Comment on "Origin of the Stokes Shift: A Geometrical Model of Exciton Spectra in 2D Semiconductors"

In a recent Letter, Yang *et al.* [1] noted that a plot of the "Stokes shift" S (the shift between optical absorption and emission) in a large number of quantum well samples falls approximately on a universal curve $S=2W/\sqrt{6\pi\ln 2}$, where W is the full width of the absorption linewidth. The authors then develop a statistical model of the well width fluctuations, based on a random potential fluctuation function, and calculate the energy distribution of the exciton states, which corresponds to the optical absorption. Within this distribution of states, some are local minima (i.e., all neighboring regions of the quantum well are narrower and correspond to higher confinement energies). Yang *et al.* calculate the distribution of these "local-minimum" states, and suggest that all recombining excitons are randomly distributed among them. The shift between the peak in the distribution of all the exciton states, versus the peak of the local-minimum states would then be the origin of the universal curve for the Stokes shift.

Within the approximations stated in their Letter, Yang *et al.* have a valid method for calculating the distribution of local-minimum states. However, numerous experiments have demonstrated that radiating excitons in direct gap quantum wells are not distributed randomly among these states, so that the model of Yang *et al.* is only an approximation to the actual situation.

Yang *et al.* state that "because the exciton lifetimes are much larger than the relaxation time for the emission of phonons, we assume that the exciton will have relaxed into a local minimum of the potential. . . when it decays. The luminescence spectrum therefore reflects the distribution of heights of local minima." In fact, numerous studies of spectral diffusion of excitons in GaAs/AlGaAs quantum wells have shown that the diffusion and cooling continue throughout the subnanosecond exciton lifetime [2–6]. When time resolved photoluminescence spectra are measured [2,3], the Stokes shift is found to continuously increase with time. The shape of the emission spectrum of the excitons is dominated by dynamic effects; *the excitons do not reach a stable, quasiequilibrium distribution before recombination occurs.* A quasiequilibrium distribution has only been observed for quantum well excitons which have a lifetime much greater than the subnanosecond lifetimes of excitons in direct gap quantum wells. For example [7], the long-lived (50 nsec) indirect excitons in a coupled quantum well have been shown to be in quasiequilibrium. Here, however, exciton-exciton interactions were shown to be important so that the excitons did not occupy the local energy minima randomly.

A static model, such as that proposed by Yang *et al.*, cannot be exact for the dynamic situation of excitons in direct gap quantum wells, since the excitons do not have sufficient time to come to thermal quasiequilibrium. A complete model to calculate the Stokes shift must include the effects of drift and diffusion [2,6], phonon emission [5,8], and lifetime.

The preceding paragraph summarizes the main point of this Comment. However, it is also worth commenting on a central assumption in the calculation of Yang *et al.* of the local-minimum states. They assume a random variation in well width, which allows them to take Gaussian distributions. In real quantum wells, well width fluctuations are unlikely to be random. As examples, the growth of quantum wells with multiple peak emission spectra through simple growth-interruption techniques [9] and the growth of quantum wires on vicinal GaAs substrates [10] manifestly demonstrate nonrandom placement of atoms on available lattice sites during epitaxial growth.

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