

Luminescence Decay Kinetics of Electron-Hole Plasma in II-VI Epitaxial Layers and Micropowders

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The luminescence decay kinetics of epitaxial layers of ZnSe and micropowders of ZnSe and CdS were measured at room temperature and at high levels of optical excitation. Comparison of the experimental results with the theoretically calculated decay kinetics showed good agreement for the initial moment of time at high concentrations of nonequilibrium charge carriers.

I. INTRODUCTION

Wide-gap semiconductor compounds II-VI and their solid solutions in the form of heterostructures, epitaxial layers, and micropowders are used to create injection lasers, electronically and optically pumped lasers, luminescent screens, photodetectors, random monochrome and “white” lasers [1-3]. The lifetimes of nonequilibrium charge carriers (NCC) in the electron-hole plasma, is important from scientific and practical points of view, since the lifetimes of NCC and their concentrations are closely related to the recombination and lasing mechanisms.

In this work, the effect of the power density of the exciting luminescence on the room temperature spectra and decay curves of the photoluminescence (PL) of ZnSe epitaxial layers and micropowders of ZnSe and CdS crystals at high excitation levels was studied. The experimental results are compared with theoretical calculations of the PL decay kinetics.

II. MATERIALS AND METHODS

In work were used ZnSe epitaxial layers from 500 nm to 2 μm thick, grown on gallium arsenide wafers by molecular beam epitaxy, as well as micropowders of ZnSe and CdS crystals with diameter of about 0.5 - 2 μm , were studied. The luminescence decay kinetics were carried out using a femtosecond Yb: KYW laser with a emission wavelength of 260 nm, with energy density up to 500 $\mu\text{J}/\text{cm}^2$, and pulse duration of 650 fs, a pulse repetition rate of 60 kHz and a Hamamatsu C4334 streak camera with a temporal and spectral resolution of 20 ps and 1 nm, respectively. Luminescence from the surface of ZnSe epitaxial layers and from ZnSe and CdS micropowders with a diameter of 5 - 10 μm was recorded at 300 K using a wide-range SDH-IV spectrometer.

III. EXPERIMENTAL RESULTS

The PL spectra of epitaxial layers and powders are located near their fundamental absorption edge. This high level of excitation provides at room temperature to generate NCC concentration up to 10^{17} - 10^{19} cm^{-3} , and luminescence is caused by the recombination of NCC in a dense electron-hole plasma. Fig. 1. shows the PL spectra of ZnSe epilayers from

the wafer surface (1, 2), ZnSe (1', 2') and CdS (1'', 2'') micropowders at low (1'', 1', 1,) and high (2'', 2', 2) excitation levels 90 and 390 $\mu\text{J}/\text{cm}^2$. An increase in the excitation level has a weak effect on the PL spectrum of epilayers and leads to a strong broadening and transformation of the PL spectra of both types of micropowders. The broadening of the spectra and the appearance of their structure in ZnSe and CdS micropowders are due to the attainment of a high NCC concentration owing to the limitation of optical amplification and diffusion of carriers by the small size of micropowders. Possible reasons for the differences in the PL spectra of epitaxial layers and powders will be discussed.

Fig. 2. shows, that the luminescence intensities as a function of time (luminescence decay) at two excitation levels of CdS and ZnSe micropowders, as well as ZnSe epitaxial layers. All kinetics have a non-exponential decay. The values of the decay parameter τ depend on the excitation level and they change with the decay time. At the end of the PL pulse the kinetics are close to exponential and the value of τ are about 1000 ps for ZnSe epilayers, 920 ps for ZnSe powder and 1070 ps for CdS powder and they are determined by the nonradiative recombination. At the initial stage of the emission, the minimal value of decay time for CdS and ZnSe powders and ZnSe epilayers are 42, 25 and 20 ps.

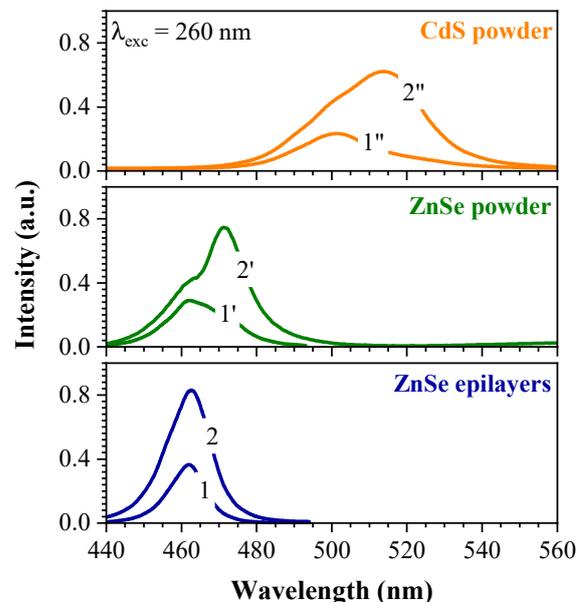


Fig. 1. PL spectra of ZnSe epitaxial layers (1, 2), ZnSe (1', 2') and CdS (1'', 2'') powders at $T = 300$ K.

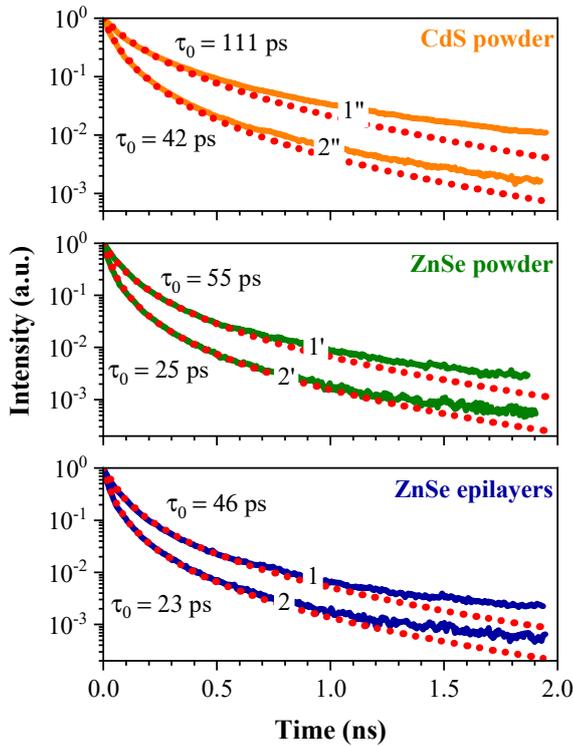


Fig. 2. Experimental (lines) and theoretical (points) PL kinetics of ZnSe epitaxial layers (1, 2), ZnSe (1', 2') and CdS (1'', 2'') powders. Excitation energy density was $90 \mu\text{J}/\text{cm}^2$ (1, 1', 1'') and $390 \mu\text{J}/\text{cm}^2$ (2, 2', 2'').

IV. THEORETICAL ANALYSIS

To describe the decay kinetics of photoluminescence at high excitation levels we will neglect the radiative recombination channels via impurity and assume that the equilibrium concentrations of both types of charge carriers (electrons and holes) are equal. In this case, to describe the rates of generation and recombination of charge carriers in the bands, we will use the following differential equation:

$$\frac{dN}{dt} = I_{exc}(t) - B(N^2 - N_0^2) - A(N - N_0). \quad (1)$$

The total concentration of electron-hole pairs N is equal to the sum of the equilibrium N_0 and nonequilibrium n concentrations. Here A and B are the nonradiative and radiative recombination coefficients. I_{exc} is the intensity of optical excitation of nonequilibrium charge carriers. We assume that the distribution of charge carriers over the depth is uniform. In (1) the photoluminescence intensity will be determined by the rate of radiative recombination described by the second term on the right-hand side. We will analyze the solution of (1) for the case when at the initial moment of time corresponding to the end of the excitation pulse $t_0 = 0$, the concentration of nonequilibrium charge carriers is maximum and is equal to n_0 . In this case, the decrease in the concentration of nonequilibrium charge carriers with time will be determined from the solution of the differential equation:

$$\frac{dn}{dt} = I_{exc}(t) - Bn^2 - An. \quad (2)$$

For high levels of excitation, which are used in our work, the concentration of nonequilibrium charge carriers is much higher than the concentration of equilibrium ones. It can be shown that in this case the concentration of nonequilibrium charge carriers after the termination of the excitation will decrease over time according to the law:

$$n(t) = \frac{An_0 e^{-At}}{Bn_0 + A - Bn_0 e^{-At}}. \quad (3)$$

The photoluminescence emission intensity is $I_{PL}(t) = B [n(t)]^2$.

As follows from the experimental kinetics (Fig. 2.) of PL, the decay of its intensity does not follow an exponential law. To characterize the state of the attenuation process at a given time, the decay parameter can be introduced [4]:

$$\tau_{PL}(t) = -\frac{I_{PL}(t)}{dI_{PL}(t)/dt}. \quad (4)$$

The equation indicates that, at least at the initial stage, the PL relaxation should not occur according to an exponential law and at each moment of time, it can be characterized by its own decay constant. The theoretical and experimental dependences of the luminescence decay kinetics are in good agreement for the initial moment of time, i.e., at high concentrations of NCC, when radiative recombination prevails. From the estimates of the electron-hole generation rates for this case, the initial concentrations n_0 were estimated, which, depending on the excitation level, are $(0.8 - 3) \cdot 10^{18} \text{cm}^{-3}$ for all samples, as well as the constants B : $(5 - 7) \cdot 10^{-9} \text{cm}^3 \text{s}^{-1}$ and A : $(4.5 - 5) \cdot 10^8 \text{s}^{-1}$. Some discrepancy between the experimental and theoretical damping curves was found at the final stage of the PL decay process. This may be due to the specifics of the excitation and registration of the luminescence pulse and will be discussed. Based on the experimental data, the PL efficiencies were also calculated for all three materials as a function of the emission time under intense excitations.

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