Density and temperature oscillations of the gaseous phase of carriers above electron-hole droplets in AgBr

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Experiments have revealed the first periodic (in time) changes in the density and temperature of the gas phase of carriers around electron-hole droplets. A qualitative interpretation is offered for the effect. The oscillation period is calculated.

In this letter we describe some temporal oscillations which we have observed in the density and temperature of the gas phase of carriers around an electron-hole droplet during steady-state excitation of AgBr crystals. The gas phase in this multivalley polar semiconductor is a mixture of excitons and biexcitons. The electron-hole droplets have a significant condensation energy, $\varphi \simeq 55$ meV, a relatively brief lifetime[1,2] $\tau_0 \sim 10^{-8}$ s, and a pronounced self-heating effect.[3] The emission spectra of AgBr are of such a nature[3] that it is more convenient to monitor the oscillations in the density of the biexciton component of the gas phase and to detect the oscillations in the temperature of this phase on the basis of its exciton component. The experiments were carried out with the help of a computer-controlled spectral apparatus, described in detail in Ref. 3. Volume excitation of the AgBr samples was carried out with a cw He-Cd laser. The power incident on the crystal was increased smoothly over a time $\sim 10^2$ s to a value an order of magnitude above the condensation threshold and then held constant within 3% for $\sim 10^4$ s by a negative-feedback system.[3] Over this entire time the integral intensity of the biexciton emission band (the EM band[3]), which is proportional to the density of biexcitons, was measured at intervals $\sim 10^2$ s. At the same intervals, the spectrum of the emission band resulting from intervalley scattering of excitons (the ES band[3,4]) was recorded. The spectral characteristics of this band (its width at the base and the position of the maximum) can tell us about the temperature of the exciton gas, $\theta = kT$.

Figures 1 and 2 show experimental results, which provide evidence of the existence of temporal oscillations with a typical period $\sim 1.5 \times 10^3$ s, observed for the density of biexcitons and for the temperature of the exciton gas above electron-hole droplets at a constant (over time) rate of generation of electron-hole pairs in the interior of the AgBr: $g \sim 10^{19}$ $\text{cm}^{-3}\cdot\text{s}^{-1}$. The oscillations in the density and the temperature are in phase. The maximum amplitude of the temperature oscillations is estimated from the data in Fig. 2 to be 2–3 K. The period of the observed oscillations generally depends on the value of g; specifically, it increases with increasing g.

The analysis of the kinetics of this observed effect involves analyzing the time evolution of the radius (R) of the droplets that are produced, the average density ($\bar{n}$) of the gas phase, and the temperature (Θ^*) of the droplets as a result of condensation

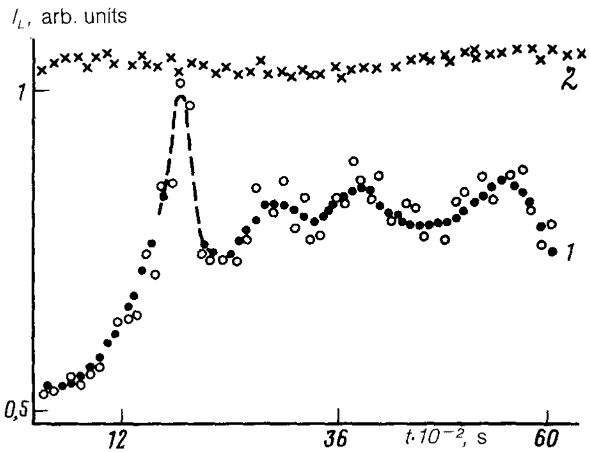

FIG. 1. 1—Intensity of the emission by excitons versus the time during steady-state excitation of AgBr crystals. ○) Experimental points; ●) result of a least-squares smoothing of the experimental points. 2—"Control" dependence, which demonstrates the response of the measurement system to a luminescence signal which remains constant over time. This signal was the emission from an exciton bound at an isoelectronic trapping center during steady-state (within 3%) excitation of AgBr at an intensity below the condensation threshold. The temperature of the constant-temperature chamber was 10 ± 0.1 K. The signal-to-noise ratio was no worse than 32 dB. The point at the maximum on curve 1 corresponds to the detection of 8×10^3 single-electron pulses from the photomultiplier.

and evaporation. For simplicity, we assume that the gas phase consists of particles of only a single species, e.g., excitons, and we assume a homogeneous case. We can then write

$$\frac{dR}{dt} = \frac{\gamma v_T}{n_0}(\bar{n} - n_T) - \frac{R}{3\tau_0}, \qquad (1)$$

$$\frac{d\bar{n}}{dt} = g - \frac{\bar{n}}{\tau} - 4\pi R^2 N \gamma v_T (\bar{n} - n_T), \qquad (2)$$

$$\frac{dc\Theta^*}{dt} = 4\pi R^2 \varphi \gamma v_T (\bar{n} - n_T) + h(\Theta^*)(\Theta^* - \Theta_l), \qquad (3)$$

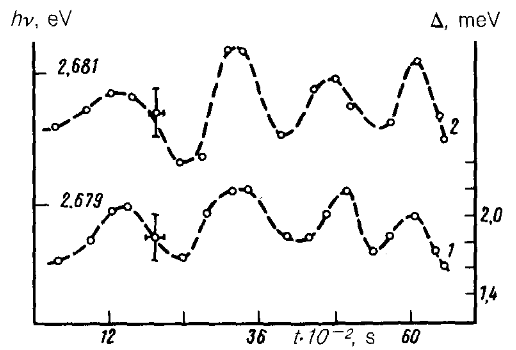

FIG. 2. 2—Time dependence of the energy position ($h\nu$) of the maximum: 1—width (Δ) of the maximum for the ES band during steady-state excitation of AgBr. The temperature of the constant-temperature chamber is 10 ± 0.1 K.

where v_T, $\gamma \sim 1$, and n_T (Ref. 5) are respectively the thermal velocity, the coefficient of adhesion to a droplet, and the thermodynamic-equilibrium concentration of the excitons above an electron-hole droplet of radius R; n_0 and N are the equilibrium density of the droplets and their concentration, respectively; τ is the exciton lifetime; c is the specific heat of the droplets; $h(\Theta^*)$ is the coefficient of heat transfer to the lattice; and Θ_l is the lattice temperature.

The initial supersaturation of the gas leads to a condensation and to the appearance of a certain number of droplets. A steady-state solution of Eq. (1) gives us two values of the droplet radius between the growth rate of the droplets is positive:

$$R_{1,2} = \frac{3\delta \dot{n} \tau_0 \gamma v_T}{2n_0} \left(1 \pm \sqrt{- \frac{8\sigma n_T(\Theta^*)}{3(\delta n)^2 \Theta^* \tau_0 \gamma v_T}} \right), \tag{4}$$

where σ is the surface tension of the droplet, and $\delta n = \bar{n} - n_T(\Theta^*)$.

Beginning at the time at which the bulk of the droplets with $R < R_1$ has decayed (this time corresponds to the first, very sharp peak on curve 1 in Fig. 1), the concentration of the remaining droplets can be regarded as constant, since the time scale for changes in this concentration during steady-state excitation is astronomically large in comparison with the times with which we are dealing.[6] Consequently, if the conditions $\bar{n},\Theta,\Theta^* = \text{const}$ are held during changes in the size of the droplets, the droplets could grow from R_1 to some other steady-state size R_2. However, since the concentration and temperature of the exciton gas decrease as a result of condensation, while the droplet temperature increases, the droplets will grow only to a size R_0, determined from the conditions

$$\frac{d}{dR}\left(\frac{\gamma v_T}{n_0}\{\bar{n} - n_T\}\right)\Bigg|_{R = R_0,\ \Theta^* = \Theta_0^*} = \frac{1}{3\tau_0}, \tag{5}$$

$$g - 4\pi R_0^3 N \frac{n_0}{\tau_0} = \frac{n}{\tau}, \tag{6}$$

$$4\pi R_0^3 \varphi \frac{n_0}{\tau_0} = h(\Theta_0^*)(\Theta_0^* - \Theta_l). \tag{7}$$

The turning point described by conditions (5)–(7) determines an instability of this system and an inverse change—a decrease—in the radius of the electron-hole droplets. To determine the period of a possible oscillatory regime, we can examine the condition under which we would have $\delta R = R - R_0 \sim e^{i\lambda t}$, and under which $\delta\bar{n},\delta\Theta^*$ would oscillate: $4\pi R_0^2 N\gamma v_T \tau_0 < 1$. We can find a minimum frequency of these oscillations:

$$\lambda = \sqrt{\frac{4\pi R_0^2 \gamma v_T}{\tau_0 c} \frac{n_T(\Theta_0^*)\varphi^2}{(\Theta_0^*)^2}}. \tag{8}$$

Using typical parameter values[3] for AgBr in (8), we find $\lambda^{-1} \simeq (1/\sqrt{0.6}) \times 10^3$ s, in extremely good agreement with the experimental value. We wish to emphasize that the relatively long period of these oscillations results from the circumstance that macroscopic parameters of the system—R, $\bar{n}$, and Θ—are oscillating. The changes in these parameters are determined in turn by the difference between fast processes: evaporation plus recombination, on the one hand, and condensation, on the other. This difference is arbitrarily small near the point described by conditions (5)–(7).

The analysis offered here requires some substantial refinements, which will be made in a more detailed paper. Nevertheless, the very observation of these oscillations due to a self-heating of electron-hole droplets is clearly of interest.

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