Bieexcitons in GaN and AlGaN epitaxial layers

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In highly excited semiconductors, it is now well established that two excitons interact attractively and form a bound state known as a bie exciton (excitonic molecule). There has been much work to date on bie exciton formation in a large variety of semiconductors. Such a characteristic phenomenon of a dense excitonic system has been studied mainly in wide-gap semiconductors because of the advantages of excitonic nature. From the viewpoint of exciton engineering, it is important to clarify nonlinear optical properties of dense excitonic phenomena. It has been pointed out that recombination processes of dense excitons in wide-gap semiconductors contribute to the formation of optical gain. Nitride-based semiconductors are among the most useful materials for applications to short-wavelength optoelectronic devices. Nitride-based semiconductors are also expected to possess large excitonic effects for optical transitions. Recent progress in growth techniques has enabled us to obtain GaN samples of high optical quality and to explore intrinsic recombination processes of dense excitons. We first revisit the optical properties of bie excitons in GaN. Since 1996, several reports have mentioned bie excitonic effects in GaN. These work consistently evaluated the bie exciton binding energy of 5.6 meV. Then, the ratio of the bie exciton binding energy (BX) relative to the exciton binding energy (BX) is estimated to be \( \frac{BX}{BX}=5.6/5.25=0.22 \). This value is found to be in close agreement with the ratio for other wide-gap semiconductors such as ZnS (BX/BX=8.0/57=0.23) and ZnSe (BX/BX=3.5/17=0.21). We have recently studied the nonlinear optical properties of excitons and bie excitons in GaN by means of time-resolved nonlinear luminescence spectroscopy, which is based on an excitation correlation technique. This spectroscopic technique, called population mixing, directly provides information about nonlinear carrier dynamics in photoexcited semiconductors. We observed a superlinear signal of nonlinear bie exciton luminescence, which indicated an enhancement of luminescence efficiency. The enhancement originated from the higher-order nonlinearity with respect to the density of bie excitons. The most probable physical process was attributed to the stimulated emission of bie exciton luminescence; that is, the formation of optical gain due to bie exciton decay processes. Next, we present the optical properties of localized bie excitons in Ga-rich AixGai-xN ternary alloy epitaxial layers. The effect of localization due to alloy disorder on bie excitons has been studied by means of photoluminescence excitation (PLE) spectroscopy. In order to evaluate the binding energy of bie excitons as a function of alloy composition, we observed a two-photon bie exciton resonance as well as an exciton resonance in the PLE spectrum of bie excitons. The two-photon bie exciton resonance indicates the direct creation of bie excitons from a ground state by a two-photon absorption process. On the basis of the energy separation between the exciton resonance and the two-photon bie exciton resonance, the bie exciton binding energy was estimated to be 8.0, 10.4, 12.6, and 15.2 meV for the sample with the aluminum composition of x=0.019, 0.038, 0.057, and 0.077, respectively. A strong enhancement of the bie exciton binding energy was observed while the exciton binding energy remained nearly unchanged in this Ga-rich range. Furthermore, we defined a Stokes shift of bie excitons in order to evaluate the degree of bie exciton localization quantitatively. If bie excitons are free from localization, bie exciton luminescence should appear at the lower-energy side from an exciton resonance by a bie exciton binding energy. However, the observed peak position of the bie exciton luminescence in the ternary alloys was located at the lower-energy side from the expected position of free bie excitons. We consider that the energy separation between the expected peak position of free-bie exciton luminescence and the observed peak position of the bie exciton luminescence indicates the degree of bie exciton localization. Therefore, we propose the energy separation as a Stokes shift of bie excitons. Both the Stokes shift of bie excitons and the Stokes shift of excitons were found to increase almost linearly with exciton linewidth. As a result, we obtained the ratio of the bie exciton Stokes shift (SXX) relative to the exciton Stokes shift (SXX) \( \frac{SXX}{SXX}=0.81 \). The strong enhancement of the bie exciton binding energy resulted from the bie exciton localization due to alloy disorder.