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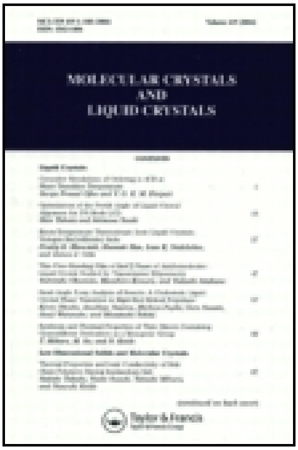

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Ferromagnetic Behaviour of TDAE-C_{60} Samples Studied with a SQUID Magnetometer

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A SQUID magnetometer has been used to study magnetic properties of TDAE-C_{60} single crystal and powder samples and another fullerene derivative APhF-Co. The measured magnetic transitions, magnetization curves and hysteresis in magnetization curves lead to the conclusion that TDAE-C_{60} behaves as a ferromagnet.

Keywords: fullerene; TDAE-C_{60}

INTRODUCTION

The discovery of the new organic magnetic material TDAE-C_{60} (TDAE stands for tetrakis dimethylamino ethylene) in 1991[1] stimulated many experiments and theoretical investigations on this material. TDAE is a strong electron donor that forms a 1:1 charge transfer compound with

C_{60}, $TDAE^+$-C_{60}^- with spin 1/2 on C_{60}^- ions.

Initial measurements of the magnetization versus external magnetic field strength $M(H)$ showed an "S" shaped characteristic but no spontaneous magnetization or a magnetic hysteresis curve was observed.[1]

Magnetic hysteresis was first reported by Suzuki *et al.*[2]. Sensitive $M(H)$ measurements were done with a SQUID magnetometer. Spontaneous magnetic order was also confirmed by the zero-field muon spin relaxation technique.[3] Tanaka *et al.*[4] tried to explain ferromagnetic properties of TDAE-C_{60} in terms of superparamagnetism where a large number of ferromagnetically correlated spins forms clusters.

The first TDAE-C_{60} single crystals were obtained in 1995[5], and magnetic studies on these samples reveal some new magnetic properties. Subsequent studies of single crystal samples have shown that TDAE-C_{60} could be described as a 3-dimensional Heisenberg ferromagnet insulator.[6]

Results of our dc SQUID magnetization measurements on TDAE-C_{60} single crystal and powder samples and on a new fullerene derivative (1-(3-aminophenyl)-1H-1,2-methano [60] fullerene cobaltocene (APhF-Co)) confirm the ferromagnetic description of magnetic properties of these samples and suggest that the essential ingredient for the ferromagnetism is the fullerene molecule rather than the electron donor molecule.

EXPERIMENTAL

All samples were prepared at J. Stefan Institute in Ljubljana: TDAE-C_{60} powder samples were synthesized according to the usual method.[1] TDAE-C_{60} single crystals were grown by the diffusion method. APhF-Co was synthesized by mixing cobaltocene in toluene solution with APhF[7]. The electron donor molecule dicyclo-pentadienyl-cobalt ($CoCp_2^+$ or cobaltocene) has spin $S = 0$ in the $CoCp_2^+$ state[8] and only electrons transferred to $APhF^-$ lead to the magnetic properties of APhF-Co.

Measurements of dc-magnetization were performed on a home built SQUID magnetometer. Magnetization has been determined from a signal

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obtained by moving a sample in an external magnetic field of strength H along the axis of superconducting detection coils in gradiometric configuration. The signal was calibrated by the common standard $CuSO_4 \cdot 5H_2O$. The measuring system allows measurements from 4.2 K till 25 K. Magnetization curves have been measured by changing the external magnetic field strength H from $0 \to H_{max} \to -H_{max} \to 0$, where H_{max} was approximately 10 000 A/m.

MEASUREMENTS AND DISCUSSION

Figure 1 shows the temperature dependence of magnetization for about 1 mm^3 large TDAE-C_{60} single crystal in a magnetic field strength H = 4 000 A/m. Above the transition temperature the magnetic signal of this sample is below the sensitivity of our measuring system. Magnetization below the transition temperature has been detected only after the sample was thermally annealed. Several newly grown single crystals samples did not show any magnetic signal before thermal annealing procedure which is in agreement with previously reported results.[9]

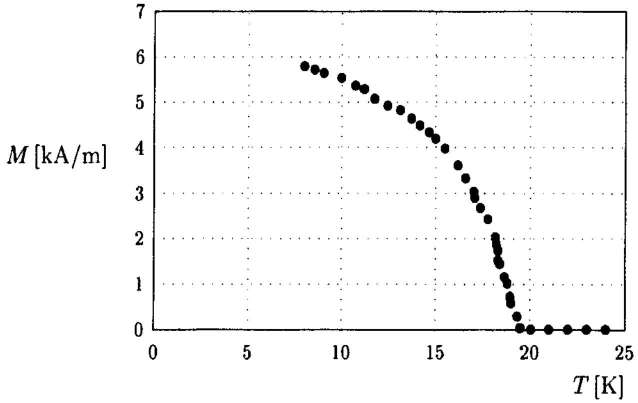

Figure 1 $M(T)$ dependence in TDAE-C_{60} single crystal sample in H = 4 000 A/m.

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The $M(H)$ dependence at 4.2 K is shown in Figure 2. The magnetization reaches saturated value of about 6 500 A/m at the magnetic field strenght $H = 3\,000$ A/m. A calculated maximal magnetization of TDAE-C_{60} single crystal sample, considering a monoclinic unit cell with $a = 15.858$ Å, $b = 12.998$ Å, $c = 19.987$ Å and $\beta = 93,37°$[5] with four free electrons per unit cell, is about 9 000 A/m. The measured value of the saturated magnetization is more than 70% of the calculated one.

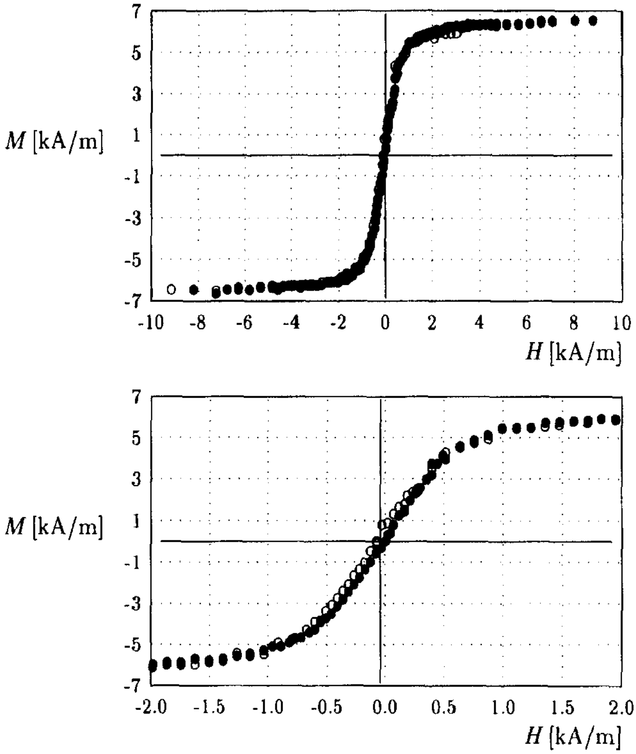

Figure 2 $M(H)$ dependence in TDAE-C_{60} single crystal at 4.2 K. Figure 2 b) shows the same measured data points as Figure 2 a) but only for small magnetic field strength H to make the hysteresis visible. ● and ○ are for increasing and decreasing H, respectively.

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A hysteresis loop in the $M(H)$ dependence has been also observed, as shown in Figure 2 b). The remanent magnetization is about 300 A/m.

Next we compare the magnetic properties (transition temperature and $M(H)$ dependencies at several temperatures below the transition temperature) between the slowly cooled sample and quenched sample. The cooling rates were about 2 K/min and 300 K/min from room temperature down to 4.2 K for slowly cooled and quenched samples respectively. We did not observe any shift of the transition temperature or changes in $M(H)$ dependencies for either cooling rate.

Similar measurements have been carried out on TDAE-C_{60} powder samples. Figure 3 shows the $M(H)$ dependence of TDAE-C_{60} powder sample at 4.2 K. The x-axis scale (the strength of the magnetic field H) is same as on the Figure 2. a), while the y-axis scale (the magnetization M) is much smaller than the magnetization of single crystal samples in the same external magnetic field.

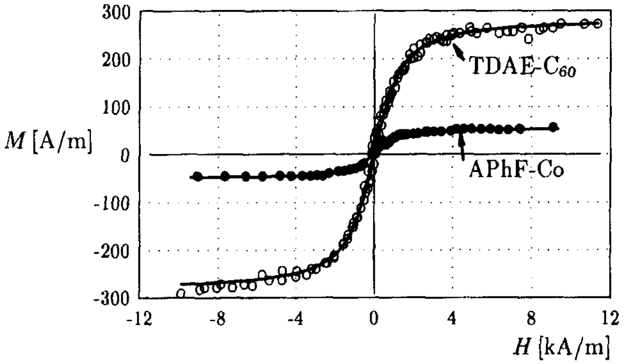

Figure 3 $M(H)$ signal in powder TDAE-C_{60} (○) and in APhF-Co (●) sample at 4.2 K. Full lines are $M(H)$ dependencies calculated from the model of superparamagnetism with the cluster density and spin of the cluster which best fit the experimental data.

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The saturation magnetization in powder sample is about 280 A/m, which is only 4% of the saturation magnetization of single crystals. The difference can be contributed to the smaller density of the powder sample and the surface effects due to a grain structure of the powder sample. The full line on the Figure 3 is a function calculated from the model of superparamagnetism. The calculated number of spins in each cluster of ferromagneticaly ordered spins is of the same order of magnitude as the number of molecules in each grain of TDAE-C_{60} powder sample as determined by scanning electron microscope. The hysteresis in magnetization curve in TDAE-C_{60} powder sample has also been observed.

A similar magnetic response has been measured on APhF-Co. Figure 4 shows the phase transition to ferromagnetic phase in APhF-Co measured in the magnetic field strength of 4000 A/m. The transition temperature is about 18 K. The value of the saturated magnetization in APhF-Co is smaller than the saturated magnetization in TDAE-C_{60} powder sample (see Figure 3) by the same factor as is the ratio of densities of these two powders. This means that the average magnetic moment per molecule of TDAE-C_{60} and APhF-Co powder samples are of the same order of magnitude: about 3% and 2% of the Bohr magneton, respectively.

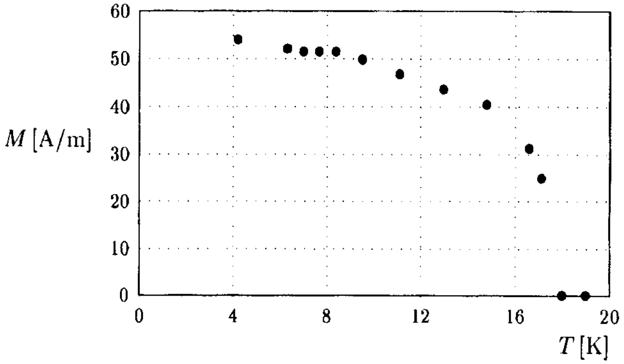

Figure 4 $M(T)$ dependence in APhF-Co.

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CONCLUSIONS

The results of our dc SQUID magnetization measurements on TDAE-C_{60} single crystals and powder samples indicate a magnetically ordered phase below 19 K, "S" shaped magnetization curves and hysteresis in the magnetization curves lead to the conclusion that TDAE-C_{60} behaves as a ferromagnet. The differences in magnetic properties between single crystal and powder sample can be attributed to the grain structure of the powder sample.

Another fullerene derivative, APhF-Co, where electron donor is cobaltocene instead of TDAE, has similar magnetic properties as TDAE-C_{60} powder sample. This can suggest that the essential ingredient for the ferromagnetic properties of these materials is the fullerene molecule.

References

[1] P.M. Allemand, K.C. Khemani, A. Koch, F. Wudl, K. Holczer, S. Donovan, G. Grüner, J.D. Thompson, *Science* **253,** 301 (1991).

[2] Suzuki, A., T. Suzuki, R.J. Whitehead, Y. Maruyama, *Chem. Phys. Lett.* **223,** 517 (1994).

[3] Lappas. A., K. Prassides, K. Vavekis, D. Arčon, R. Blinc, P. Cevc, A. Amato, R. Feyerherm, F.N. Gygax, A. Schenck, *Science* **267,** 1799 (1995).

[4] K. Tanaka, A.A. Zakhidov, K. Yoshizawa, K. Okahara, T. Yamabe, K. Yakushi, K. Kikuchi, S. Suzuki, I. Ikemoto, Y. Achiba, *Phys. Rev, B* **47,**7554 (1993).

[5] R. Blinc, K. Pokhodnia, P. Cevc. D. Arčon, A. Omerzu, D. Mihailović D., P. Venturini, L. Golič, Z. Trontelj. J. Lužnik, Z. Jagličić, J. Pirnat, *Phy. Rev. Lett.* **76,** 523 (1996).

[6] D. Arčon, P. Cevc, A. Omerzu, R. Blinc, *Phys. Rev. Lett.* **80,** 1529 (1998).

[7] A. Mrzel, P. Umek, P. Cevc, A. Omerzu, D. Mihailović, *Carbon,* **36,**603 (1998).

[8] K. Kim, D.J. Liddle, D.J. Cleary, *J. Phys. Chem.* **94.** 3205 (1990).

[9] Z. Jagličić, Z. Trontelj, J. Lužnik, J. Pirnat, R. Blinc, *Solid State Comm.* **101,** 591 (1997).