CHANGE OF ELECTRON CONFIGURATION OF OXYGEN MOLECULES ADSORBED ON NANOCARBONS

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Introduction

Oxygen adsorbed on a surface has been of great interest due to a possibility to form magnetic layers at low temperatures [1,2]. A surge of interest to study oxygen adsorption on carbon in last years is related to a finding of the way to control conduction properties of carbon nanotubes via the absorption [3]. Note that even such a control was experimentally demonstrated, its interpretation is still debatable. In particular, the change of properties of the adsorbed gas has not been monitored directly yet. Then, the question about occurrence of the charge transfer between an absorbed oxygen molecule and the substrate remains open. The present communication is an attempt to demonstrate that the electron state of the oxygen molecules adsorbed on the nanocarbons is changed. The method we propose consists of the in-situ measurements of the net magnetization of adsorbed oxygen.

Experimental

The magnetization measurements were carried out with a commercial SQUID (superconducting quantum interference device) magnetometer (Quantum Design, MPMS 7). The method to extract an information on the oxygen magnetic moment was the same as in our recent paper [4]. We measured the overall magnetic moment of carbon material put in a gelatine capsule before and after oxygen adsorption (which occurred just inside the SQUID chamber at moderate temperatures above about 55 K due to a leakage). Then the difference was considered as the net magnetic moment of oxygen adsorbed. It is worth noting that the use of the same sample without sealing-unsealing of the container for measurements (as is usually done) allowed one to get the oxygen magnetic moment with a very high precision.

As we showed [4] the shape of *M-H* (magnetic moment vs. applied magnetic field) curves at temperature of 2 K could be successfully used in order to characterize the magnetic state of oxygen in the case of adsorption occurring on the randomly distributed surfaces, while the absolute value of magnetic moment, M_0, at 2K at the maximal applied magnetic field (7 T in our experiment) is proportional to the number of adsorbed oxygen molecules bearing own spins. Note that the randomness of adsorbing surface orientations leading to the randomness of orientations of spins contributing to

the magnetic moment enables a clear interpretation of the experimental data unlike the case of a flat surface when possible tilt angle (as well as its distribution) of oxygen molecules makes the interpretation difficult.

We studied the magnetization of oxygen adsorbed on the surface of onion-like carbon nanoparticles (CN) and on the surface of multi-walled carbon nanotubes (MWNT). As in our previous paper [4] the use of powder provides the randomness of adsorbing surface orientations. After the occurrence of oxygen adsorption inside the SQUID chamber (that was realized at 60 K in our experiment) we cooled samples down and then did not increase the temperature above 40 K in order to prevent the additional oxygen adsorption. Then, the variation of the magnetization of the adsorbed oxygen was monitored at different temperatures and magnetic fields.

Results and Discussion

Measuring magnetic moment of adsorbed oxygen in the field cooling and then in the field warming, we found that the corresponding curves differ at the temperature region between about 15 K and 30 K showing a magnetic irreversibility. The field-warming curve lies below the field cooling one. We also found that the difference between the field-cooling and field-warming magnetic moment increases (being scaled) with the decrease of the applied magnetic field. These experimental results are similar to those reported in [5], where the magnetic moment decrease was related to the transition between different magnetic phases of oxygen.

In order to get a quantitative information about the found irreversibility we succeeded the following experiment. The sample was cooled from 40 K down to the temperature of 2 K, and then, the *M-H* curves were measured at this temperature low temperature. After that, the sample was warming up to 40 K, where no irreversibility was found for magnetic moment of adsorbed oxygen and cooling was done at different regime (either the different applied magnetic field or the different temperature sweeping rate). Repeating this process many time, we found that the variation of the value of M_0 consisted about 30 % in our experiment. Note that the cooling at smaller magnetic field or at smaller temperature sweeping rate (larger time being within irreversibility region) led to smaller value of M_0. At the same time for all state achieved at 2 K the shape of the *M-H* curves was exactly the same. It means that being scaled (to the same value of M_0) the *M-H* curves coincided. The latter allows us to claim that only the number of oxygen spins contributing to total magnetic moment at 2 K in our experiment depended on the regime we achieved this low temperature while no different magnetic phases were found at 2 K. Note that such interpretation of results is totally different from that in [5].

Conclusions

The found variation of the magnetic moment M_0 (depending on the regime we passed the irreversibility region) without change of the shape of the *M-H* curves allows us to claim that we directly monitored the decrease of the number of adsorbed oxygen

molecules bearing own spins. This decrease of spins has been ascribed to the change of electron configuration of the adsorbed oxygen molecules.

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