RHEED intensity oscillation of C_{60} growth on GaAs substrates

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Abstract

Intensity oscillation of reflection high-energy electron diffraction (RHEED) is observed during C_{60} layer epitaxial growth on GaAs (111)B and (111)A substrates. The frequency of the oscillation coincides well with growth rate of C_{60} layers, suggesting that C_{60} grows by repeating nucleation and a step flow growth as with GaAs and other semiconductor materials. Unusual oscillation is observed in the initial C_{60} layer growth on GaAs (111)B substrates with (2 x 2) reconstruction. The initial layer growth is completed at approximately half monolayer coverage by C_{60} molecules. This phenomenon is explained by the model that C_{60} absorption sites are limited due to As-trimers absorbed on (111)B surfaces. This model is strongly supported by the fact that no such effect is observed on GaAs (111)A substrates where no As-trimer is absorbed.

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1. Introduction

Extensive investigations have been done for the physical and chemical properties of C_{60}, and have revealed their potentialities in superconductivity [1] and photoconductivity. C_{60} molecules are highly symmetric and crystallize into a face-centered cubic structure on crystalline substrates such as Si and GaAs, in spite of the large lattice mismatch between C_{60} and the substrates [2-6]. Reflection high-energy electron diffraction (RHEED) is used to observe growth and surface conditions of semiconductor films, and RHEED intensity oscillation technique enables dynamic effects to be investigated [7]. In the present work, to investigate growth mechanism of C_{60} layers, we observe RHEED intensity oscillation during epitaxial growth of C_{60} layers on GaAs substrates, and Clear RHEED intensity oscillation has been successfully observed. The frequency of the oscillation proportionally increases with the C_{60} flux, and coincides well with the growth rate of C_{60} layers. This suggests that C_{60} layers grow with repeating nucleation and a step flow mode as with growth of GaAs and other semiconductor materials.

2. Experimental procedure

C_{60} layers are grown on GaAs (111)B and (111)A substrates by using solid source molecular beam epitaxy. GaAs substrates are first etched in an alkaline etchant, and loaded in the growth chamber. Native oxide layers of GaAs surfaces are removed by a thermal flash at 580 °C in As_4 atmosphere. After growing a GaAs buffer layer at 500 °C, the substrate temperature is lowered to 200 °C in As_4 atmosphere, and C_{60} layer growth is performed. 99.5 % C_{60} powder is used for the C_{60} source. C_{60} beam equivalent pressure (BEP) is varied between 8.0×10^{-7} and 2.4×10^{-6} Torr. RHEED measurements are performed with an electron beam along the <011> azimuth of the GaAs substrates. The intensity of a RHEED specular reflection is captured by a video camera system, and the intensity oscillation is analyzed by a personal computer.

3. Results and discussion

RHEED patterns of C_{60} layers grown on GaAs (111)B and (111)A substrates exhibit a six-fold symmetry and indicate that the epitaxial orientation is [111] direction on both substrates [5]. The observed streak intervals of the patterns indicate that the lattice constant of epitaxial cubic C_{60} films coincides well with the value of bulk cubic C_{60} crystals.

Figure 1 shows the intensity of the specular beam in the RHEED pattern of a C_{60} layer on a GaAs (111)B (2 x 2) structure. The lateral axis denotes a deposition thickness in monolayers (MLs). After a few monolayer depositions, regular RHEED intensity oscillation is observed. Figure 2 shows the frequency of the oscillation as a function of C_{60} BEP. The frequency proportionally increases with the C_{60} BEP, and coincides well with growth rate of

C_{60} layers obtained from the film thickness. Therefore, the RHEED intensity oscillation during C_{60} growth is caused by the same mechanism as that of GaAs and other semiconductor growth. C_{60} layers grow with repeating nucleation and a step flow mode.

Next, we discuss the process of the initial C_{60} layer growth. After C_{60} shutter opening, a shoulder pointed by an arrow is found at 0.5 ML, and the minimum point is at 1.0 ML. After that, intensity peaks appear at 1.5 ML, 2.5 ML, 3.5 ML and so on. This unusual oscillation suggests a peculiar arrangement of the first-layer C_{60} molecules on a GaAs (111)B (2 x 2) structure. Figure 3 shows a model of a C_{60} first-layer on a GaAs (111)B (2 x 2) structure. Since the surface of a GaAs (111)B (2 x 2) structure has a periodic lattice structure by absorbed As-trimers as shown in Fig. 3 [8], the C_{60} absorption sites should, therefore, be limited due to the steric structure between C_{60} molecules and As-trimers. In this case, the density of C_{60} absorption sites is estimated to be approximately 50 % of the full coverage. Therefore, the first-layer growth should be completed at 0.5 ML C_{60} deposition. The shoulder of 0.5 ML deposition probably indicates this event. The first peak appears at 1.5 ML deposition, indicating that the regular layer by the C_{60} growth takes place after first-layer deposition.

In order to verify the C_{60} first-layer arrangement model on the (111)B (2 x 2) surface, RHEED intensity oscillation is also investigated during the growth of C_{60} on a GaAs (111)A

(2 x 2) structure. Figure 4 shows an absorption model of first-layer C_{60} molecules on a GaAs (111)A surface with (2 x 2) reconstruction. The GaAs (111)A (2 x 2) structure is gallium terminated with one of four surface gallium atoms missing [9], and the surface periodicity is much finer than the C_{60} molecules size. Therefore, C_{60} molecules will be crystallized in a close packed form from the first-layer. Figure 5 shows the intensity of the specular beam in the RHEED patterns of C_{60} layers on a GaAs (111)A (2 x 2) structure as a function of C_{60} deposition thickness. Clear RHEED intensity oscillation is observed, but no peculiar characteristic is detected. After the C_{60} shutter opening, a shoulder pointed by an arrow is found at 1.0 ML, and the intensity peaks appear at 2.0 ML, 3.0 ML and so on. This indicates that the C_{60} molecule is close packed from the first-layer. This result strongly supports the model that a configuration of a C_{60} first-layer is determined by steric structures between C_{60} molecules and substrate surfaces.

4. Conclusions

Clear RHEED intensity oscillation during epitaxial growth of C_{60} layers on GaAs (111)B and (111)A substrates is observed. The frequency of the oscillation proportionally increases with C_{60} BEP, and coincides well with the growth rate of C_{60} layers. Therefore, C_{60} layers grow with repeating nucleation and a step flow mode as with the growth of GaAs and other semiconductor materials. In the initial C_{60} layer growth on a GaAs (111)B (2 x 2) structure, a shoulder is found at 0.5 ML, and intensity peaks appear at 1.5 ML, 2.5 ML and so on. This peculiar oscillation has been explained by the considering the structures of the surface. The surface structure of GaAs (111)B (2 x 2) is largely modified by the absorption of As-trimers. As a result, available surface sites density of C_{60} molecules is approximately 50 % of the full coverage. Thus, the first flat surface appears after 0.5 ML deposition. This model is strongly supported by the fact that no such effect is observed on GaAs (111)A substrates where no As-trimer is absorbed.

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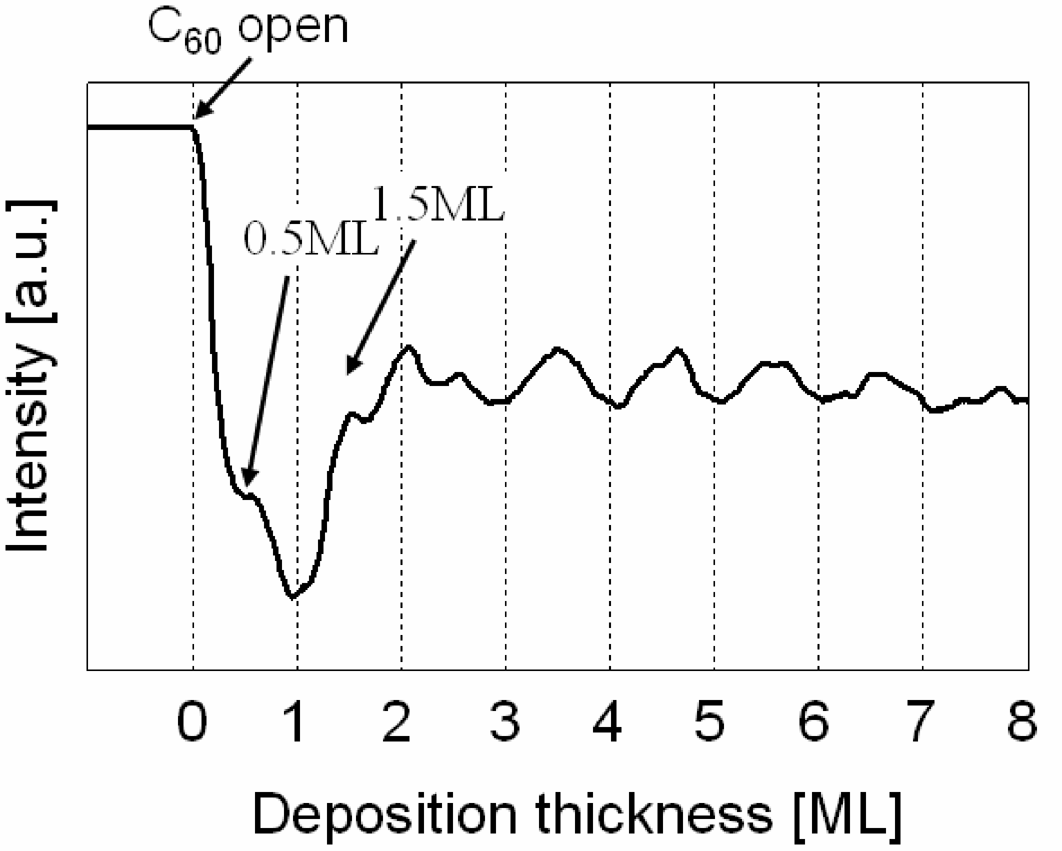
C_{60} open
0.5ML
1.5ML
Intensity [a.u.]
0
1
2
3
4
5
6
7
8
Deposition thickness [ML]

Fig. 1

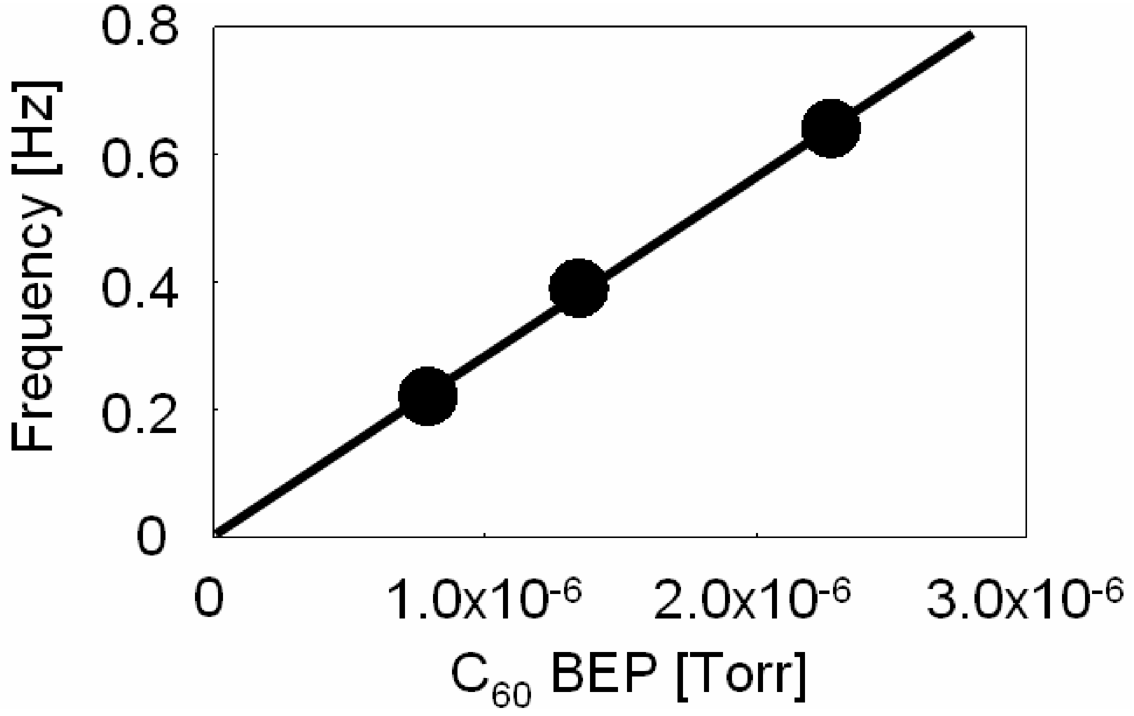
0.8
0.6
0.4
0.2
0
Frequency [Hz]
0
1.0×10^{-6}
2.0×10^{-6}
3.0×10^{-6}
C_{60} BEP [Torr]

Fig. 2

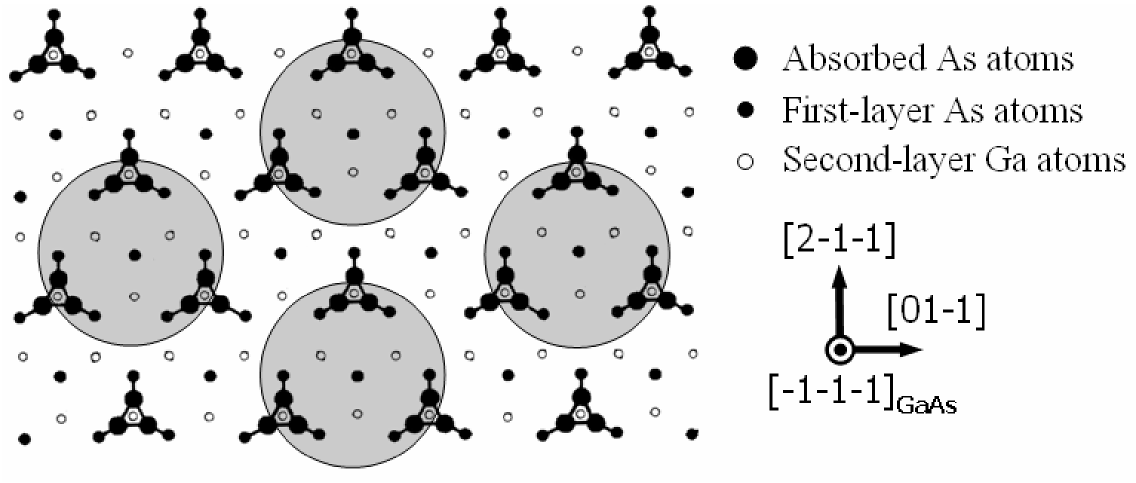
Absorbed As atoms
First-layer As atoms
Second-layer Ga atoms
[2-1-1]
[01-1]
[-1-1-1]GaAs

Fig. 3

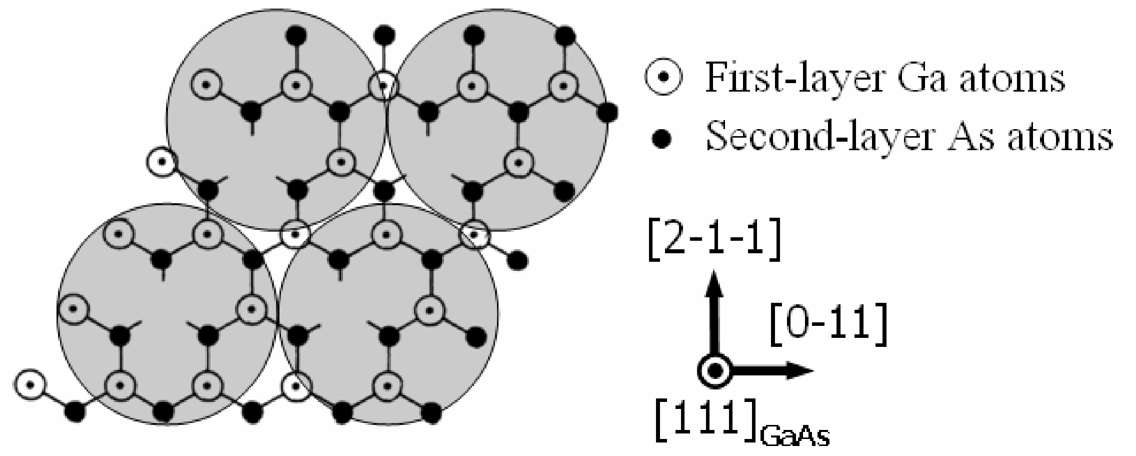
First-layer Ga atoms
Second-layer As atoms
[2-1-1]
[0-11]
[111]GaAs

Fig. 4

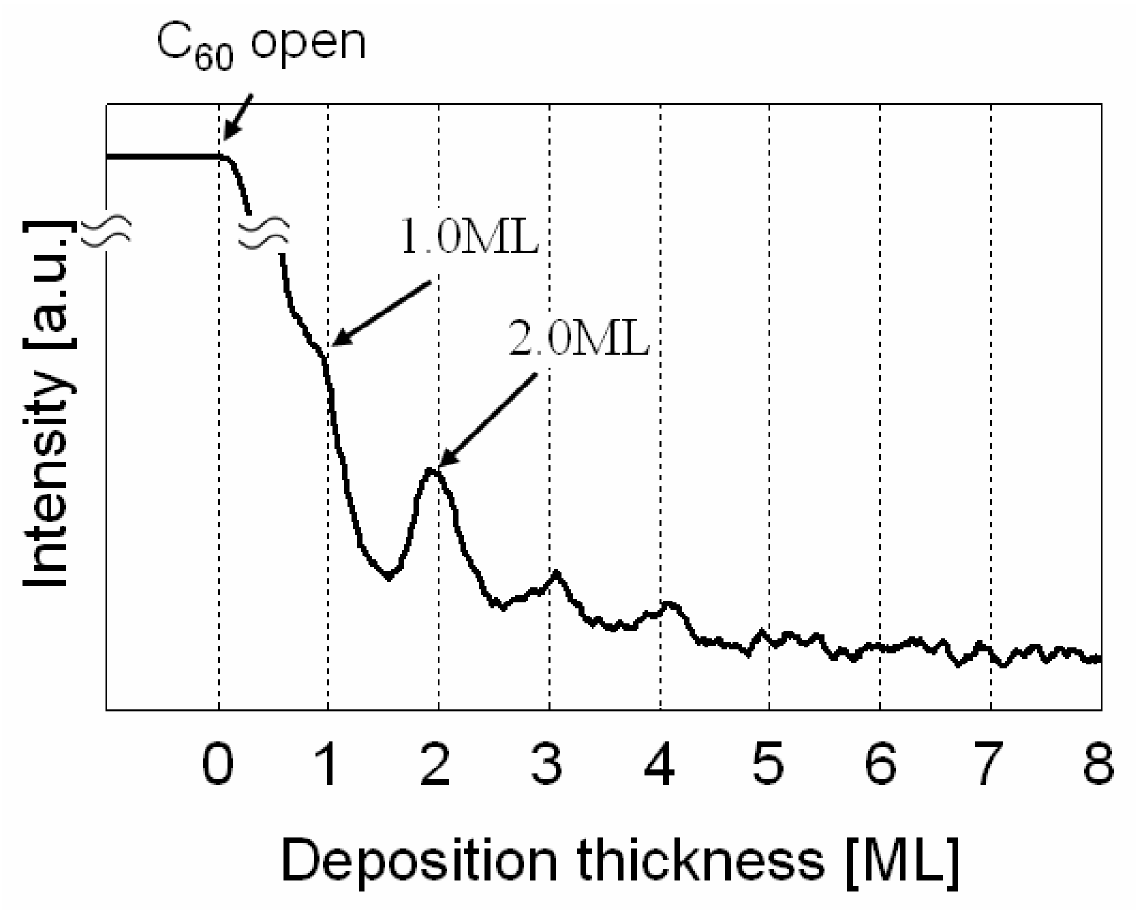
C60 open
1.0ML
2.0ML
Intensity [a.u.]
0
1
2
3
4
5
6
7
8
Deposition thickness [ML]

Fig. 5