいかなかった場合、その理由を記述してください。）<br>Experimental method and results. If you failed to conduct experiment as planned, please describe reasons. |
|---|
| The electronic phase diagram of cubic alkali fullerides, $A_xA'_{3-x}C_{60}$ (A, A'= alkali metals) exhibits striking similarities with those of unconventional superconductors such as the cuprates. Superconductivity emerges from an antiferromagnetic strongly correlated Mott insulating state through bandwidth control *via* outer wavefunction overlap of the constituent molecules and is accompanied by a dome-shaped dependence of the critical temperature, $T_c$. This is experimentally achieved by tuning a parameter such as physical (through application of external pressure) or chemical (through substitution of ions with varying size) pressure. Moreover, we have also established that such bandwidth control transforms the Mott insulating state first into an unconventional correlated Jahn-Teller (JT) metal (where localized electrons coexist with metallicity and on-molecule Jahn-teller distortions persist), and then into a Fermi liquid with a less prominent molecular electronic signature. This normal state crossover is mirrored in the evolution of the superconducting state, with the highest $T_c$ found at the boundary between unconventional correlated and conventional weak-coupling BCS superconductivity where the interplay between extended and molecular aspects of the electronic structure is optimized to create the superconductivity dome [**SCIENCE ADVANCES** *1*, e1500059 (2015)]. |

## 2. 実験方法及び結果（つづき） Experimental method and results (continued)

In this experimental period, we attempted to use the $\mu^+$SR technique in its zero-field (ZF) variant to study the electronic ground states as we move across the complex bandwidth-controlled phase diagram. We attempted to probe more deeply the Mott-Jahn-Teller insulator to Jahn-Teller metal crossover, which is controlled by varying bandwidth (or unit cell volume, $V$). Both our magnetization and $^{13}$C NMR measurements had shown that the Jahn-Teller metal exhibits a strongly enhanced spin susceptibility relative to that of a conventional Fermi liquid, characteristic of the importance of strong electron correlations. In the insulating phase, $1/^{13}T_1$ is governed by antiferromagnetic spin fluctuations – such fluctuations remain important in the Jahn-Teller metal regime but gradually diminish with decreasing $V$ as $(U/W)$ decreases and conventional Fermi liquid behavior appears for underexpanded fullerides.

ZF-$\mu^+$SR data of a representative overexpanded fulleride with composition $Rb_{0.75}Cs_{2.25}C_{60}$ and a superconducting $T_c$ = 31.4 K were collected in the temperature range 5 to 240 K. $Rb_{0.75}Cs_{2.25}C_{60}$ is an insulator at high temperature but there is a crossover to the Jahn-Teller metal state at ~130 K as established by complementary magnetization, NMR and IR measurements. In Fig. 1, we show the ZF time-dependent $\mu^+$SR spectra of $Rb_{0.75}Cs_{2.25}C_{60}$ at selected temperatures. At high temperatures, the spectra are characteristic of the presence of weak static nuclear dipole moments, which result in a small depolarization rate, $\sigma \sim 0.04\ \mu s^{-1}$, together with a very slow relaxation, arising from fluctuating electronic moments. Cooling down below 190 K leads to the emergence of a second relaxing component whose relaxation rate, $\lambda$ increases rapidly and saturates near the onset of the insulator-to-metal crossover at ~130 K. A second increase in the relaxation rate of the dominant component now also occurs at the metal-to-superconductor transition on further cooling. Fig. 2 shows the temperature dependence of the relaxation rate as we cross the boundaries between the high-temperature insulating state towards the anomalous Jahn-Teller metallic state and finally, the superconducting state. The significance of these preliminary results are under further consideration but they already provide good evidence for the emergence of frozen electronic moments as the phase diagram boundary from the Mott-Jahn-Teller insulator to the Jahn-Teller metal is crossed on cooling.

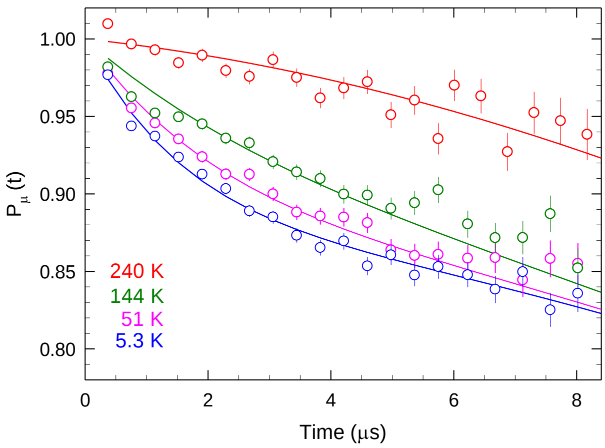


Fig. 1: Temperature evolution of the ZF $\mu^+$-spin polarization, $P_\mu(t)$ for $Rb_{0.75}Cs_{2.25}C_{60}$ at 5.3, 51, 144, and 240 K. The solid lines represent the fits to the model function employed.

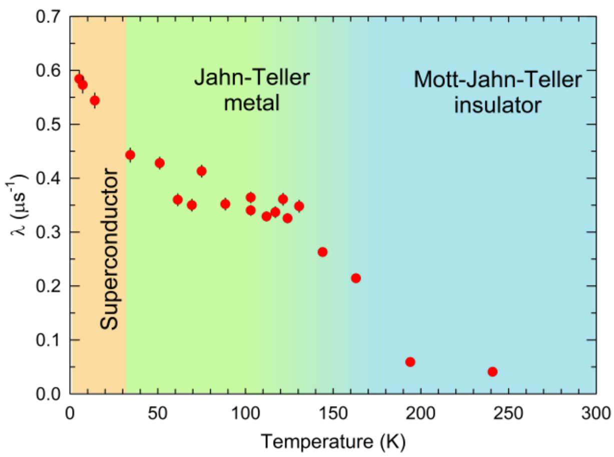


Fig. 2: Temperature dependence of the ZF relaxation rate, $\lambda$ in $Rb_{0.75}Cs_{2.25}C_{60}$ showing the emergence of static disordered magnetic component below 190 K which coincides with the Mott-Jahn-Teller insulator-to-Jahn-Teller metal crossover established by complementary magnetization and $^{13}$C NMR measurements.