Biexcitons 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 biexciton (excitonic molecule). There has been much work to date on biexciton 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 biexcitons in GaN. Since 1996, several reports have mentioned biexcitonic effects in GaN. These work consistently evaluated the biexciton binding energy of 5.6 meV. Then, the ratio of the biexciton binding energy (BXX) relative to the exciton binding energy (BX) is estimated to be BXX/BX=5.6/25=0.22. This value is found to be in close agreement with the ratio for other wide-gap semiconductors such as ZnS (BXX/BX=8.0/37=0.22) and ZnSe (BXX/BX=3.5/17=0.21). We have recently studied the nonlinear optical properties of excitons and biexcitons 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 biexciton luminescence, which indicated an enhancement of luminescence efficiency. The enhancement originated from the higher-order nonlinearity with respect to the density of biexcitons. The most probable physical process was attributed to the stimulation of biexciton luminescence; that is, the formation of optical gain due to biexciton decay processes. Next, we present the optical properties of localized biexcitons in Ga-rich AlxGa1-xN ternary alloy epitaxial layers. The effect of localization due to alloy disorder on biexcitons has been studied by means of photoluminescence excitation (PLE) spectroscopy. In order to evaluate the binding energy of biexcitons as a function of alloy composition, we observed a two-photon biexciton resonance as well as an exciton resonance in the PLE spectrum of biexcitons. The two-photon biexciton resonance indicates the direct creation of biexcitons 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 biexciton resonance, the biexciton 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 biexciton binding energy was observed while the exciton binding energy remained nearly unchanged in this Ga-rich range. Furthermore, we defined a Stokes shift of biexcitons in order to evaluate the degree of biexciton localization quantitatively. If biexcitons are free from localization, biexciton luminescence should appear at the lower-energy side from an exciton resonance by a biexciton binding energy. However, the observed peak position of the biexciton luminescence in the ternary alloys was located at the lower-energy side from the expected position of free biexcitons. We consider that the energy separation between the expected peak position of free-biexciton luminescence and the observed peak position of the biexciton luminescence indicates the degree of biexciton localization. Therefore, we propose the energy separation as a Stokes shift of biexcitons. Both the Stokes shift of biexcitons and the Stokes shift of excitons were found to increase almost linearly with exciton linewidth. As a result, we obtained the ratio of the biexciton Stokes shift (SXX) relative to the exciton Stokes shift (SX), SXX/SX = 0.81. The strong enhancement of the biexciton binding energy resulted from the biexciton localization due to alloy disorder.