Comments

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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"

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X. Zhang et al. [Phys. Rev. B 41, 1376 (1990)] argue that our data on the thermal quenching of excitons bound to nitrogen pairs in GaP [Sturge et al., Phys. Rev. B 15, 3169 (1977)] are incorrect for some pairs, and, from this, draw far-reaching conclusions about the exciton-phonon interaction. In this Comment we point out that the data on which Zhang et al. base their argument, which were previously reported by some of the authors in Chang (Zhang) et al. [Scientia Sinica (Ser. A) 25, 942 (1982)], have already been shown to be incorrect, and that our simple model accounts for all the reliable data.

In a recent paper X. Zhang et al.¹ (hereinafter referred to as ZDHB) criticize our work² on the thermal quenching of excitons bound to nitrogen (N) and nitrogen pairs (NN_i, where the nitrogen atoms form an *i*th nearestneighbor pair) in GaP. In Ref. 2 we determined the exciton population as a function of temperature by monitoring the phonon sideband (PS) intensity when the nophonon (NP) line was selectively excited. ZDHB argue that the I(PS)/I(NP) ratio is strongly temperature dependent, so that our method gives incorrect results.

The data on the temperature dependence of the NN₁ NP intensity given by ZDHB (their Fig. 3) are, within experimental error, in agreement with our data² on the PS.³ ZDHB report activation energies of 133 ± 10 , 42 ± 4 , and 22 ± 4 meV, compared with our 115, 37, and 20 meV (no error bars were given, but they were of the same order). The only difference is that in Ref. 2 it was shown that the 20-meV activation only occurs at high intensity, when hole traps are saturated (the hole traps can be emptied, and the 20-meV component eliminated, by illuminating with infrared radiation). Figure 7 of Ref. 2 shows the limiting cases of complete saturation and none: at intermediate levels all three activation energies are seen, as in ZDHB.

Comparison of our data with those of ZDHB show that we differ on the temperature dependence of the PS intensity, not on that of the exciton population; this is a difference of data, not of interpretation, as implied by ZDHB. The essential difference between our technique and that of ZDHB is that we pumped each bound exciton selectively, so that only one exciton was observed at a time, while ZDHB, by pumping above the band gap, excited all excitons simultaneously. Although it is not so stated in Ref. 2, we did, in fact, check for a possible temperature dependence of the I(NP)/I(PS) ratio in the cases of the NN₁ and of the N excitons and found that so long as self-absorption in the NP line is avoided, the ratio is independent of temperature in these two limiting cases.

Zheng and his collaborators^{4,5} compared the spectra obtained with selective and band-gap pumping for excitons bound to several different pairs and found that, for selective pumping into a PS, so that only one exciton is excited at a time, the I(PS)/I(NP) ratio in luminescence is indeed independent of T. For above-band-gap pumping, on the other hand, the PS's from different excitons overlap, and the I(PS)/I(NP) ratio can appear to be strongly temperature dependent if great care is not taken to identify the individual PS correctly. Note that the relative strengths of the different NN_i excitons depend strongly on temperature, so that attribution of a PS to the wrong exciton can lead to an apparent, but spurious, temperature dependence of the I(PS)/I(NP) ratio, as reported by ZDHB. As remarked above, self-absorption in the NP line can also lead to a spurious temperature dependence, since the linewidth depends on temperature.

In a subsequent paper,⁶ three of the authors of ZDHB

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draw far-reaching theoretical conclusions from their putative observation of a strong temperature dependence of the I(PS)/I(NP) ratio in this system. It is not the purpose of this Comment to discuss the validity of this theory, but to point out that it appears to have been developed to account for an effect for which there is no real experimental evidence. We are grateful to Professor Weikun Ge and Yong Zhang for drawing our attention to Ref. 1, and for helpful discussions. The work at Dartmouth was supported by the U.S. Department of Energy under Grant No. DEFG-02-ER-45330. Some of this work was done in collaboration with K. F. Rodgers, now deceased.

- ¹X. Zhang, K. Dou, Q. Hong, and M. Balkanski, Phys. Rev. B 41, 1376 (1990) (ZDHB); see also H. Chang (X. Zhang), C. Hirlimann, M. Kanehisa, and M. Balkanski, Sci. Sin. (Ser. A) 25, 942 (1982).
- ²M. D. Sturge, E. Cohen, and K. F. Rodgers, Phys. Rev. B 15, 3169 (1977).
- ³This agreement is rather surprising. We found (see, for example, Fig. 2 of Ref. 2) that even when it was possible to sort out the various overlapping spectra, above-band-gap pumping gave complex results which were difficult to interpret, be-

cause of the temperature dependence of the capture cross sections.

- ⁴Y. Zhang, Q. Yu, J. Zheng, B. Yan, B. Wu, W. Ge, Z. Xu, and J. Xu, Solid State Commun. **68**, 707 (1988). Note that this paper refers to "mixing" of the sidebands from different excitons when pumped above the band gap; what is actually meant is "overlap." ZDHB do not refer to this paper, nor to Ref. 5.
- ⁵J. Zheng and Y. Zhang, Sci. Sin. 29, 862 (1986); 29, 870 (1986).
- ⁶Q. Hong, X. Zhang, and K. Dou, Phys. Rev. B 41, 2931 (1990).