The unusual Raman and optical properties of black phosphorus

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Abstract

In this talk, I will summarize recent results on the Raman and optical characterization of black phosphorus (BP), which show interesting unusual features [1-3]. It will be shown that the angular dependence of the polarized Raman spectrum obtained in experiments can only by adjusted by group theory if the components of the Raman tensor are taken to be complex [1]. This feature is attributed to BP's complex dielectric tensor, from which the Raman tensor is derived. The structural and vibrational properties of the atoms near different BP edges were also studied using polarized Raman spectroscopy [2]. The observed results, likewise, exhibit deviations from group theory's predictions, with unexpected Raman modes appearing only at the edges. The observed features could be reproduced by density functional theory (DFT) simulations, which showed them to be the result of atomic rearrangements at the edges. Finally, the nonlinear optical properties of BP were investigated via third harmonic generation pumped by a near infrared femtosecond laser [3]. It was found that few-layer BP yields third harmonic intensities that are 1 to 2 orders higher than obtained in bulk samples, and 3 orders of magnitude higher than achieved in graphene. This nonlinearity enhancement is attributed to resonances with 2D exciton states. In addition to direct mechanical exfoliation, few-layer BP was also obtained via laser thinning with the third-harmonic generation pump. In this case, the optical nonlinearity could be probed while thinning occurred, which can be exploited for mapping the light-crystal interaction as a function of BP thickness.

References

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