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Microfluidic-Assisted Assembly of Fluorescent Nanodiamonds for Precise Temperature Measurement

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Summary:

In all-optical quantum sensing with fluorescent nanodiamonds (FNDs), enhancing the optical signal from excited FNDs is essential. Here, we demonstrate that the template-assisted assembly of FNDs allows the positioning of aggregated FNDs and improves the precision of temperature measurement. Our assembly process combines a polydimethylsiloxane microfluidic device with vertical through-holes and degas operation, enabling not only the aggregation of FNDs for increasing fluorescence intensity but also their high yield positioning over millimeter-scale areas.

Keywords: nanodiamonds, nitrogen vacancy center, temperature sensor, templated self-assembly

Background

Fluorescent nanodiamonds (FNDs) with nitrogen vacancy (NV) centers function as quantum sensors for measuring temperature, magnetic fields, and electric fields. FNDs are suitable for temperature sensors owing to their superior thermal conductivity, chemical resistance and optical durability [1,2]. To achieve precise all-optical quantum temperature sensing with a temperature-dependent fluorescence spectrum, a high fluorescence intensity is required. Although boosting the excitation laser power is one method, it might cause objects heating. Instead of this approach, increasing the NV center density in FNDs has been proposed; however, it is limited to enhancing the fluorescence intensity because of a concurrent rise in other defects [3]. We propose a templated self-assembly approach to form aggregated FNDs for fluorescence intensity enhancement, that also address the particle-to-particle variance in fluorescence properties [1]. By employing a polydimethylsiloxane (PDMS) microfluidic device, this microfluidic-assisted assembly method achieves both precise measurement and patterning of FND clusters on a substrate.

Description of the New Method or System

In this assembly method (Fig. 1), FND clusters were formed on the substrate by drying the FND suspension and aggregating the FNDs along the vertical direction of microchannels. To achieve this process with a single supply of a few microliters of the FND suspension, we employ a PDMS microfluidic device that consists of two layers: a pattern layer with vertical through-holes defining the FND cluster shape/position and a channel layer with channels supplying the FND suspension to the through-holes. When closely attached to the substrate, the pattern layer creates dead-end channels via the through-holes. Here, conducting a degas operation after introducing the FND suspension into the channels would result in the generation of bubbles from the PDMS due to its high porosity, preventing the channels from being completely filled with the FND suspension. Thus, a 1-h degas operation is performed before supplying the FND suspension. When a device is returned to atmospheric pressure and a quick supply of small sample volume of the suspension is performed, any residual gas in the through-holes is absorbed into PDMS, generating a driving force for the suspension to fill the dead-end channels [4]. Then, the FND suspension are dried in the through-holes to aggregate FNDs. Finally, peel off the device from the substrate. This method allows the formation of FND clusters with large numbers of NV centers to improve measurement precision through increased fluorescence intensity and positioning FND clusters on the substrate over millimeter scales.

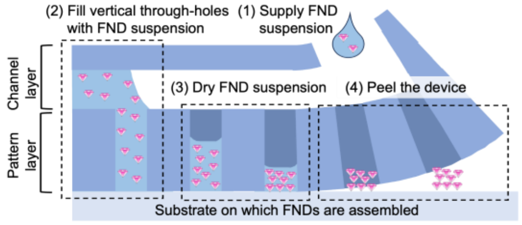

Fig.1. Conceptional illustration of the microfluidic-assisted assembly of FNDs and process sequence.

Results

Potentials of the proposed assembly process was demonstrated by arraying FND clusters. The PDMS microfluidic device was fabricated using standard soft lithography (Fig. 2). The pattern layer has 10-µm diameter through-holes with 20-µm spacing underneath the channel layer with an area of 16 mm^2. Using FNDs with an average size of 750 nm, FND cluster sites were arrayed on PDMS substrate with a yield of over 99% under FND suspension concentration of 0.05, 0.1, and 0.2 w/v% (Fig. 3a). With increasing suspension concentration, a larger amount of FNDs aligned in the vertical direction (Fig. 3b). The height of the FND clusters obtained with a concentration of 0.2 w/v% was double compared with those at 0.1 w/v%.

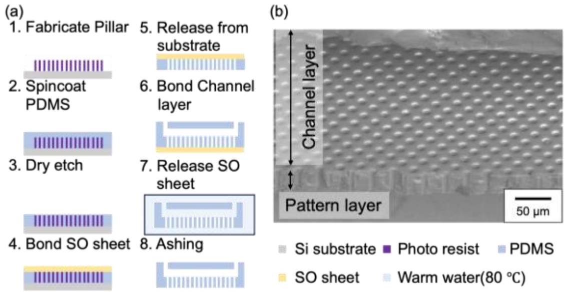

Fig. 2. (a) Device fabrication process. (b) SEM image of the cross section of the microfluidic device.

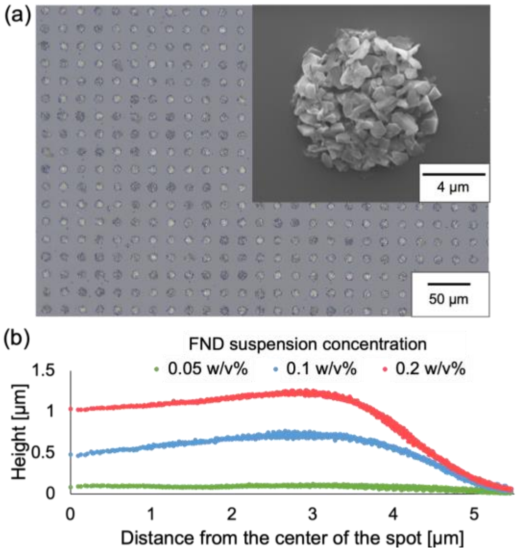

Fig. 3. (a) Optical and SEM images of the FNDs array on the PDMS substrate. (b) Height profile of the FND clusters for different FND suspension concentrations.

Next, the temperature dependence of the peak wavelength of the zero-phonon line (ZPL) of the NV center [1] was evaluated for temperature sensors. At a controlled temperature on a hot plate, the FND cluster array's ZPL peak wavelength shifted linearly by 9.4 ± 2.5 pm/K (n = 10 clusters), demonstrating its applicability for temperature sensors (Fig.4).

Finally, the fluorescence intensity and measurement precision were investigated. Figure 5 indicates enhanced fluorescence intensity with higher suspension concentrations, i.e., a larger number of FNDs in the clusters. Furthermore, the sensitivity, which indicates the precision of the temperature measurement in 1-s, was improved by increasing the fluorescence intensity. The FND clusters produced by the 0.2 w/v% suspension concentration showed approximately twice the sensitivity compared to the 0.1 w/v% concentration. These experimental results demonstrate that the microfluidic-assisted assembly process enables not only the positioning of FND clusters on the substrate but also the enhancement of fluorescence intensity and sensitivity. Therefore, this fabrication process for temperature sensors with FNDs can also be used to develop various quantum sensors.

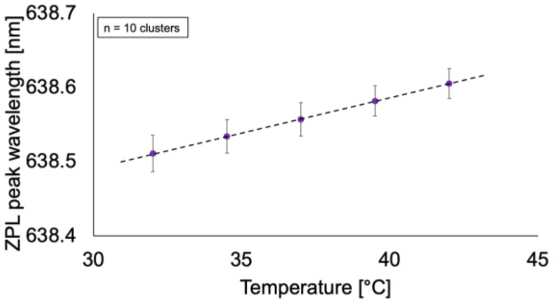

Fig. 4. Temperature dependence of ZPL peak wavelength. Error bars are standard deviation of 10 FND clusters.

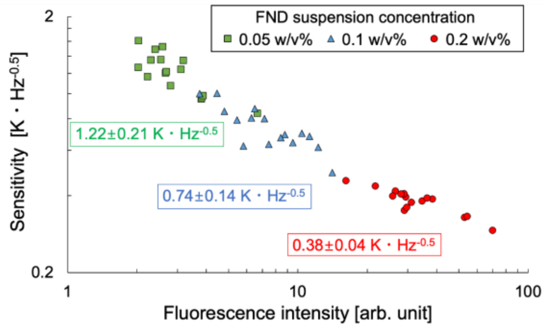

Fig. 5. Relationship between fluorescence intensity and sensitivity for different suspension concentrations. Values in the graph are the average sensitivity obtained at each concentration using 0.5 mW laser.

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