Orientational phase transition in C\(_{60}\) single crystal under high pressure

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Abstract By using a piston-cylinder type hydrostatic high-pressure unit, the electrical dc conductance of bulk C\(_{60}\) single crystal was measured in the pressure range of 0 to 0.6 GPa at 290 K. With increasing pressure, a discontinuous increase of the conductance was observed at the pressure of 0.176 GPa, which can be ascribed to the orientational ordering phase transition. In the simple cubic phase, the increase of conductance with increasing pressure is faster than that in the face-centered-cubic phase.

Keywords: C\(_{60}\), high pressure, conductivity.

The unique cage structure of the C\(_{60}\) molecules has evoked immense interest of researchers. One of the most prominent characteristics of solid C\(_{60}\) is the rotational properties of C\(_{60}\) molecules. At room temperature and ambient pressure, solid C\(_{60}\) adopts a face-centered-cubic (fcc) structure\(^{[1]}\). The molecules which are orientationally disordered, rotate nearly freely and independently of each other\(^{[1]}\). Below \(-260\) K, the electrostatic interaction of the molecules produces a freezing of the free molecular rotation\(^{[2]}\). In this state, the four molecules of the fcc conventional unit cell become orientationally nonequivalent, and the C\(_{60}\) crystal undergoes a first-order phase transition to a simple-cubic (sc) structure.

The rotational fcc-sc transition has also been observed to take place at room temperature under high pressure. Many experimental techniques, including Raman spectroscopy\(^{[3]}\), X-ray diffraction\(^{[4]}\), differential thermal analysis\(^{[5]}\) and neutron diffraction\(^{[6]}\), have been used to determine the orientational fcc-sc phase transition under high pressure. And the results show that the orientational fcc-sc phase transition occurs in a pressure range of 0.2—0.5 GPa at 300 K for C\(_{60}\) single crystal. However, almost all of the reported orientational fcc-sc phase transition pressures were measured by using a diamond anvil cell or an aluminum pressure cell, which were very likely to introduce errors in calibrating the pressure especially within the lower pressure range. So it is still necessary to do some further investigations on the pressure-induced orientational fcc-sc phase transition for C\(_{60}\).

The orientational phase transition affects physical properties such as the thermal conductivity, the attenuation of sound, the magnetic susceptibility, enthalphy and specific heat capacity. Although Regueiro et al. reported an electrical resistance measurement result under high pressure by using diamond anvil technique in the pressure range of 5—23.4 GPa\(^{[7]}\), there still lack reports on the pressure-induced conductance behavior in the lower-pressure regime at room temperature.

In this note, we present D. C. electrical conductance variation with hydrostatic pressure of
C\textsubscript{60} single crystals. A dramatic conductance jump was observed at 0.176 GPa. It is also observed that the slope of electrical conductance versus pressure changes significantly before and after phase transition point.

1 Experimental

The C\textsubscript{60} single crystal was grown by using vapor sublimation method. The sample was cut into a 1 mm \( \times \) 2 mm \( \times \) 0.5 mm rectangle and cleaned with acetone. The electrodes were prepared by evaporating silver pads through a patterned mask on the [111] plane of the C\textsubscript{60} single crystal. Then copper wires of 0.1 mm in diameter were attached onto the silver pads by silver paste. In order to minimize the influences of the ambient environment, the sample was first annealed at 500 K for 1 h in a vacuum of \( 1.3 \times 10^{-3} \) Pa, and then protected with a layer of BF4 glue.

High hydrostatic pressure was generated in a piston-cylinder type vessel which can be compressed in two directions by a 400-ton hydraulic press with double pistons and four pillars. Transformer oil was used as pressure transmitter. In the pressure cavity there was a manganin stress gauge. The hydrostatic pressure was calculated from the resistance of the manganin stress gauge which was measured with a Keithley model 2400 source/meter. This experimental setup can control the pressure from 0.0001 to 3 GPa continuously and precisely.

The D. C. electrical conductance measurements were performed by using the standard two-wire configuration with a computer-controlled Keithley model 2400 source/meter. The choice of the two-wire configuration was based on the large resistance value (\( \sim 50 \) M\textOmega\ at room temperature and ambient pressure) of our C\textsubscript{60} samples, so the contact resistance and wire resistance were negligible. In order to minimize the possible relaxation effect caused by the pressure change, a 5-min delay time was set before taking the data. All the experiments were done at 290 K.

2 Results and discussion

Figure 1 shows the reversible pressure dependence of the electrical conductance of a C\textsubscript{60} single crystal sample. In the pressure range below 0.176 GPa, the conductance of the sample increases with increasing pressure. As the pressure increases further, there is a sudden conductance jump. The slope of the conductance changes with pressure following this conductance jump is larger than that in the lower pressure range. If we normalize the conductance data to that measured at ambient pressure, then the slopes before and after the phase transition are calculated to be 2.6 and 7.1 GPa\textsuperscript{-1}, respectively. We have measured several single