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Reply to "Comment on 'Radiative and nonradiative recombination of bound excitons in GaP:N. I. Temperature behavior of zero-phonon line and phonon sidebands of bound excitons' and 'Radiative and nonradiative recombination of bound excitons in GaP:N. IV. Formation of phonon sidebands of bound excitons'"

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For excitons trapped at deep centers such as N in GaP, the phonon sidebands are composed of a phonon-assisted momentum-conserving indirect transition and a multiphonon transition. As a result, the temperature dependence of phonon sidebands can be different from that of the corresponding zero-phonon lines. We suggest distinguishing the term "replica" due only to multiphonon processes from the term "sideband." A full-kinetic-analysis method is suggested for use in unraveling the complicated temperature dependence of the luminescence of bound excitons in GaP:N.

The main point of the preceding Comment of Ge *et al.*[1] and the Comment of Sturge and Cohen[2] on our recent papers[3–5] on the radiative and nonradiative recombination of bound excitons in GaP:N is whether or not the temperature behavior of zero-phonon lines (ZPL) is the same as that of the corresponding phonon sidebands (PS). Ge *et al.*[1] claimed that the TA and LA PS of NN_3 had been mistaken for the LO PS of NN_4–NN_6 in our papers,[3,5] leading to an anomalous temperature behavior of the latter.

In our work, we did notice that there were some broad weak bands near $NN^*_{4,5,6}$ (LO PS of $NN_{4,5,6}$) when the temperature was low enough. We did follow closely the evolution of these broad bands with temperature and deducted them when we deconvoluted the spectra. The deconvolution of spectrum always produces some error, especially for these unresolved broad bands. To avoid such difficulties brought on by a deconvolution of the spectrum, we focused our attention instead on NN_1 and NN_3 and their PS NN^*_1 and NN^*_3, where the ZPL (including NN_1 and NN_3) up to 170 K and PS (including NN^*_1 and NN^*_3) over 135 K, to convince ourselves that their temperature behavior is really different.

Concerning the TA- and LA-phonon sidebands of ZPL, we believe they are in fact due to the indirect recombination on the basis of momentum-conservation considerations. The TA- and LA-phonon energies are nonzero only when their wave vector **K** is not at the Γ point in the Brillouin zone (BZ). For the TA and LA PS, the momenta of the phonons involved are nonzero. Consequently, the initial state must be an electron at any non-Γ point in the BZ (with a hole at the Γ point). That is why the sidebands due to acoustic phonons are always much broader than the LO(Γ) PS. Therefore, we should distinguish between these two different kinds of process, the former being due to a phonon-assisted momentum-conserving (MC) indirect recombination and the latter being due to multiphoton transition related to the configuration-coordinate (CC) coupling. Both may be called phonon sidebands, but only the latter should be considered as a replica or satellite line. For a long time, these two processes have been studied separately in different systems. The isoelectronic-impurity-N-doped GaP provides us with a system in which both MC and CC processes coexist. For GaP:N, the wave function of electrons trapped at N is strongly localized at the X point of the BZ.[7] The indirect recombination process cannot therefore be neglected. We should emphasize that the direct transition cannot take place unless the wave vector **K** of the phonons involved is zero. Therefore, the Huang-Rhys parameter S can be used only for the CC component of the sidebands. In our opinion, in some cases such as GaP:N, one cannot simply consider the temperature dependence of the PS as being that of the ZPL as has been done by Sturge, Cohen, and Rodgers in Ref. 6.

Dai, Gundersen, and Myles[8] treated the MC process thoroughly. The MC process includes phonon-assisted transitions from essentially all wave vectors in the BZ and not just certain critical points such as the X and Γ points.[5,8] Our experimental results show that the indirect recombination assisted by LO(X) phonons is much stronger than that assisted by the acoustic phonons (cf. Fig. 1 of Ref. 4). For the optic PS, therefore, we should consider the CC transition and the MC recombination at the same time. That is the main point of our two-component (TC) model.

Ge *et al.*[1] claimed that our experimental results—the ratio of the LO PS to their corresponding ZPL is strongly temperature dependent—imply an anomalous strong temperature dependence of the Huang-Rhys parameter S. One knows that in the CC model, S is temperature independent (precisely speaking, is very weakly temperature dependent). In our opinion, the LO PS in GaP:N comes from not only the CC recombination. We did not consider the ratio (R) PS/ZPL as Huang-Rhys parameter S. We have also never meant that the variation of R with temperature implies the S is temperature dependent.

Sturge and Cohen[2] pointed out that they pumped each bound exciton selectively, so that only one exciton was observed at a time. As already pointed out in our previous work,[4] this interpretation is incorrect, because of the existence of the energy transfer between the isolated N and various N-N pair centers. We excited selectively the exciton trapped at NN center and observed the luminescence from other shallower NN_i center. Having considered this point, we suggest performing a kinetic analysis in which various radiative and nonradiative (including the energy transfer) processes were involved. It may be a complicated procedure, but a necessary one. Having done so, we obtained almost the same value for the activation energy by using the experimental data from the above-band-gap excitation or the selective excitation. We have found that under selective excitation, the population of excitons at various NN_i centers changes with temperature and excitation density. This will be the subject of a forthcoming paper.

Ge *et al.*[1] also claimed that the shallower centers NN_4-NN_6 quench faster than the deeper center NN_3 and the thermally activated energy transfer from the shallower centers to the deeper ones further enhances the PL intensity from the NN_3 center. We disagree with that. First, that the shallower centers quench faster than the deeper center is valid only for relatively low temperatures. When the temperature is high enough, because of the larger activation energy of the centers, the quench of deep centers will be faster than the shallower centers. Second, we cannot simply say that the PL of NN_3 will be enhanced owing to the energy transfer from NN_4-NN_6 to NN_3, considering the energy transfer mentioned above between N and N-N pair centers and that the energy can be transferred not only from shallower centers to deeper ones but also from deeper centers to shallower ones.[4] In order to understand the effect of the above complicated energy transfer processes on the photoluminescence of each NN_i, we believe a full kinetic analysis will be required.[3]

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