Thermally Induced Acoustic Emission from Polycrystalline Buckminsterfullerene

Tibor Braun,[a] Péter Berényi,[b] András Illényi[b] and Sabirdjan Sakiev[b]

[a] *Institute of Inorganic and Analytical Chemistry, L. Eötvös University, PO Box 123, 1443 Budapest, Hungary*
[b] *Acoustic Research Laboratory, Hungarian Academy of Sciences, Budapest, Hungary*

Acoustic emission activity has been detected in C_{60} polycrystals at 260 K and at 190–200 K by heating and cooling the C_{60} crystals in the 180–300 K interval; the main acoustic activity is attributed to the fcc–sc phase transition in the polycrystalline material.

Acoustic emission (AE) has been found to accompany many physical and chemical processes in solids and in solid–liquid systems.[1–4] The AE appears as a transient elastic wave generated by the rapid release of energy within a material. The detection and analysis of the released AE signals can be performed with a piezoelectric transducer in contact with the sample investigated or to the outside of a reaction cell. This allows information to be obtained on physical mechanisms connected to deformations, transformations and defects throughout the matrix. We report here the first observation of thermally induced AE in polycrystalline C_{60} in the temperature range 180–300 K. Intense AE occurs in a relatively narrow temperature interval T at about 260 K. As previous investigations have shown an orientational ordering phase-transition in solid C_{60}[3,4] at 260 K it is supposed that the observed AE is related to that phase transition.

The investigated polycrystalline C_{60} (F & J Ltd., Moscow, Russia, crystals of 0.2–1.2 mm size, 99.9% purity) was previously examined by DSC and X-ray diffraction. The AE equipment used is shown in Fig. 1. The C_{60} crystals were placed in the cavity of the waveguide (2) and slightly compressed. In order to prevent water condensation during cooling of the cryostat chamber, the crystals were covered with a piece of cotton-wool, which was replaced after each thermal cycle. The intensity of the AE was measured with the arrangement according to Fig. 1(*a*). For analysing the momentary shape of the acoustic signals measurements were made according to Fig. 1(*b*). With this arrangement the real-time shapes of the thermoelastic signals were recorded in the investigated temperature interval. Cooling of the cryostat was performed using liquid nitrogen.

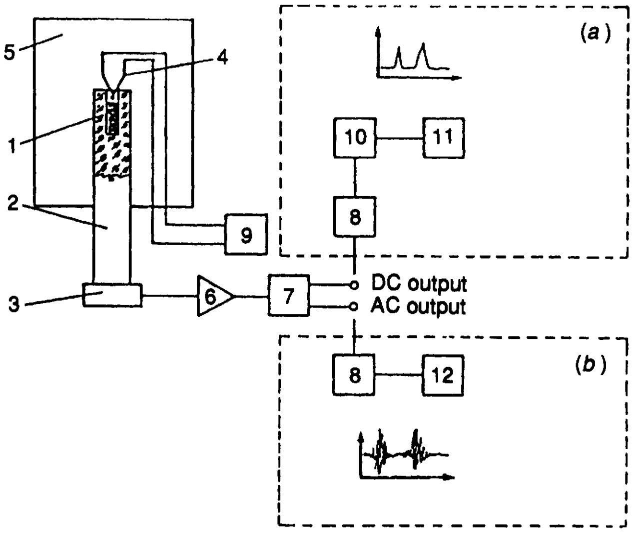

Fig. 1 Schematic diagram of experimental AE apparatus; (1) C_{60} samples, (2) glass waveguide (length = 100 mm, p = 7 mm, with sample cavity, width = 2 mm, length = 10 mm), (3) acoustic resonant sensor (Brüel & Kjaer 8313), (4) thermocouple (Cu–constantan), (5) cryostatic chamber, (6) preamplifier (Brüel & Kjaer 2637), (7) main amplifier (Brüel & Kjaer 2638), (8) digital storage oscilloscope (Iwatsu DS 6612), (9) microvoltmeter (Solartron 7071), (10) PC (IBM AT 286), (11) printer, (12) graphics plotter

Fig. 2 shows the AE signals as a function of temperature (180–300 K) for the empty sample cavity and for the cavity filled with graphite granules. As can be seen there are no AE signals detected above the noise level.

Fig. 3 shows the results of the same experimental cycle with the C_{60} sample. The measurement was repeated in Fig. 4 with a high resolution of the temperature range. Here the discrete lines show the integrated values of the AE signals within temperature intervals of 0.1 K in the range 243–264 K. An intense AE activity was observed in the temperature interval

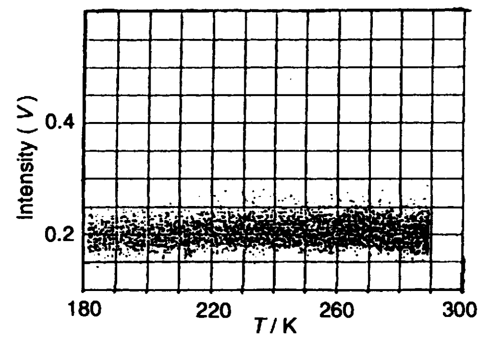

Fig. 2 Temperature dependence of the AE signal for a graphite sample and for the empty cavity

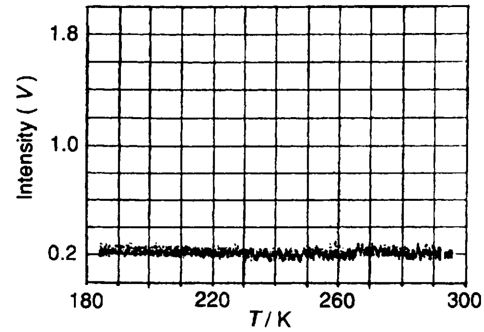

Fig. 3 Temperature dependence of single AE signals for a polycrystalline C_{60} sample

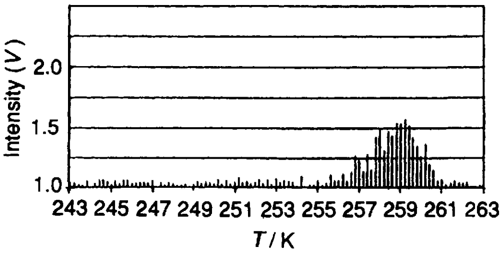

Fig. 4 Temperature dependence of the integrated (over 0.1 K) AE signals for a polycrystalline C_{60} sample

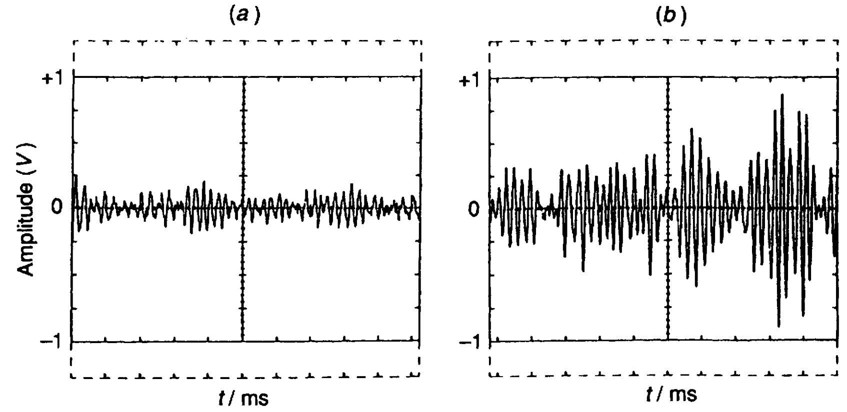

Fig. 5 Real-time shape of AE signals (*a*) at 262.7 and (*b*) at 256.8 K

256–260 K. The rate of temperature change in the cryostat was *ca.* 1° min^{-1}.

In order to investigate the shape (with time) of the AE signals, measurements at different temperatures were performed. Fig. 5(*a*) shows the amplitudes of the AE signals at 262.7 K (*i.e.* at a temperature above the intense AE activity. The average U_{Ae} was 8 mV, *i.e.* + 6 dB over the noise level of the preamplifier and the frequency is 30–50 kHz. Corresponding measurements at 256.8 K, *i.e.* in the range of intense AE activity [Fig. 5(*b*)], shows that the amplitude of the AE signals is *ca* 5–6 times that of those measured at 262 K, amounting to 40–60 mV with frequencies of 15–20 kHz.

It should be noted that in some experimental cycles an AE activity of comparable intensity appeared also in the region 190–205 K (Fig. 6). However contrary to the AE activity at 256–260 K, which was fully reversible, that at 190–205 K manifested itself only with a fresh C_{60} sample and did not appear during the next cooling–heating cycles. The origin of the intense AE activity around 260 K is probably related to the fcc–sc phase transition already reported.[5,6] This transition is accompanied by a discontinuous decrease in the lattice parameter and this could generate the AE activity. AE activities from phase transitions have already been reported in metals, alloys and in some chemical compounds.[1–4] As far as we know no such activity has been observed for fullerenes and/or fullerene compounds.

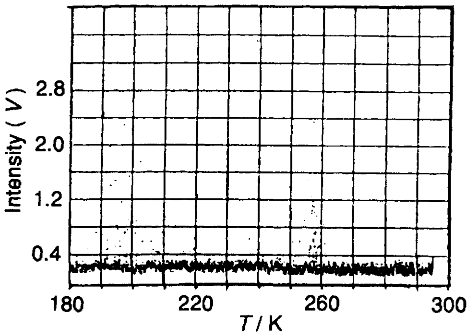

Fig. 6 Temperature dependence of the AE signal for a C_{60} sample at the first cooling–heating cycle

The irreversible AE activity observed at 190–200 K is attributed to the appearence of local, mechanical, thermal stresses in the C_{60} crystals. This effect seems to be similar to the situation when the material is exposed to external, mechanical stress.[1]

Our investigations will continue to obtain a more in depth understanding of the AE activity at the orientational ordering phase transition temperature in C_{60} and also for the explanation of the transient AE activity around 190–200 K.

T. B. gratefully acknowledges the financial help provided by the Hungarian OTKA Commission within grant No. T007642.

Received, 28th February 1994; Com. 4/01208I

References

1 R. V. Williams, *Acoustic Emission*, Adam Hilger Ltd., Bristol, 1980.
2 A. P. Wade, D. B. Sibbald, M. N. Bailey, R. M. Belchamber, S. Bittman, J. A. McLean and P. D. Wentzell, *Anal. Chem.*, 1991, **63**, 497A.
3 O. Lee, Y. Koga and A. P. Wade, *Talanta*, 1990, **37**, 861.
4 A. P. Cook, L. E. Bowman and A. P. Wade, *J. Crystal. Growth*, 1993, **131**, 395.
5 P. A. Heiney, *J. Phys. Chem. Solids*, 1992, **53**, 1333.
6 P. A. Heiney, J. E. Fischer, A. R. McGhie, W. Romanow, A. M. Denenstein, J. P. McCauley and A. B. Smith, III, *Phys. Rev. Lett.*, 1991, **66**, 2911.