*УДК 537.9: 537.635*

**J. Chigvinadze, V. Buntar, G. E. Zaikov,**
**O. Yu. Emelina, S. Ashimov, T. Machaidze, G. Donadze**

# INVESTIGATION OF MAGNETIC PROPERTIES OF $RB_3C_{60}$ SINGLE CRYSTAL FULLERENE USING TORQUE EXPERIMENTS

*Keywords: Magnetic state, $Rb_3C_{60}$ fullerene, superconductivity, normal state.*

*For the first time, magnetic properties of fullerides have been studied using a torque technique. A $Rb_3C_{60}$ single crystal has been investigated in these experiments. It was shown that this method is sensitive to the structural phase transition in both superconducting and normal states. In the superconducting state a transition at $T_c = 28$ K was observed and a pinning force of the magnetic vortices has been estimated. In the normal state, the transition from s.c. to f.c.c. structure has been indicated. Also a very strong magnetic response has been observed at temperatures T ~ 200 - 250 K, which may be explained in terms of the rearrangement on the magnetic system in the material.*

*Ключевые слова: Магнитное состояние, $Rb_3C_{60}$ фуллерена, сверхпроводимость, нормальное состояние.*

*Впервые, магнитные свойства фуллеридов были изучены с помощью метода крутящего момента. Монокристаллический $Rb_3C_{60}$ был исследован в этих экспериментах. Было показано, что этот метод является чувствительным к структурным фазовым переходам в обоих сверхпроводящем и нормальном состояниях. В сверхпроводящем состоянии переход наблюдался при Tc = 28 K и была оценена сила пиннинга магнитных вихрей. В нормальном состоянии, был указан переход структуры от s.c. к f.c.c. Также очень сильный магнитный отклик наблюдался при температурах T ~ 200 - 250 K, что можно объяснить с точки зрения перестройки магнитной системы в материале.*

## Introduction

The appearance of superconductivity in alkali fullerides has led to extensive efforts in attempting to understand their electronic, magnetic, structural, and dynamic properties and to elucidate the origin of their high $T_c$. In particular, the question of whether or not such a large value of $T_c$ can be caused by coupling to phonons alone is still to be answered and the answer strongly depends on the normal state properties. Despite the apparent simplicity of the structure of $A_3C_{60}$ fullerides (where A is an alkali metal), some important issues are not yet fully resolved.

It is well known that $A_3C_{60}$ fullerides, as well as pure $C_{60}$, are weak diamagnetics with the appearance of strong diamagnetism at the transition to the superconducting state. At high temperature, solid $A_3C_{60}$ forms a face-centered-cubic (f.c.c) phase. In this phase, $C_{60}$ molecules freely rotate with a reorientation time scale of the order of $10^{-11}$ seconds. With lowering temperature, the transition to s.c. structure occurs, which comes from the fast reorientation of the $C_{60}$ molecules, and an anisotropic uniaxial rotation of fullerene molecules takes place. This transition has been observed for $K_3C_{60}$ by NMR experiments [1, 2], but the exact transition temperature was hard to evaluate since, in these experiments, the peak related to this transition overlapped with a peak related to appearance of T` site and is very wide. No special effect on the magnetic properties at the temperature of the solid state transition from s.c. to f.c.c. structure has been observed.

$A_3C_{60}$ fullerides are extremely air sensitive and usually are sealed in a glass or quartz capsules. Therefore, most of the magnetic measurements on these materials have been done with SQUID magnetometers. Therefore, an alternative method of investigation of the magnetic properties may give new information about the magnetic structure of $A_3C_{60}$ superconductors in the normal state.

## Experimental Method and Samples

In these experiments we apply the mechanical torque method in order to study the magnetic properties of alkali doped fullerenes. This method has been extensively used to investigate critical parameters of superconductors such as $T_c$, critical field $H_c$, and energy dissipation in the mixed state [3, 4]. A cylindrical (in the ideal case) sample is suspended on a thin elastic thread and torque oscillations of small amplitude of $1^0$ or $2^0$ are generated with a short-time impulse. After that the sample performs free axial-torsion oscillations in an external magnetic field, $\vec{H}$, which is perpendicular to the axis of the sample. The temperature dependence of the frequency, ω (or of the period, *t*), and of the dissipation of the oscillations, δ, is measured at different magnitudes of the magnetic field.

If there are no fixed (pinned) magnetic moments in the sample, neither the dissipation nor the frequency of the oscillations depend on the external magnetic field. For example, when either i) the external magnetic field does not penetrate the substance, which is the case for a superconductor in an external field smaller than the lower critical field $H_{c1}$, or ii) the inner magnetic moments are either zero or disoriented and not fixed.

The appearance of pinned magnetic dipoles produces a nonzero magnetic moment $\vec{M}$ in the sample. The interaction between $\vec{M}$ and $\vec{H}$ makes a torque $\tau = MH \sin\alpha$, where α is the angle between $\vec{M}$ and $\vec{H}$. This additional moment τ affects the oscillating system and makes the dissipation and the frequency of oscillations dependent on the external magnetic field. The sensitivity of this method is very high, $10^{-17}$ W [4].

In our experiments we used a crystal of $Rb_3C_{60}$ that was made from a single crystal of $C_{60}$ by doping it with Rb using the method of vapor phase doping (*). Details of the sample preparation and its characterization can be found in Refs. 5 and 6 (sample R28 therein). The physical dimensions of the crystal are 3.3 x 2.7 x 1.3 $mm^3$. It consists of a few single crystalline grains of the radius about 350 μm [5]. The sample is sealed in a quartz capsule to prevent exposure of the material to air.

## Results and Discussions

In figure 1 we present temperature dependences of both period and dissipation of the oscillations of the sample monitored in the external magnetic field of 100 mT with increasing temperature from the temperature of liquid He. At temperature $T_C$ = 28 K there is a step-like transition to the normal state on both *t* and δ. This transition temperature is slightly lower than that obtained in Ref. 6 for this crystal. This difference is because the thermometer with the electrical cables cannot be fixed on the oscillating sample directly and is placed some distance from it.

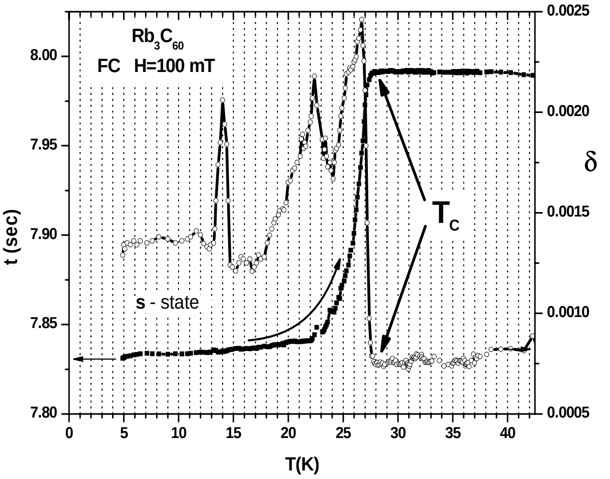

**Fig. 1 – Temperature dependence of the period (solid squares) and of the dissipation of the oscillations (open circles) of $Rb_3C_{60}$ in the superconducting state and at temperatures close to the transition, $T_C$, in the external magnetic field $\mu_0H$ = 100 mT**

It is clearly seen that the dissipation in the superconducting state is larger than that in the normal state. This is because, in the superconducting state, the material is penetrated by magnetic field in the shape of vortices. These vortices are partly pinned on structural defects and the pinning force depends on several factors like size and number of the defects, and thermal fluctuations. Vortices in the material tend to align along the external magnetic field, which increases the dissipation of the oscillations. When the material undergoes the transition to the normal state, vortices do not exist in the sample anymore and, therefore, do not prevent the oscillation of the sample.

The larger the amplitude of the oscillations, the larger is the force applied on a pinned vortex by the magnetic field. By measuring the period *t* and the dissipation δ at different amplitudes of oscillations one can find the critical amplitude $\varphi_C$ at which vortices unpin from structural defects. Using the value of $\varphi_C$ one can find the value of the critical moment $\tau_C$ and the strength of the bulk pinning force $F_p$ as [7]:

$$F_p = \frac{3}{4}\frac{\tau_c}{R^3 L};$$

where R and L are the radius and the length of the cylindrical sample respectively. The results of the estimation of the bulk pinning force at temperature T = 4.2 K are presented in figure 2.

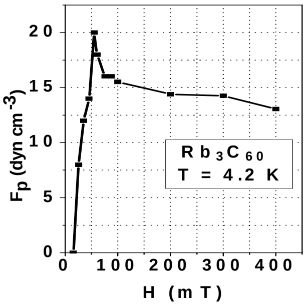

**Fig. 2 – The bulk pinning force as a function of the applied external magnetic field at T = 4.2 K**

With the increasing external magnetic field from zero, vortices penetrate the sample from its surface to the center and the bulk pinning force grows steeply. At the external field $H_{ext} = H^*$ (in our experiments $H^* \sim$ 50 mT) the sample is completely penetrated by vortices. Further increasing of the external magnetic field leads to increasing numbers of vortices, (i.e. the volume density of vortices in the sample), and therefore to the decreasing of the bulk pinning force.

The temperature dependence of the dissipation of the oscillations in the normal state is presented. At around $T_1$ ~ 270 K a peak can be observed. This peak may be associated with the first order phase transition from s.c. to f.c.c. structure. This transition is related to the freezing of libration modes and an orientation of $C_{60}$ molecules. If each $C_{60}$ molecule is considered as a diamagnetic dipole, then the orientation of molecules leads to the orientation of magnetic moments in the system and, therefore, strong response on δ(T) and ω(T). As expected, the transition temperature in $Rb_3C_{60}$ is higher than that in the pristine $C_{60}$ at T = 263 K due to the presence of Rb atoms in the interfullerene space.

There is a second peak at $T_2$ ~ 200-250 K. The amplitude of the peak of the dissipation (as shown in Fig. 3) is several orders of magnitude, which is above of the sensitivity of our experimental device. Moreover, this peak is much larger than the one at the s.c. – f.c.c. phase transition. In our experiments, the sample stops oscillating and we cannot generat the oscillations with a short-time impulse any more. We are not aware of any earlier experiments showing such strong magnetic response of fullerides materials.

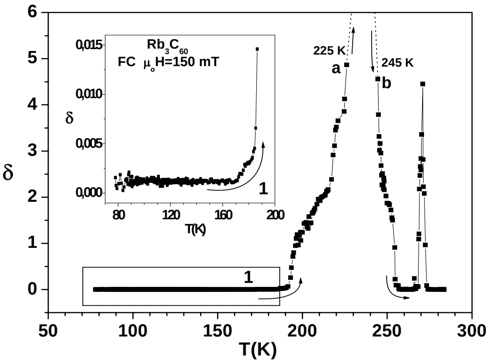

**Fig. 3 – Temperature dependence of the dissipation δ of the oscillations at the external magnetic field $\mu_0 H$ = 150 mT**

In earlier NMR experiments, some anomalies of the temperature dependence of spin-lattice relaxation times have also been observed in this range of temperatures in $K_3C_{60}$ [8, 9] and $Na_2CsC_{60}$ [10]. However, this anomaly was very weak and, as the authors reported, its amplitude was in the order of experimental error. In our experiments, the magnitude of the effect is huge. Since the effect appears to be so strong in our torque experiments, which are sensitive to the presence of fixed/oriented magnetic moments, it may mean that at this temperature there is a reorganization of the magnetic structure in the material.

In is unclear now what kind of magnetic structure is formed in $A_3C_{60}$. These magnetic moments cannot be the moments of $C_{60}$ molecules since most of the molecules are already oriented at $T_1$ = 280K and, moreover, the magnetic effect is much stronger than that at the phase transition. One may speculate that some persistent currents may appear, or we may assume that the magnetic moments involved in the crossover are due to distortions of the $C_{60}$ molecules or the $A_3C_{60}$ lattice. In general, magnetism is mainly related to properties based on electron spins. The solid lattice, though important for some of effects, usually does not provide an important contribution to magnetism. However, there is a specific type of materials for which the Jahn-Teller effect plays a very important role determining both structural and magnetic properties.

(*) Sample preparations have been done in the Institute for Material Physics, Vienna University, Vienna, Austria.

## References

1. Y. Yoshinari, H. Alloul, G. Kriza, and K. Holczer, *Phys.Rev.Lett.* **71**, 2413 (1993).
2. Y. Yoshinari, H. Alloul, V. Brouet, G. Kriza, K. Holczer, and L. Forro, *Phys. Rev, B* **54**, 6155 (1996).
3. J. Chigvinadze, *JETP,* **63**, p. 2144 (1972)
4. J. Chigvinadze, *JETP,* **65**, p. 1923 (1973)
5. V.Buntar, M.Haluska, H.Kuzmany, F.M.Sauerzopf, and H.W.Weber, *Supercond. Sci. Technol,*. **16**, 907 (2003)
6. V. Buntar, F.M. Zauerzopf, H.W. Weber, M. Halushka, and H. Kuzmany, *Phys.Rev. B* **72**, 024521 (2005).
7. M. Fuhrmans, C. Heiden, *Cryogenics* **8**, 451 (1976).
8. Y. Yoshinari, H. Alloul, G. Kriza, and K. Holczer, *Phys.Rev.Lett.* **71**, 2413 (1993).
9. Y. Yoshinari, H. Alloul, V. Brouet, G. Kriza, K. Holczer, and L. Forro, *Phys. Rev, B* **54**, 6155 (1996).
10. P. Matus, H. Alloul, G. Kriza, V. Brouet, P.M. Singer, S. Garaj, and L. Forro, *Phys. Rev. B* **74**, 214509 (2006).

---

© **J. Chigvinadze -** mean researcher, Departmet of condenced matter physics, E. Andronikashvili Institute of Physics, Tbilisi, Republic of Georgia; **V. Buntar -** Department of Physics and Astronomy, McMaster University, Hamilton, Canada; **G. E. Zaikov** - professor, Department of technology of plastic materials, Kazan National Research Technological University, chembio@sky.chph.ras.ru; **O. Yu. Emelina** - assistant, Department of technology of plastic materials, Kazan National Research Technological University; **S. Ashimov -** senior researcher of Departmet of condenced matter physics, E. Andronikashvili Institute of Physics, Tbilisi, Republic of Georgia; **T. Machaidze -** researcher of Departmet of condenced matter physics, E. Andronikashvili Institute of Physics, Tbilisi, Republic of Georgia; **G. Donadze -** researcher of Departmet of condenced matter physics, E. Andronikashvili Institute of Physics, Tbilisi, Republic of Georgia.