

## Comment on “Plasmon-Enhanced Ultraviolet Photoluminescence from Hybrid Structures of Graphene/ZnO Films”

In a recent Letter, Hwang *et al.* [1] observed enhanced ultraviolet photoluminescence from hybrid structures of graphene/ZnO films. The authors attributed this phenomenon to the resonant excitation of graphene plasmon by an analogy to similar behavior of surface plasmon induced at metal/ZnO interfaces. Here we think that this analogy is not appropriate and claim that the influence of plasmons alone cannot explain all the experimental results.

(i) They have pointed out the distinctive feature of graphene plasmon from surface plasmon (SP) close to the high frequency limit with increased momentum: while the SP frequency from the metal/dielectric interface saturates, graphene plasmon maintains a linear dispersion relation up to higher energies. Here, the authors have adopted the formula by the random phase approximation and neglected the frequency-dependent dielectric function for ZnO. However, as Jablan and Yu have shown [2,3], the dispersion relation is derived by calculating the electromagnetic field in the air/graphene/dielectric system. If we take the optical constants of ZnO [4] and employ the semiclassical model for conductivity of graphene, we can give a numerical result for the dispersion curve of the air/graphene/ZnO system. The distinctive feature is almost the same as Hwang *et al.* presented, but a difference in the physical regime should be noticed: for the metal/ZnO interface, the phase velocity  $\omega/q$  of SP is comparable to the velocity  $c$  of light in vacuum, so SP can easily couple with the free electromagnetic field; but in the case of the air/graphene/ZnO system,  $\omega/q \ll c$ , and the plasmon mode is in the nonretarded region [5]. In this region, momentum mismatch between graphene plasmons and photons is very large, the transformation efficiency into photons would be very low. In fact, the in-plane wave number of the plasmon mode is on the order of  $10^9 \text{ m}^{-1}$ , while the photon wave number is on the order of  $10^7 \text{ m}^{-1}$ . Thus graphene plasmons can transform into propagating photons only by scattering with the small surface corrugation ( $\sim 1.5 \text{ nm}$ ) of ZnO. Unfortunately, the size of this kind of corrugation cannot be proven by the experiment.

(ii) Another important feature of SP is that its field component perpendicular to the metal/dielectric interface is evanescent, which decays exponentially. For the conventional plasmon mode, the skin depth of dielectric material is on the order of half the wavelength of light involved [6], about a hundred nanometers in the visible and ultraviolet region. While in the case of the air/graphene/SiO<sub>2</sub>/ZnO system, the value of the wave vector perpendicular to the interface is on the order of  $10^9 \text{ m}^{-1}$ . Thus the skin depth into the spacer is about 1 nm, much smaller than the value for SP. If enhanced photoluminescence is expected, the thickness of the spacer should be on the order of 1 nm, the Purcell factor calculated from [7] confirms this

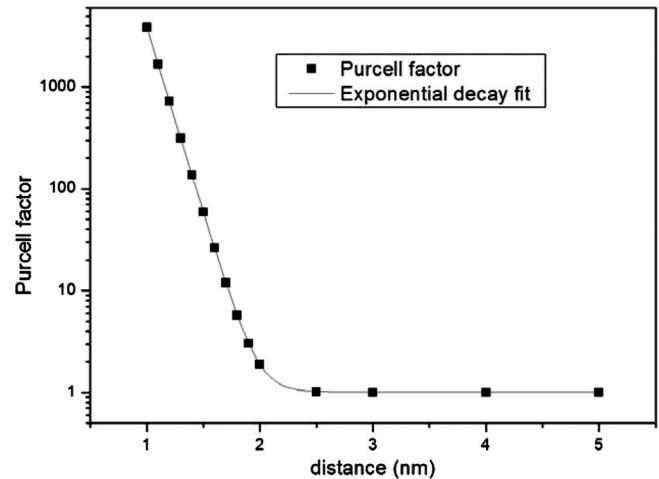


FIG. 1. Purcell factor at the band gap energy of ZnO ( $\sim 3.3 \text{ eV}/376 \text{ nm}$ ) as a function of the distance from light emitting area to graphene using SiO<sub>2</sub> as a spacer.

conclusion as shown in Fig. 1. Since the skin depth here cannot explain the data in Fig. 3(a) in [1], we think that plasmon resonance alone cannot account for the observed experimental results.

(iii) In our opinion, the plasmons should be excited by light incident from the ZnO side of the graphene/ZnO system in order to observe the plasmon coupling effect directly in PL experiments. Since the absorption coefficient of ZnO reaches  $17 \mu\text{m}^{-1}$  at 325 nm, ZnO films with a thickness of less than 50 nm should be employed so that the laser beam can excite the entire film [8]. We also suggest that time-resolved photoluminescence with this geometry should be done to study the different spontaneous emission rates of a bare ZnO film and a hybrid structure of graphene/ZnO film. This would help us to investigate the recombination mechanism behind this plasmon related enhancement.

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Wei Yang and Xiaodong Hu

State Key Laboratory for Artificial Microstructure and Mesoscopic Physics and School of Physics, Peking University, Beijing 100871, China

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- [1] Sung Won Hwang, Dong Hee Shin, Chang Oh Kim, Seung Hui Hong, Min Choul Kim, Jungkil Kim, Keun Yong Lim, Sung Kim, Suk-Ho Choi, Kwang Jun Ahn, Gunn Kim, Sung Hyun Sim, and Byung Hee Hong, *Phys. Rev. Lett.* **105**, 127403 (2010).

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- [2] Marinko Jablan, Hrvoje Buljan, and Marin Soljaccaronicacute, *Phys. Rev. B* **80**, 245435 (2009).
- [3] V. Bludov Yu *et al.*, *Europhys. Lett.* **92**, 68001 (2010).
- [4] P.L. Washington, H. C. Ong, J. Y. Dai, and R. P.H. Chang, *Appl. Phys. Lett.* **72**, 3261 (1998).
- [5] J.M. Pitarke *et al.*, *Rep. Prog. Phys.* **70**, 1 (2007).
- [6] William L. Barnes, Alain Dereux, and Thomas W. Ebbesen, *Nature (London)* **424**, 824 (2003).
- [7] I. Gontijo, M. Boroditsky, E. Yablonovitch, S. Keller, U.K. Mishra, and S.P. DenBaars, *Phys. Rev. B* **60**, 11 564 (1999).
- [8] C.W. Lai, J. An, and H.C. Ong, *Appl. Phys. Lett.* **86**, 251105 (2005).