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Nanoscale characterization of few-layer hexagonal boron nitride by scattering-type scanning near-field optical microscopy

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Abstract

We demonstrate a method for determining the number of h-BN layers with scattering-type scanning near-field optical microscopy (s-SNOM). We found that s-SNOM near-field signals show a h-BN thickness dependence and that the number of layers can be determined by comparing the measured data with analytical calculations. We used this method to investigate the in-plane distribution of the number of h-BN layers grown on a sapphire substrate, and then we identified three to five layers of h-BN, which is consistent with electron microscope observations. The present method also revealed that their domain size is about 100–200 nm. These results indicated that s-SNOM provides a non-destructive way for determining the number of h-BN layers with high spatial resolution.

1. Introduction

Hexagonal boron nitride (h-BN) is a layered material with a hexagonal lattice structure consisting of boron and nitrogen atoms. It has a large bandgap, and it has neither dangling bonds nor charge traps on its surface, which, if present, degrade the excellent physical properties of two-dimensional (2D) materials. These unique properties make h-BN a leading candidate as a dielectric substrate for 2D-material-based devices. In fact, 2D materials on h-BN show high carrier mobility [1], high photoluminescence intensity [2], and a long propagation length of plasmon polaritons [3]. However, most studies are currently using micrometer-size h-BN flakes mechanically exfoliated from bulk crystals.

For large-scale device applications, the growth of wafer-scale h-BN has been widely investigated by using chemical vapor deposition on catalytic transition metal substrates [4]. However, how to control the number of layers remains an issue. The dielectric properties of h-BN have thickness dependence, and thus controlling the layer number is one of the critical steps for fabricating 2D-material-based devices with h-BN. To achieve the homogeneous growth of h-BN with controlled thickness, we need to clarify the growth mechanism, which requires a measurement method for characterizing the in-plane distribution of the layer number. Raman spectroscopy is widely used to characterize the structural properties of 2D materials. The relative peak intensity and relative peak position of the two Raman signals provide a measure of the layer number of graphene [5] and MoS_2 [6], respectively. h-BN exhibits a characteristic Raman peak at around 1370 cm^{-1} attributed to the in-plane phonon mode. Monolayer h-BN can be identified from an upward shift of the Raman peak position [7]. However, the position also depends on local strain, so the determination of the number of h-BN layers is not as simple as in the case of other 2D materials.

In this study, we investigated h-BN layers grown by metal-organic chemical vapor deposition (MOCVD) using scattering-type scanning near-field optical microscopy (s-SNOM), which is a nanoscale optical characterization method based on atomic force microscopy (AFM). We performed an analytical calculation to examine the relationship between the near-field amplitude and the number of h-BN layers.

2. Experimental

h-BN was grown on *c*-plane sapphire by MOCVD. The precursors of h-BN were triethylboron and ammonia, and the carrier gas was hydrogen. First, an AlN buffer layer was formed on the sapphire by nitridation of its surface under ammonia flow, and then h-BN was grown on it.

We used a commercial s-SNOM system (Neaspec GmbH) to determine the number of layers of the h-BN. In the s-SNOM system, strong optical fields at the tip apex, induced by focusing an infrared beam from a quantum cascade laser, excite the sample, and the scattered light is recorded [schematically shown in Fig. 2(c)]. In this experiment, we imaged the sample at a frequency ω = 1370 cm^{-1}, where the strong signal is expected to be obtained due to the in-plane optical phonon mode of h-BN.

3. Results and discussion

First, we directly observed the number of layers of the MOCVD-grown h-BN by transmission electron microscopy (TEM). The low-magnified cross-sectional TEM image indicates that h-BN layers were continuously grown on the AlN buffer layer [Fig. 1(a)]. The number of h-BN layers can be directly counted in the high-magnified images. In this sample, the number of h-BN layers fluctuated depending on observation positions, ranging from three [Fig. 1(b)] to five [Fig. 1(c)]. Note that TEM observation requires destructive processing of samples.

The h-BN layers were investigated by s-SNOM to determine the layer number without any processing. Here, we measured a different area of the same sample used for the TEM observation. Although the height image in Fig. 2(a) indicates a height variation of several nanometers, it cannot reveal the number of layers. In Fig. 2(b), we plot the normalized near-field amplitude ($s = s_{sample}/s_{sapphire}$), where s_{sample} and $s_{sapphire}$ are raw signals from the sample and bare sapphire substrate, respectively. The image shows mainly the two parts: s = 3.0 and s = 3.2–3.3. This could reflect the difference in the

number of layers.

To reveal the relationship between the near-field amplitude and the number of h-BN layers, we performed an analytical calculation based on the finite dipole model [8, 9], where the tip is modeled as a perfectly conducting spheroid. The s-SNOM near-field signals can be explained by the scattering coefficient proportional to the effective polarizability of the tip. The effective polarizability depends on the function of the tip-sample distance and quasistatic reflection coefficient, and the reflection coefficient depends on the permittivity of the sample and the number of layers. In this calculation, we used a layer structure of air/h-BN/AlN/sapphire. Figure 2(d) shows the calculated near-field amplitude as a function of the number of h-BN layers and the optical frequency ω. At around ω = 1370 cm^{-1}, s shows a larger change with the number of h-BN layers, whereas it shows a small change when ω is far from 1370 cm^{-1}. Figure 2(e) shows the data at ω = 1370 cm^{-1} extracted from Fig. 2(d). Our calculation suggests that s-SNOM images measured at around ω = 1370 cm^{-1} can distinguish the number of h-BN layers.

By comparing the value of s in the s-SNOM experiments shown in Fig. 2(b) with the calculation, we found that 100–200-nm h-BN domains with three layers (s = 3.2–3.3) and five layers (s = 3.0) were grown in this sample. The number of layers is consistent with the TEM observation. s-SNOM images give us the information about not only the number of layers but also their in-plane distribution.

3. Conclusions

The number of h-BN layers grown on a sapphire was characterized by s-SNOM. The normalized near-field amplitude depends on the layer number, which can be explained by a finite dipole model. We demonstrated that the number of h-BN layers can be counted by s-SNOM with nanometer-order spatial resolution.

Acknowledgements

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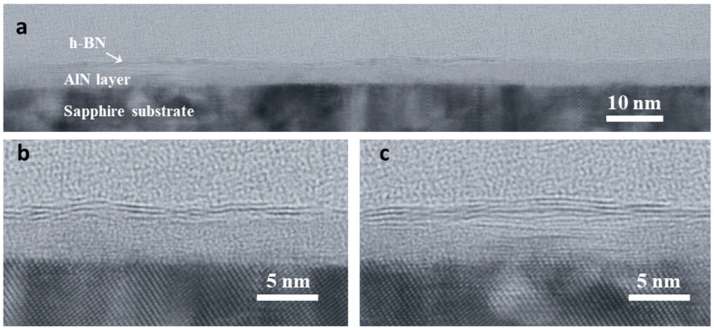

Fig. 1 Cross-sectional TEM images of the h-BN grown by MOCVD. **(a)** Low- and **(b), (c)** high-magnified images.

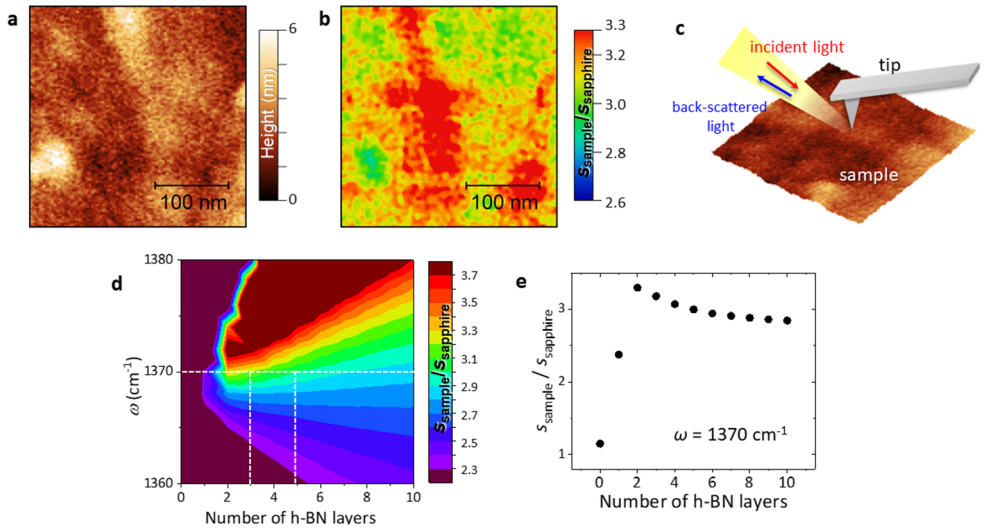

Fig. 2 (a) Height image and **(b)** the normalized near-field amplitude ($s_{sample}/s_{sapphire}$) images at an optical frequency ω = 1370 cm^{-1}, both of which were obtained for the MOCVD-grown h-BN using s-SNOM. **(c)** Schematic of s-SNOM measurement. **(d)** Calculated near-field amplitude ($s_{sample}/s_{sapphire}$) as a function of the number of h-BN layers and ω. **(e)** Layer number dependence at ω = 1370 cm^{-1}.