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Origin of carrier heating in semiconductor nanocrystals: excess energy of photoexcited electrons or Auger processes?

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Abstract. Heating of carriers photoexcited with different excess energies into semiconductor nanocrystals of different radii is studied. The effective carrier temperature is evaluated by analysing the shape of the luminescence band with nanocrystal size dispersion taken into account. The heating is shown to be determined rather by the initial excess energy of the photoexcited electron–hole pairs than by Auger processes even in nanocrystals with considerable quantum confinement.

1. Introduction

The recent interest in energy relaxation of carriers excited in semiconductor nanocrystals is partially caused by the importance of the phenomenon to the operation of quantum dot lasers [1] as well as by intriguing peculiarities of this quasi-zero-dimensional object sharing properties of both bulk crystals and free atoms or molecules. In small nanocrystals, a drastic decrease in efficiency of energy relaxation by emission of LO phonons [2] as well as blockade to emission of acoustic phonons [3] was predicted; however, new mechanisms of energy relaxation [4–6] have been pointed out as possible substitutes for direct phonon emission to circumvent the predicted bottleneck. In nanocrystals with weak quantum confinement, a considerable increase of the effective carrier temperature over the lattice temperature was demonstrated by analysing the luminescence band shape [7]. In doing so, one obtains an important parameter characterizing the energy relaxation in the system of photoexcited carriers.

The present paper aims to demonstrate that the investigation of carrier temperature may also be utilized to reveal the *origin* of carrier heating in semiconductor nanocrystals. Nanocrystals large enough for an ensemble of carriers to be excited were investigated. The idea of the method is rather straightforward. In principle, the heating of photoexcited carriers without imposing an external electric field can be either extrinsic, i.e. caused by *initial* excess energy of the photoexcited electron–hole pairs, or intrinsic, i.e. originating from exothermal processes inside the system of *thermalized* carriers (the Auger processes resulting in transition of one of the three interacting carriers to a highly energetic state after recombination of the other two carriers, the exciton–exciton interaction with radiative recombination of one of the interacting excitons while the other exciton gains energy to satisfy energy and momentum conservation). The intrinsic heating depends on carrier density, while the extrinsic heating is mainly governed by the amount of excess energy of the photogenerated carriers. So, the strong dependence of the heating caused by the

Auger processes on the carrier density and insensitiveness of the Auger heating to the excess energy may be utilized for identification of the role of these processes in the dynamics of a dense electron–hole system generated in a nanocrystal.

For a long time, Auger processes have been claimed to be of great importance in recombination processes in nanocrystals [8] though there were no direct experiments demonstrating their role up to now. In bulk crystals, Auger processes are often the dominating recombination process in highly excited narrow-band-gap semiconductors, but are ineffective in crystals with band gap exceeding approximately 0.6 eV [9], since energy and momentum conservation causes an exponential decrease of the probability of the Auger processes with increasing band gap. Taking into account the real structure of conduction and valence bands, involvement of phonons etc slightly modifies the rate of the Auger processes, but the calculated lifetime of the electron–hole plasma (EHP) due to the Auger recombination in such semiconductors as CdSe or CdS is several orders of magnitude lower than the observed lifetime at accessible carrier densities. It is worth noting, however, that violation of momentum conservation near heteroboundaries of nanocrystals results in a high rate of *nonthreshold* Auger recombination [10]. Recently, the Auger recombination has been pointed out as the mechanism populating the trap states at the surface of a nanocrystal [11]. The possibility of highly efficient Auger carrier capture is demonstrated in self-assembled semiconductor dots [12]. Also it has been shown that the Auger processes initiate heating of carriers in GaAs/AlAs heterostructures [13]. So the Auger processes may be of importance in nanostructures even of semiconductors with comparatively large band gap.

To use the effective temperature as a probe of the carrier dynamics in nanocrystals, a thorough study of luminescence and analysis of its band shape should be carried out. So the first part of the present paper is devoted to an investigation of the reliability of determining the effective carrier temperature in nanocrystals. Afterwards, the luminescence of carriers excited in nanocrystals without excess energy is compared with luminescence under photogeneration of carriers with large excess energy.

2. Experimental details

Luminescence was studied under quasi-steady conditions. The samples were excited by harmonics of the radiation of a Q-switched YAG:Nd³⁺ laser (pulse duration was 10 ns) operating at a repetition rate of 12.5 Hz. The luminescence spectra were analysed by using a Jobin Yvon double monochromator HRD-1 and recorded by using a photomultiplier and a boxcar integrator with subsequent digital data acquisition. The measured spectra were corrected for spectral response of the monochromator and photomultiplier.

To study the band shape of the luminescence of highly excited nanocrystals, two samples containing CdS nanocrystals in a glass matrix were investigated. The samples were fabricated by using the diffusion controlled growth technique [14] in identical conditions except the annealing time resulting in different average nanocrystal size in the two samples. The average size of nanocrystals was also estimated by analysing the low-energy absorption spectra of the samples. The average nanocrystal radius \bar{a} of the first sample was 100 nm. The radius \bar{a} is still considerably smaller than the light wavelength, so that the Maxwell–Garnett theory should be applied when studying the absorption of light; however, the glass matrix does not significantly affect the net absorption of this composite system [15]. All the processes of carrier relaxation and recombination in each nanocrystal contained in this sample proceed essentially as in a bulk crystal since the quantum confinement is negligible. So we chose this sample as an object with bulk-like nanocrystals.

The other sample contained nanocrystals with average radius of 7.5 nm. Here, the lowest band-to-band transition is shifted by 28 meV due to the quantum confinement of the free carriers. The nanocrystals in this second sample are, however, still large enough to yield excitation of an ensemble of electron–hole pairs. One electron–hole pair per nanocrystal with $\bar{a} = 7.5$ nm corresponds to the bulk density of $6 \times 10^{17} \text{ cm}^{-3}$. So a system of up to several tens of electron–hole pairs can be created in such a nanocrystal under quasi-steady photoexcitation without inducing optical damage of the sample.

Naturally, the so-called photodarkening of the samples should be considered when exciting the sample with light as intense as in our experiments. The effect results mainly in increasing the absorption of the sample as well as in decreasing the lifetime of the photoexcited electron–hole pairs. These permanent changes are apparently caused by photomodification of the surface of the nanocrystals [16]. To avoid interference of these permanent modifications with the transient effects under investigation, our investigation was carried out only after the samples were exposed to the intense laser irradiation long enough for saturation of all the photomodification processes to be completed. Stability of the luminescence quantum yield of the samples was utilized as an indication of the saturation. Furthermore, the photodarkening effect is considerably diminished by lowering the temperature of the sample. So all the samples under investigation were exposed to the photoexcitation only after being cooled down to the temperature of liquid nitrogen.

A sample of a commercial Corning glass filter (2-60) containing $\text{CdS}_x\text{Se}_{1-x}$ nanocrystals was chosen as an object for our experiments on the influence of the excess energy on the carrier heating. The sulphur content x was estimated to be 0.47 from the spectral separation of CdS-like and CdSe-like LO modes in the Raman spectrum [17] by using the results presented in [18]. The band gap of the $\text{CdS}_{0.47}\text{Se}_{0.53}$ at liquid nitrogen temperature, calculated by using a linear extrapolation between CdS and CdSe, is 2.18 eV. The average nanocrystal radius in the sample is approximately 5 nm [19]. Consequently, the effective band gap in the nanocrystals under investigation is blue-shifted on average by 80 meV. So the quantum energy of the second harmonic (2.330 eV) of the YAG:Nd³⁺ laser radiation used for the excitation is rather close to the band gap, while the third harmonic (3.495 eV) excites the electrons with large excess energy.

3. The effective temperature of carriers

The luminescence spectra of the sample with 100 nm average radius nanocrystals are illustrated in figure 1(a). The spectra were measured at 77 K temperature. The excitation intensities used in our experiments were high enough to generate sufficient density of carriers for the Mott transition from a system of excitons to a system of free carriers to occur. The transition is studied in more detail elsewhere [20]. As usually observed in bulk crystals, the high-energy slope of the observed single dominating band is exponential, reflecting the exponential behaviour of the energy distribution function of carriers which governs the band shape in this spectral region. The decrease of the steepness of the slope with increasing excitation intensity is obviously caused by heating of the EHP generated in the nanocrystals. The high-energy side of the EHP band can be fairly well fitted by using the expression describing recombination between carriers in parabolic bands without conservation of the wavenumber. The latter assumption is rather plausible in nanocrystals with strong influence of surfaces due to a large surface-to-volume ratio. So the full lines in figure 1(a) illustrate the fitting of the experimental spectra with the expression:

$$I_{LUM}(h\nu) \propto (h\nu - \tilde{E}_g)^2 \exp\left(-\frac{h\nu - \tilde{E}_g}{k_B T}\right). \quad (1)$$

Here, $h\nu$ is the energy of the emitted photon, \tilde{E}_g is the normalized band gap, k_B is the Boltzmann constant and the Boltzmann distribution of carriers is assumed. The expression (1) is derived without taking into account the carrier–phonon interaction [21] and the damping in the EHP [22] which are responsible for the line shape in the region of the low-energy slope of the EHP luminescence band. Consequently, this region is not properly described by expression (1). Moreover, the low-energy side of the EHP band is overlapped with other luminescence bands. These low energy bands appear more clearly in the luminescence spectra at lower excitation intensities and probably are caused by recombination through impurities and by luminescence of excitons. However, the processes mentioned above have no significant influence on the high-energy slope of the band at high excitation intensities which is of interest in our study in order to evaluate the EHP effective temperature T_C . The dependence of the EHP temperature on the excitation power density, evaluated by fitting luminescence bands at different excitation intensities, is presented in figure 1(b). At moderate carrier densities, T_C should just reflect the quasi-temperature of nonequilibrium long-wavelength LO phonons produced predominantly via their successive emission by photogenerated electrons with a large excess energy [23]. The full line in figure 1(b) represents the T_C against pump intensity I_{EXC} dependence under the assumption that

$$[N(T_C) - N(T_L)] \propto I_{EXC} \quad (2)$$

where $N(T_C)$ and $N(T_L)$ are the Bose–Einstein distribution functions for LO phonons of CdS (phonon energy 37.8 meV) at the EHP temperature T_C and the lattice temperature (80 K), correspondingly. The dependence calculated without any adjustable parameters describes the experimental data fairly well.

The red-shift of the EHP luminescence band proceeding with increasing excitation intensity is caused by the band gap renormalization due to many-body effects in the EHP which was earlier observed in the CdS nanocrystals by using luminescence [20] and transient absorption techniques [24]. When evaluated by using the expression for band shrinkage against EHP density derived for bulk crystals [25], the observed maximal shift of 40 meV corresponds to the density of $3 \times 10^{18} \text{ cm}^{-3}$. The direct evaluation of the EHP density is obscured due to the inaccuracy of defining the carrier-density-dependent effective lifetime of the carriers. However, even a rough estimation giving the value of the order of 10^{19} cm^{-3} allows one to conclude that the density calculated from the band gap renormalization is lower than that estimated according to the excitation conditions. The discrepancy could be explained by carrier capture by deep traps via multiphonon emission enhanced by increased temperature of the EHP [26].

Luminescence spectra of the sample with 7.5 nm average radius nanocrystals are illustrated in figure 2(a). The spectra are dominated by a single band with the high-energy side exponentially decreasing with the energy of the emitted photon. The decrease of the slope with increasing excitation intensity seems to be caused by heating of carriers. However, the heating in nanocrystals with considerable quantum confinement should be treated more thoroughly. First of all, the energy spectrum in these nanocrystals is discrete. However, except for the really small nanocrystals where excitation of many electron–hole pairs is unrealistic, the effective joint density of states is distorted in comparison with the bulk crystals only near the band edge. Small separation between the states of holes due to their large effective mass and mixing of the valence subbands, thermal broadening, lifting the restrictions imposed for optical transitions by selection rules due to deviations from the spherical symmetry of the nanocrystals etc cause a rather smooth spectrum of the effective joint density of states in the region where the high-energy wing of the luminescence of the

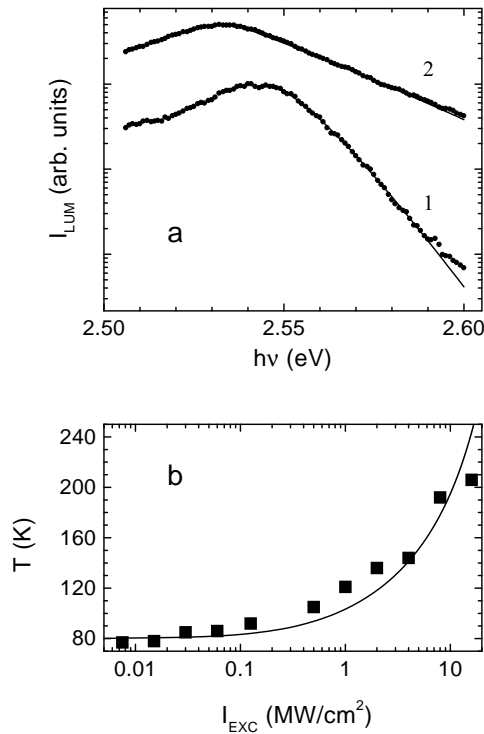


Figure 1. Luminescence spectra (dots) at two excitation power densities, 0.12 (1) and 16 MW cm^{-2} (2), and their fitting with the calculated band shape (a); and carrier temperature against excitation density dependence (b) determined from fitting the luminescence band (squares) and calculated according to equation (2) (full line). The sample contains CdS nanocrystals with average radius of 100 nm; lattice temperature equals 77 K.

highly excited nanocrystals is observed. The bulk-like character of the spectrum of the joint density of states far above the band gap is consistent with the extensive results on linear and non-linear absorption and is confirmed by the exponential slope of the luminescence band. Taking into account the various energy relaxation channels mentioned in the introduction, such a distribution is rather plausible.

However, a straightforward fitting of the luminescence band shape with expression (1) describing the exponential high-energy slope gives rather confusing results presented by squares in figure 2(b). The temperature evaluated this way asymptotically approaches the value 130 K, well above the lattice temperature (77 K).

So the fitting of the luminescence band with expression (1) gives plausible results when the temperature is above a certain value and completely fails to be appropriate on decreasing the temperature to be evaluated. The behaviour may easily be caused by the distribution of nanocrystal size. The observed luminescence is actually a superposition of quanta emitted from nanocrystals of different size. Since the energy spectrum in each individual nanocrystal is shifted due to quantum confinement according to its size, the corresponding luminescence spectra are different. To take the effect into account, we calculated the resulting luminescence band shape by integrating contributions of all the different nanocrystals. The band shape of the luminescence emitted from any individual nanocrystal was described by expression (1). However, the effective band gap \tilde{E}_g was

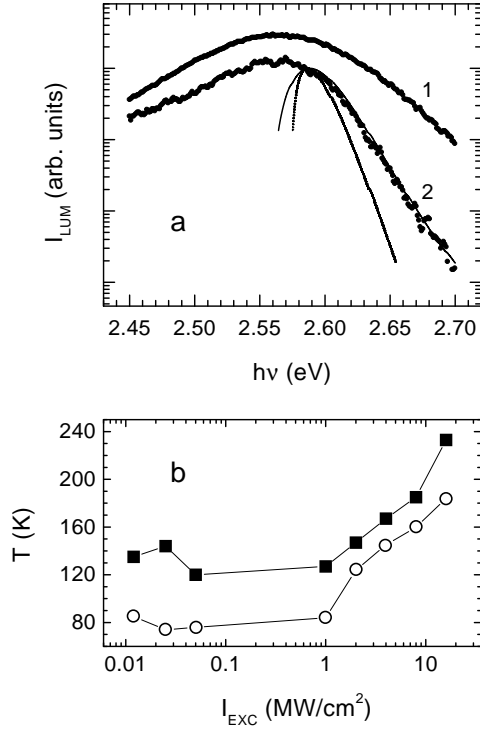


Figure 2. Luminescence spectra (dots) at two excitation power densities, 0.12 (1) and 16 MW cm⁻² (2), and their fitting with the calculated band shape with (full line) and without (dotted line) size dispersion taken into account (a); and carrier temperature against excitation density dependence (b) determined from fitting the luminescence band with (circles) and without (squares) the size dispersion taken into account. The sample contains CdS nanocrystals with average radius of 7.5 nm; lattice temperature equals 77 K.

calculated as a function of the nanocrystal radius a :

$$\tilde{E}_g(a) = \tilde{E}_g(\infty) + \frac{\hbar^2 \pi^2}{2\mu a^2} \quad (3)$$

with $\tilde{E}_g(\infty)$ as the bulk-crystal value of the band gap and μ as the reduced effective mass of electron and hole. The relative weight of the nanocrystals with a certain radius was taken to be determined by the size distribution function. Though the size distribution in samples fabricated by using the diffusion controlled growth technique should follow the Lifshitz–Slezov function [14], we approximated the distribution by the Gauss function. So the shape of the luminescence band was calculated according to expression

$$I_{LUM}(h\nu) \propto \int_0^\infty \Theta(h\nu - \tilde{E}_g(a)) [h\nu - \tilde{E}_g(a)]^2 \exp\left[-\frac{h\nu - \tilde{E}_g(a)}{k_B T}\right] \exp\left[-\frac{(a - \bar{a})^2}{2\sigma^2}\right] da. \quad (4)$$

Here, the effective band gap \tilde{E}_g is defined by expression (3), σ is the dispersion parameter and $\Theta(x)$ denotes the Heaviside function.

As expected, the influence of the size distribution is negligible for the sample with average nanocrystal radius of 100 nm. However, the band shape in the sample with

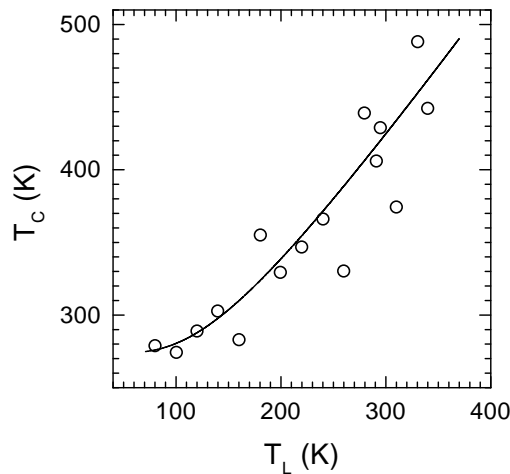


Figure 3. Dependence of effective carrier temperature on lattice temperature at constant excitation intensity (circles—experiment, full line—calculation according to equation (2)).

$\bar{a} = 7.5$ nm is strongly affected by the size distribution. It is worth noting that the high-energy tail of the calculated band is still very close to exponential. However, the slope is determined not only by the effective temperature but also by the size distribution which is described by the dispersion parameter in the Gauss function. The fitting result is illustrated in figure 2(a) by a full line. The dispersion parameter $\sigma = 1.1$ nm, i.e. 15% of the average radius, was found to ensure the best fit and was kept constant in the fitting of all the spectra measured in this sample. Now the temperature at the lowest excitation intensities, determined from the fitting with the size distribution taken into account, approaches 77 K (see figure 2(b)). The fitting curve for the luminescence band shape calculated with $T_C = 77$ K by simply using the expression (1) is presented for comparison in figure 2(a) by a dashed line.

So, the exponential slope of the luminescence is influenced by two effects: a constant contribution due to the size distribution of the nanocrystals and heating of the carriers in the nanocrystals. To test this conclusion we measured the luminescence spectra of the sample with 7.5 nm average nanocrystals at different lattice temperatures (see figure 3). The excitation intensity was sufficient to cause a substantial heating of carriers (280 K at 77 K of lattice temperature) and was maintained constant while measuring all the spectra. According to expression (2), the T_C versus T_L dependence at $I_{EXC} = \text{constant}$ should follow the full line in figure 3. In spite of the rather large scatter of experimentally determined carrier temperatures the fitting can be considered as being fairly satisfactorily.

Concluding the study of the band shape we can assume that the carrier heating takes place even in nanocrystals with considerable quantum confinement. The effective temperature of the carriers can be estimated by analysing the luminescence band shape at high excitation intensities while the high-energy slope of the band at the lowest excitation intensities is determined rather by size distribution than by the temperature.

4. The origin of the carrier heating

In order to examine whether the heating of carriers is determined by their density or by the excess energy of the photoexcited electron-hole pairs delivered to the system of the relaxed

carriers directly or through the system of LO phonons, we excited the nanocrystals with light quanta of different energy. The second harmonic of the YAG:Nd³⁺ laser radiation (quantum energy $h\nu_2 = 2.330$ eV) is rather close to the band gap of the CdS_{0.47}Se_{0.53} nanocrystals with radii of 5 nm while the quanta of the third harmonic ($h\nu_3 = 3.495$ eV) of the same radiation excite the carriers well above the band gap.

Evolution of the luminescence spectra with increasing excitation density of the third-harmonic radiation is illustrated in the inset of figure 4. The band broadens and shifts to the high-energy side, apparently reflecting the dominance of radiative recombination involving donor or acceptor states at the lowest excitation levels, increasing contribution of excitons and, finally, band-to-band recombination of free electron–hole pairs. The high-energy tail of the band at elevated excitation levels is exponential and its slope decreases with increasing excitation intensity (see figure 4), as observed in CdS nanocrystals. So it is plausible to interpret the decreasing slope by heating of carriers in the nanocrystals.

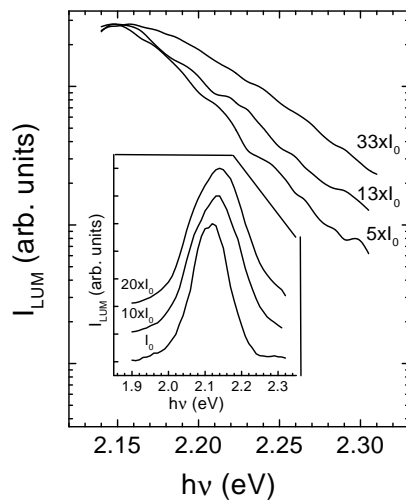


Figure 4. Modification of high-energy slope of the luminescence band with increasing excitation power density (indicated above the corresponding spectra, $I_0 = 0.1$ MW cm⁻²). The whole band is presented in the inset. The sample contains CdS_{0.47}Se_{0.53} nanocrystals with average radius of 5 nm. The spectra are arbitrarily shifted vertically for clarity.

Furthermore, we compared the luminescence spectra with the spectra of luminescence excited by generating carriers without excess energy. Since the absorption coefficient for the 3.495 eV quanta exceeds that for the 2.330 eV quanta by 20 times, the different absorption depth was taken into account when evaluating the density of photogenerated carriers. Though the general features of the transformation of the band shape are identical for both kinds of excitation, we do not observe any change in the high-energy slope of the band when excited with quanta without excess energy. In this case, the heating seems not to occur in the whole range of intensities of excitation generating carriers at densities corresponding to the densities of carriers excited with excess energy resulting in their considerable heating. So the presented comparison indicates that it is the excess energy and not the carrier density that mainly governs the heating of carriers in the nanocrystals under investigation.

To confirm the conclusion, we carried out an experiment with a two-beam excitation of the same sample containing nanocrystals of CdS_{0.47}Se_{0.53}. Beams of the second (2.330 eV) and the third (3.495 eV) harmonics of the laser radiation were focused onto the same 250 μm

diameter spot at the surface of the sample. The sample was excited separately by each of the beams as well as simultaneously by both of them. The resulting luminescence spectra are presented in figure 5 (the spectra are arbitrarily shifted vertically for easier comparison of their shape). The intensity of the third harmonic was chosen to cause a considerable heating above the lattice temperature (77 K), so that the high-energy slope of the spectrum corresponds to the carrier temperature of 250 K (curve 2 in figure 5).

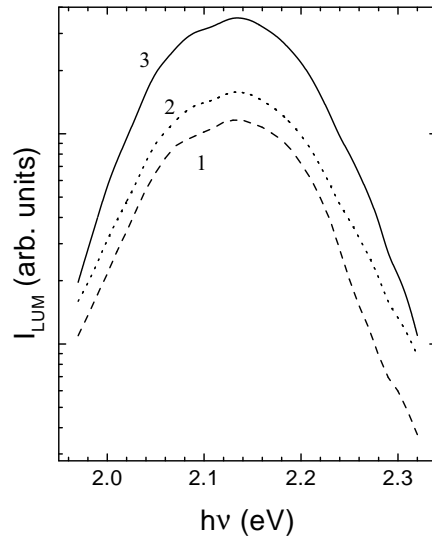


Figure 5. Spectra of luminescence of 5 nm average radius $\text{CdS}_{0.47}\text{Se}_{0.53}$ nanocrystals at equivalent intensities of photoexcitation with quantum energy 2.330 eV (1), 3.495 eV (2) and synchronously with both of the beams (3). The spectra are arbitrarily shifted vertically for clarity.

The intensity of the second harmonic was adjusted to excite the same luminescence intensity as in the first case. In spite of the different distribution of carriers into the depth of the sample caused by different absorption coefficients for the two energies of the exciting quanta, the equal luminescence intensity seems to satisfactorily reflect the equal carrier density. The conclusion is supported by the identical band shape in the vicinity of the band peak that is rather sensitive to carrier density. The corresponding spectrum excited with the second harmonic is presented by curve 1 in figure 5. The high-energy slope of the band corresponds to 80 K, i.e. equals the lattice temperature.

The carrier density generated by simultaneous excitation by both of the beams is twice as high as in the case of excitation by any single beam, but only half of the carriers enter the system with excess energy while the other half are excited nearly resonantly. Now the carrier temperature determined from the high-energy slope of the luminescence band is equal to 130 K. Consequently, the generation of additional carriers without excess energy caused a decrease of T_C instead of an increase which one could expect if the heating were caused by the Auger processes with the rate in strong dependence on carrier density. The cooling can easily be explained by sharing of the excess energy between all the carriers in the system. Naturally, the excess energy of the photogenerated electron-hole pairs affects the temperature of the relaxed carriers via strong interaction of the carriers with the system of LO phonons and that is the energy balance between these two systems that determines their effective temperatures. However, at the present stage, we do not necessarily need

detailed evaluation of the effective temperatures to conclude the origin of the heating in nanocrystals under investigation.

5. Conclusions

The obtained results imply that carrier heating may occur at elevated photoexcitation intensities even in nanocrystals with considerable quantum confinement. The effective carrier temperature can be evaluated by analysing the band shape of luminescence from these nanocrystals, provided that the nanocrystal size distribution is taken into account. The heating is caused by excess energy of the photogenerated electron–hole pairs and the Auger processes play no important role in nanocrystals large enough for an ensemble of electron–hole pairs to be excited. The latter conclusion is important for interpreting various results on optical nonlinearities when the dynamics of carrier density is described by a rate equation. It seems that, when the experimental results are not properly described by the rate equation including only the first- and second-order recombination terms, we have to look for more sophisticated origins of the discrepancies (enhanced surface recombination [27], thermally activated nonradiative capture of carriers via multiphonon emission [7] etc) instead of just including a term of the third order which describes the Auger processes.

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