Biexcitons and their dephasing processes in ZnO

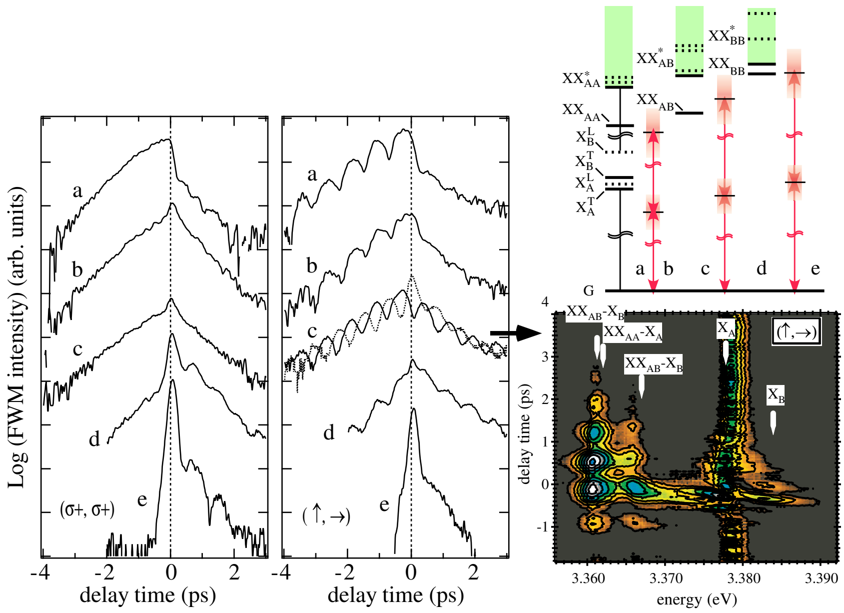
S. Adachi[1], K. Hazu[2], T. Sota[2], SF. Chichibu[3], G. Cantwell [4], D. C. Reynolds[5], and C. W. Litton[5]

[1]*Department of Applied Physics, Hokkaido University, and CREST, Japan Science and Technology Agency, N13 W8, Kitaku, Sapporo 060-8628, Japan,* [2]*Department of Electrical, Electronics, and Computer Engineering, Waseda University, Shinjuku, Tokyo 169-8555, Japan,* [3]*Institute of Applied Physics, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-8573, Japan,* [4]*Eagle-Picher Technologies, LLC., 200B. J. Tunnel Blvd., Miami, Oklahoma 74354, USA,* [5]*Materials and Manufacturing Directorate, Air Force Research Laboratory, Wright-Patterson Air Force Base, Ohio 45433, USA*

adachi-s@eng.hokudai.ac.jp, phone/fax: +81-11-706-6669

Nonlinear optical processes are extremely sensitive to the interactions between elemental excitations. They provide direct information on processes that are inaccessible to other spectroscopic techniques. Therefore, the investigations of many body effects including their higher-order effects through nonlinear spectroscopy such as four-wave mixing (FWM) are going to be important direction of research in condensed matter physics. II-VI semiconductors have advantages to investigate exciton-exciton interactions due to their large exciton binding energies, however, only little is known about coherent properties of biexcitons [1] and their scattering processes because of the faster dephasing, in part, due to sample quality compared to GaAs. In this work, we studied that the dephasing dynamics of excitons and biexcitons in a free-standing *c*-face ZnO (2 x 3 x 0.5 mm) by using four-wave mixing technique.

Figure shows time-integrated FWM traces as a function of delay-time τ_{12} for $(\uparrow\rightarrow)$ (right panel) and $(\sigma_+\sigma_+)$ (left panel), respectively, varying the central excitation energy ($\boldsymbol{E}\perp\boldsymbol{c}//\boldsymbol{k}$). The exciton density is estimated as $\sim 10^{16}$ cm^2. As seen the figure, the FWM signals of ZnO are very sensitive to the excitation energy by reflecting many related energy states. The uppermost trace, whose excitation energy is far below the lowest A-hole exciton, has the strong emission almost only for $\tau_{12} < 0$, which should be related to TPCs of bound biexcitons (XX_{AA} and XX_{AB}). Because the formation of XX_{AA} is forbidden for $(\sigma_+\sigma_+)$, the beating appears only for $(\uparrow\rightarrow)$. In particular, A exciton-B exciton correlation i.e. bound and unbound heterobiexcitons, which can be generated even in cocircular polarizations, has a significant qualitative impact on the optical response [2]. This long rise also allows us to extract the dephasing rate of biexciton. The beatings due to the interference between XX_{AA}-XX_{AB} TPCs dominate the first four signals from the uppermost trace. As increasing more the excitation energy, the stronger emission around $\tau_{12} \sim 0$ grows, which is induced by the excitation of continua and their destructive interference. From the FWM measurements varying light polarizations, directions ($\boldsymbol{k}//\boldsymbol{c}$ or $\boldsymbol{k}\perp\boldsymbol{c}$) and temperatures, we can get the valuable knowledge about valence-band ordering, biexciton binding energies, dephasing of biexcitons and excitons, and polariton nature of ZnO [3].



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