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Characterizations of C_{60} Films by Measuring Internal Friction

内部摩擦測定による C_{60} 薄膜の物性評価

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1. Introduction

Due to high symmetry of C_{60} molecule and weak van-der-Waals interaction between them in crystal phase, solid C_{60} exhibits various properties with varying temperature. An elastic anomaly of solid C_{60} at around 160 K has been observed on temperature dependences of the sound velocity and the internal friction. This anomaly is considered to correspond to C_{60} jumping between inequivalent orientations[1]. It is well known that a structural phase transition between sc and fcc appears in C_{60} crystals around 260 K. This phase transition has been revealed in many measurements such as x-ray diffraction[2], sound velocity[1,3], dielectric constant[4], electrical conductivity[5], NMR[6], and thermal conductivity[7]. At temperatures higher than 300 K Vickers hardness of C_{60} crystal showed an anomalous behavior that the hardness increases with increasing temperature and reached a maximum at around 370 K[8]. The calorimetric measurement also showed a slightly hysteretic properties of specific heat of C_{60} crystal in the temperature range of 310~370 K[9]. Moreover, the properties of C_{60} crystal at around 420 K were also been studied by several groups. Raman scattering study showed that above 400 K the pentagonal pinch mode changes in frequency and becomes sharp compared with that at room temperature[10]. This result indicats that partial orientational order remained above 260 K and only above 400 K the C_{60} molecules rotate freely and isotropically. Calorimetric measurement showed that there was a peak of heat capacity in C_{60} crystal at 425 K[9]. The internal friction measured by means of the free-free bar apparatus showed that a λ-shaped peak was detected in C_{60} films at 426 K[11]. The capacitance and dissipation factor curves have a pronounced feature at 435 K, confirming a phase transition above 400 K[12].

In this study, the C_{60} films are grown on Si substrate by means of evaporating C_{60} powder in vacuum. The internal friction of the C_{60}/Si sample is measured by reed-vibriting method, in which electrostatic drive and laser displacement sensor are used. We also calculated several crystallographic parameters of C_{60} crystal as a function of temperature. By comparing the theoretical and experimental results, the structural and dynamic properties of C_{60} molecules in crystal phase will be discussed.

2. Experimental

The C_{60} films were deposited on the Si substrate of $40 \times 5 \times x$ mm[3] at 473 K by sublimation in vacuum with residual gas pressure below 2.0×10^{-6} Torr. The thickness of the C_{60} films on the Si substrate was about 0.5 μm. The internal friction of C_{60} films was evaluated by subtracting the internal friction of Si substrate from that of the C_{60} film/Si substrate sample.

3. Results and Discussion

Figure 1 shows temperature dependences of internal friction of C_{60} films in the temperature range of 100~500 K for two frequencies of 224 and 444 Hz. Several peaks of the internal friction are observed. Except the peak 4, they shift to higher temperatures with increasing frequency. The peaks 1 and 2 are attributed to relaxation processes of C_{60} molecule between different orientations in sc lattice. The peak 3 is due to order-disorder phase transition corresponding to sc and fcc structures. The peak 4 at 360 K is from Si substrate, and independent of frequency. The peak 5 is related to a transition between solid and ‘liquid’ phases because of the evaporation temperature of 460 K for C_{60} crystal.

In order to clarify mechanisms of the internal friction happened in the C_{60} films, we calculated both diameter and nearest-neighbor distance of C_{60} molecules in sc and fcc crystal as a function of temperature. The results obtained are shown in Fig. 2. These values are defined as follows:

$$\delta D(\mathrm{T}) = D(\mathrm{T}) - D_0,\ \ \delta d(\mathrm{T}) = d(\mathrm{T}) - d_0$$

where D_0 and d_0 is diameter and nearest-neighbor

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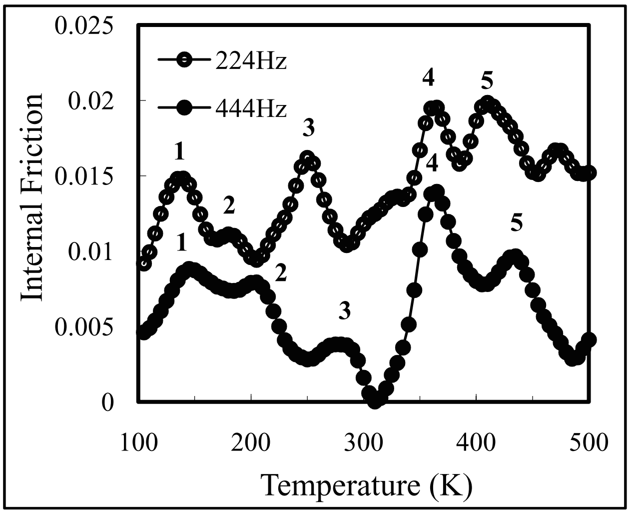

Fig. 1 Temperature dependences of the internal friction of C_{60} films. The open and closed circles represent the results measured at frequencies of 224 Hz and 444 Hz, respectively.

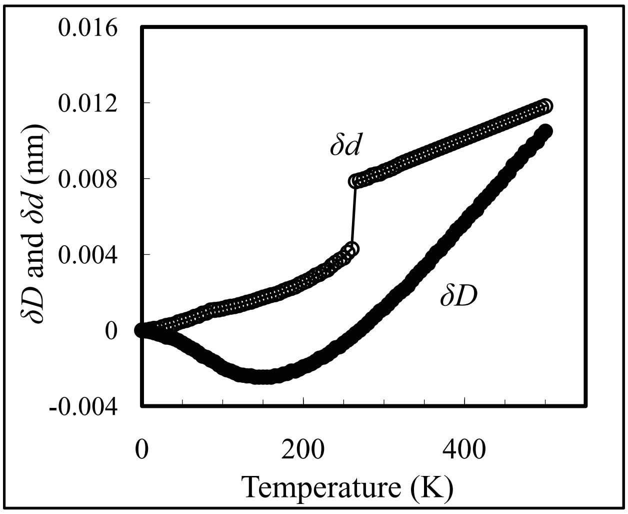

Fig. 2 The diameter (δD) and the nearest-neighbor distance (δd) of C_{60} molecules in sc and fcc crystal phases as a function of temperature.

distance of C_{60} molecules at 0 K, respectively. Also, we used that $d(\mathrm{T}) = 1.4154/\sqrt{2} = 1.001$ nm and $D(\mathrm{T}) = 0.71$ nm at temperature of 270 K. Except a sharp variation at about 260 K, δd is proportional to temperature. On the other hand, there is a minimum of δD at about 150 K. The difference between δd and δD shows several special points at temperature, below 100 K, around 150 K, at 260 K and above 260 K. Below 100 K the decrease in the $\delta d - \delta D$ results in a glassy phase in the C_{60} film. In the range of 150～200 K, the large difference between δd and δD is advantageous to orientational order relaxation of C_{60} molecules. Around 260 K the sc-fcc structural phase transition can be certainly detected by measuring internal friction. The decrease of the $\delta d - \delta D$ above 260 K, leading to the solid~liquid phase transition, results in a peak around 420 K for internal friction.

4. Conclusion

The internal friction of the C_{60}/Si sample was measured by using reed-vibriting method. Several relaxation peaks were observed on the temperature dependence of internal friction of C_{60} film. These relaxation processes are related to the variation in both the diameter and the nearest-neighbor distance of C_{60} molecules in crystal phase.

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