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High-density effects due to interaction of self-trapped exciton with (Ba, 5*p*) core hole in BaF_2 at low temperature

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The effect of VUV undulator excitation intensity on the emission shape and decay time of BaF_2 crystal at low temperature has been observed. The findings are explained in terms of quenching of Auger-free luminescence (cross luminescence) by self-trapped exciton via Förster mechanism energy transfer.

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We have been recently observed the effect of quenching of Auger-free luminescence (AFL) in BaF_2 due to interaction (Ba^{3+}, *5p*) uppermost core hole with electron excitation created in the same VUV absorption process [1]. The nature of the secondary excitations was not discussed and in principle it may be the trapped electron near the bottom of the conduction band or self-trapped exciton (STE) or defects (e.g., F-H pairs [2]), etc.

In order to study the effect of interaction for core hole with STE at least one of the species must be created in crystals at high concentration (more than $\sim 10^{18}$ cm^{-3}). In the former case the photon energy more than 18 eV and in the latter one more than 10 eV has to be used [3]. Unfortunately, there are a lot of known problems with using VUV laser in this photon range and we have used another opportunity. The undulator at the UVSOR storage ring (Okazaki, Japan) provides an intense quasi-monochromatic radiation around 30 eV with bandwidth $\sim 5\%$ and the incident photon flux on the sample is about 10^{14} photon/(s·mm^2), while the electron current is 100 mA and the excited spot on the sample is 1 mm^2 [4]. It is well-known that the absorption coefficient of BaF_2 in VUV region [5] is very high and the lifetime of STE has the millisecond components at low temperature [6]. It is simple to calculate that the steady-state concentration of STE N is

$$N[\text{exciton/cm}^3] \cong$$
$$\cong I[\text{mA}]\, N_0[\text{photon/s}\cdot\text{cm}^2\cdot\text{mA}]\, \delta\, K[\text{cm}^{-1}]\, \tau[\text{s}], \qquad (1)$$

where I is the beam current in the storage ring; N_0 is the photon flux on the sample at beam current $I = 1$ mA; δ is the creation efficiency of STE in the sample; K is absorption coefficient of VUV excitation photon; τ is the longest lifetime for STE. Relation (1) is valid when the time decay of STE is much longer than interval between excitation pulses, e.g. 11 ns for multi- and 177.6 ns for single bunch mode operation UVSOR.

Using formula (1), in our experimental conditions we could choose $\delta \sim 0.1$, $K \sim 10^6$, $\tau \sim 10^{-2}$ at $T = 10$ K. As a result we have the following simple expression for steady-state concentration of STEs under undulator irradiation:

$$N[\text{exciton/cm}^3] \cong \text{I}[\text{mA}]\cdot 10^{17}\,. \qquad (2)$$

Thus, under undulator excitation at low temperature the mean distance R between STEs is well enough for dipole–dipole interaction. For instance, at $I = 100$ mA the $R \sim 5$ nm. It should be mentioned that at room temperature the lifetime of STE is about 10^{-6} s and accumulation effect is negligible for our purpose. The Al-filter with about 150 nm thickness was used in order to remove the visible scatter and straight light from beamline. The LHT flow type cryostat provided a temperature of the sample across 10–300 K. The emission spectra were analyzed by a grating visible monochromator and detected by micro-channel-plate photomultiplier (HAMAMATSU R2287U-06). For time decay measurements the time-correlated single photon method was used [7].

In Fig. 1 the two emission spectra for BaF_2 normalized at the maximum of STE are presented. It is clear seen that the relative intensity of AFL (~ 5.6 eV) decreases with the beam current in storage ring. The measurements were done under $I_1 = 35$ mA (big crosses) and $I_2 = 168$ mA (small crosses) at 10 K. All emission curves which were measured between I_1 and I_2 fall in the middle. Figure 2 shows the decay time profiles for BaF_2 at 10 K (lower curve) and at room temperature (upper curve) at the same beam current in storage ring. It is clear seen the effect of cooling on the decay rate of AFL. Both decay curves exhibit a complicated non-exponential decay law. The mean decay time was 0.9 ns at room temperature and 0.4 ns at $T = 10$ K. As both the time duration (full width at half maximum is equal 500 ps) and interval between bunches much faster than the lifetime of the STE at $T = 10$ K we can consider uniform distribution of the STEs near the surface layer of BaF_2. In this case the time decay law has to be governed by the Förster mechanism [8,9] of quenching for AFL:

$$J(t) \sim \int L(t') \exp\left\{-\left[\frac{t-t'}{\tau_{rad}} + \gamma N(t-t')^{1/2}\right]\right\} dt' , \tag{3}$$

where γ is determined by the overlap integral of the luminescence spectrum of AFL and the absorption spectrum of STE; N is the steady-state concentration

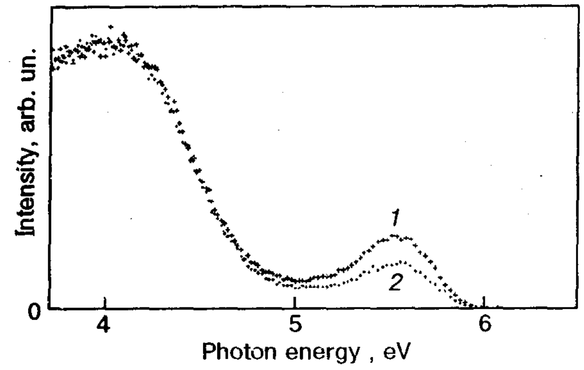

Fig. 1. The emission spectra of BaF_2 at $T = 10$ K under 30 eV undulator excitation normalized at maximum of STE band (4 eV): at beam current in UVSOR storage ring $I = 35$ (*1*) and 168 mA (*2*).

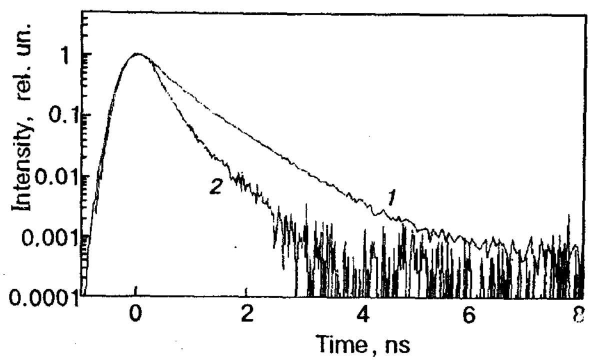

Fig. 2. The decay curves of BaF_2 at $T = 300$ (*1*) and 10 K (*2*) under 30 eV undulator excitation.

of STEs defined by relation (2) and $L(t')$ is the excitation profile [1]. Thus, from (2) and (3) we can conclude that the strength of quenching of AFL due to interaction with STE is proportional to the beam current in the storage ring and increases under cooling. Within the framework of this model the experimental results could be explained as follows. The relation between emission intensity for STE and AFL depends on their concentrations. In case of AFL the radiative time decay is about 0.9 ns and the mean concentration of core hole created by one bunch is less than ~ 10^9 cm^{-3}. Thus, the amount of STEs which annihilate due to interaction with core holes is much less than the steady-state concentration of the STEs. Thus, the decrease in the relative intensity of AFL in emission spectrum BaF_2 with beam current in Fig. 1 could be successfully explained by the interaction of the STE with uppermost core holes. The same explanation might be used in case of decay acceleration under cooling. When the temperature decreases (Fig. 2, curves *1, 2*) the steady-state concentration of the STEs increases and the quenching of the AFL due to interaction of the core hole with STE becomes more remarkable. The latter findings can't be explained by the near surface loses because the time decay of AFL under moderate excitation intensity ~ 10^{11} photon/s has not showed any changes [10]. The data obtained are very important for understanding scintillation process in BaF_2, because energy 32 eV of the closely spaced (Ba^{3+}, 5*p*) core hole and STE should be created in the same absorption process [1,11,12] and thus decrease the yield of fast nanosecond luminescence.

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