Supplemental information

Ultrasensitive detection of exosomal miRNA
with PMO-graphene quantum dots-functionalized
field-effect transistor biosensor

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**Table S1.** Sequences and involved oligonucleotide probes. Related to STAR Methods.

| Probe name       | Sequences (5’- 3’)                                      |
|------------------|--------------------------------------------------------|
| PMO probe        | NH₂-TCAACATCAGTCTGATAAGCTA                              |
| PMO-Cy5 probe    | NH₂-TCAACATCAGTCTGATAAGCTA-Cy5                        |
| miRNA21          | UAGCUUAUCAUGACUGAUUGUUGA                                |
| OM-miRNA21       | UAGCUUAUCAUGACUGAUUGUUGA                                |
| TM-miRNA21       | UUGCUCUAUCAUGACUGAUCAUUGA                               |
| miRNA10b         | UACCCUGUAGAACCAGAAUUGUG                                  |
**Table S2.** Performance comparison of different sensing methods for detection of miRNA. Related to Figure 4.

| Method                                                                 | Detection Range | LOD   | Reference               |
|------------------------------------------------------------------------|-----------------|-------|-------------------------|
| AuNPs decorated Graphene FET sensor                                    | 1 fM to 100 pM  | 0.37 fM | Li et al., 2021         |
| Flexible graphene FET sensor                                           | 10 fM to 100 pM | 10 fM  | Gao et al., 2020        |
| PASE modified Graphene nanosheet FET sensor                           | 10 fM to 10 nM  | 10 fM  | Kim et al., 2021        |
| Graphene Oxide/Graphene Layered Structure FET sensor                  | 10 fM to 100 pM | 10 fM  | Huang et al., 2021      |
| CRISPR/Cas12a mediated electrochemiluminescence sensor                | 1 fM to 10 nM   | 0.331 fM | Wang et al., 2021      |
| Microgels Fluorescence sensor                                         | 10 fM to 10 nM  | 10 fM  | Caputo et al., 2019     |
| liquid-phase SERS biosensor                                           | 100 fM to 5 nM  | 1.45 fM | Wu et al., 2021         |
| Surface acoustic wave sensor                                           | 0.5 nM to 5 nM  | 0.19 nM | Cogal et al., 2021      |
| **GQDs-PMO functionalized Graphene FET sensor**                       | **100 aM to 1 nM** | **0.085 fM** | This work             |
**Figure S1.** Raman spectra of RGO and RGO-PLL. Related to Figure 3.

**Figure S2.** AFM images before and after functionalization of GQDs-PMO. (a) The PLL-RGO surface. (b) The GQDs-PMO modified surface. Scale bar, 200 nm. Related to Figure 3.
Figure S3. Optimization of experimental parameters. (a) The ratio of GQDs to PMO (PMO concentration from 0.5 to 2.5 μM). (b) The concentration of the PLL film dropped on the RGO-FET chip surface. (c) Hybridization time for GPPR-FET and target miR21. (d) Hybridization temperature between target miR21 and GPPR-FET. Error bars represent the standard errors (n = 3). Related to Figure 4.
**Figure S4.** Response of the PMO-functionalized G-FET sensor to varying concentrations of miR21 (from 1 fM to 1 nM). Error bars represent the standard errors (n = 3). Related to Figure 4.

**Figure S5.** Stability of GPPR-FET sensor. Error bars represent the standard errors (n = 3). Related to Figure 4.
Figure S6. The 7 days stability of GPPR-FET sensor. Error bars represent the standard errors (n = 3). Related to Figure 4.