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Ultra-low threshold optically pumped random laser emission behavior of highly oriented pyrolytic graphite

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ABSTRACT

Graphite is a semimetal and thus has no bandgap; photoluminescence is not expected from relaxed charge carriers. Here, however, an ultralow threshold optically pumped random lasing behavior in highly oriented pyrolytic graphite (HOPG) was observed under 488 nm Ar^+ laser excitation at room temperature. In addition, this lasing behavior differs from the conventional random lasing process, i.e., the emission exhibits an ultra-low lasing threshold and a linear dependence on the laser excitation intensity in a low power range, and it saturates at high excitation densities. The theoretical model which accounts for the optically pumped random lasing behavior in HOPG has been proposed.

1. Introduction

Due to its unique structural properties, graphite has stimulated tremendous interest in both fundamental physics studies and potential applications in recent years, especially, since the successful preparation of single-layer graphene by mechanical exfoliation [1]. Besides its low cost, non-toxic, excellent mechanical and chemical stability, and superior electrical and thermal conductivity [2], it exhibits potential applications for transferable and high-power optoelectronics devices [3]. Since it has no bandgap [4], photoluminescence is not expected from relaxed charge carriers. And there have been little theoretical and experimental reports of lasing from graphite until now.

In this paper, we report the observation of a significant random lasing behavior from highly oriented pyrolytic graphite (HOPG) under excitation by 488 nm Ar^+ laser. Lasing emission was found to occur across the visible spectral range (1.65–2.07 eV). In addition, this lasing behavior differs from the conventional random lasing process, i.e., the emission exhibits an ultra-low lasing threshold and a linear dependence on the laser excitation intensity in a low power range, and it saturates at high excitation densities. The qualitative model to clarify the mechanism of this emission behavior in HOPG was proposed.

2. Experimental

Commercially available HOPGs were employed as samples. We have simulated the deformation of HOPG using molecular dynamic (MD) methods with a reactive empirical bond order potential [5]. The simulation was performed on a graphite supercell of 1992 atoms ($2.46 \times 2.46 \times 4$ nm^3 in dimension) within an canonical ensemble at 3000 K. The structural model as plotted in Fig. 1(a) was fully relaxed for 2 ns with a MD time of 1 fs. The crystalline quality and orientation of the HOPG samples were investigated by X-ray diffraction (XRD) using a D/Max-2400 ($CuK_{\alpha 1}$:λ=0.154056 nm). Photoluminescence (PL) measurements were carried out at room temperature by a Jobin Yvon HR320 spectrometer using an Ar^+ laser with an excitation wavelength of 488 nm. Optical filters have been used to obtain single-mode operation of Ar^+ laser during the test. The relative reflection spectra were measured by a UV–vis–NIR spectrophotometer (UV-3600) with integrated sphere.

3. Results and discussion

As shown in Fig. 1(a), HOPG consists of graphene layers that are stacked like a "house of cards" with many distortions in the structure. The presence of wrinkle-like structures and moiré superlattices are usually attributed to the rotation of the top graphene layer with respect to the second layer [6]. Fig. 1(b) shows the typical XRD pattern of HOPG. The dominant diffraction peak at ~26.4 corresponds to the hexagonal graphite (002), the peaks at ~42.5, ~44.9, ~54.3, and ~77.5 can be indexed to (100), (101), (004), and

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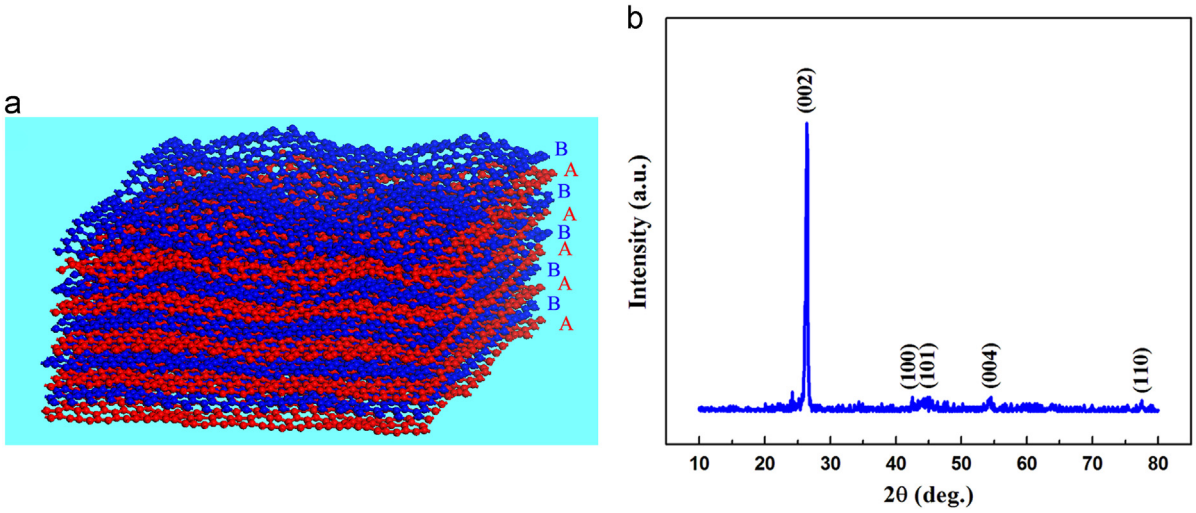

Fig. 1. (a) A schematic representation for simulating the deformation of HOPG with the layer sequence ABAB… using the molecular dynamic (MD) methods with a reactive empirical bond order potential diagram. (b) The representative XRD pattern of HOPG sample.

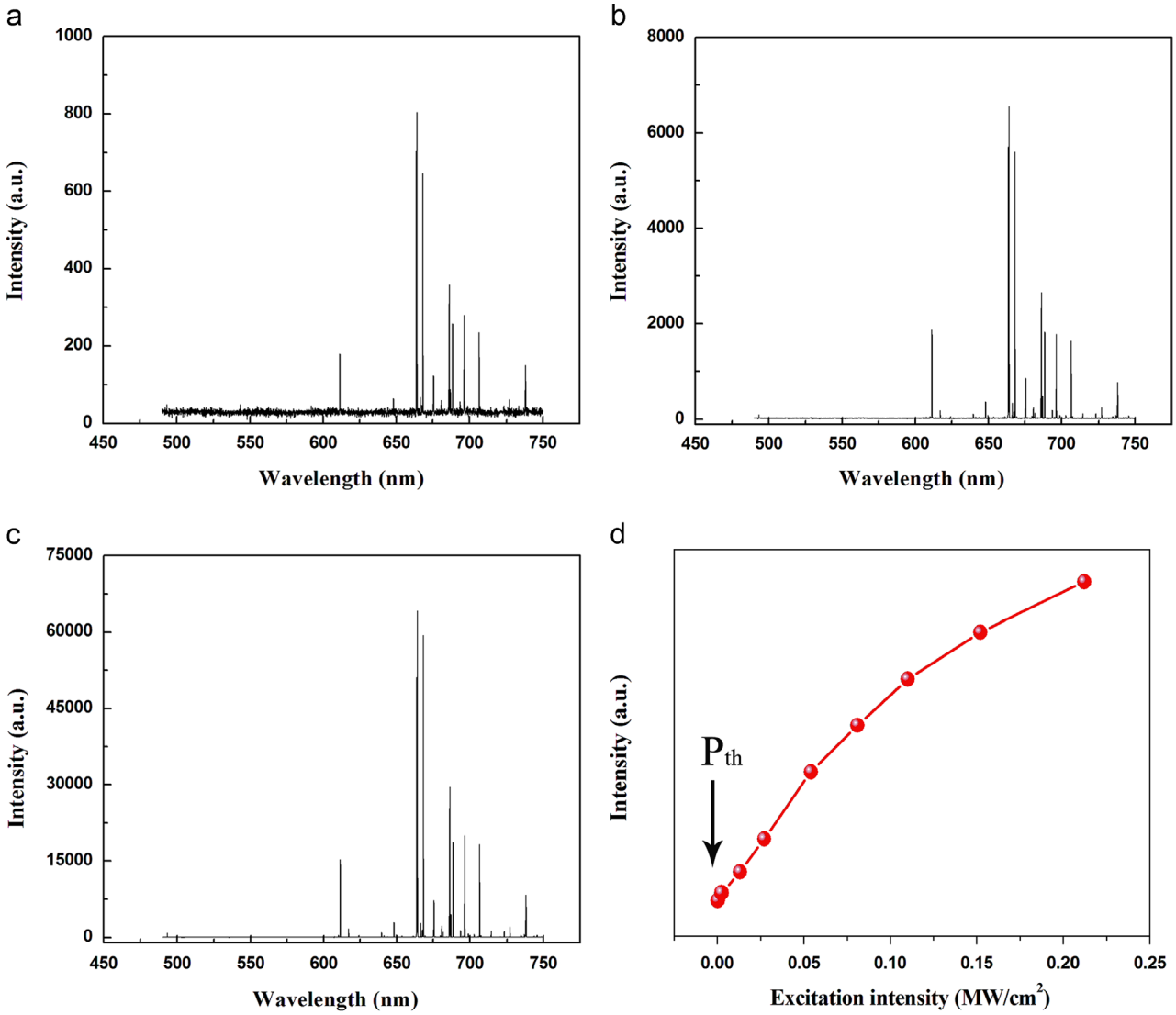

Fig. 2. Lasing spectra of HOPG versus different pump power. (a) 1‰, (b) 1%, and (c) 100%. The maximum output power of Ar^+ laser is 15 mW. (d) The emission intensity of the spectrum as a function of the pump power intensity.