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Laser-induced heat radiation in borate glass

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Abstract. The heat radiation due to laser-induced heating of local inhomogeneities in borate glass was investigated experimentally. When borate glass is excited by a Q-switched YAG:Nd^{3+} laser, the heat radiation is reliably detectable without glass damage. The optical properties of the anti-Stokes emission investigated are described on the basis of a simple spherical-particle model. There is agreement between the experimental and calculated data. The local temperature is estimated to be about 10 000–15 000 K.

1. Introduction

According to the well-known definition of luminescence, to detect luminescence correctly it is necessary to eliminate both scattered light and heat radiation (HR) from the integral optical signal. The HR intensity is usually assumed to be negligible in comparison with the luminescence intensity. However, it is known that even an ordinary pulsed laser (not an extra-powerful one) can cause incandescence of some local inhomogeneities in condensed matter [1] and in gaseous phases [2]. Due to the high local temperature, a variety of processes occur in the overheated absorption centres. For example, as is shown in [3] for NaCl crystals, laser radiation can cause melting, vaporization, formation of cracks, and even migration of the absorption centre. Also, the phenomenon is complicated due to the variation of the inhomogeneities as regards size and shape, and optical and thermal properties.

To perform experiments correctly, an investigator needs to be aware of the phenomenological properties of possible HR. This paper presents the results of an experimental investigation of the laser-induced HR in alkali borate glass.

Real glasses usually contain a number of microscopic inclusions and structure inhomogeneities that arise, for example, from impurities in charge materials, from crucible wall grit, and from dust particles. The local absorption coefficient of such inhomogeneity can be high enough to cause laser-induced heating of this absorption centre up to the incandescence temperatures. Sometimes such centres initiate glass damage by the laser beam [1]. However, it is revealed that alkali borate glass is quite suitable for experimental investigation of laser-induced incandescence. The peculiarity is that borate glass produces visible HR at relatively low laser excitation intensities at which no glass damage takes place. This provides the possibility of measuring various optical properties of the phenomenon without frequent replacements of damaged glass samples being required.

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2. Experimental details

The experiments were performed with glasses of the following formula: $E_2O{\cdot}nB_2O_3$, where $n = 2$–8 and E = Li, Na, K. Some of the glasses were doped with 0.1–1 wt% of mercury-like ions (Tl^+, In^+, Pb^{2+}, Sn^{2+}, etc). The glasses investigated are transparent in the spectral region for which the experiments were performed.

The glass preparation procedure was as follows. The charge materials of commercial purity were used without any special treatment. The glass was formed in platinum crucibles at a temperature of 1300 K in the open air. The annealing time was about eight hours at a temperature of 650 K. The annealed glass samples were cut to dimensions of 5–10 mm, then ground and polished.

A Q-switched YAG:Nd^{3+} laser (wavelength ∼1066 nm, pulse duration ∼30 ns, second-harmonic wavelength ∼532 nm) with a computer-controlled spectrometer was employed for the measurements. Special care was taken to prepare the laser beam with a smooth crosswise intensity distribution. Sub-nanosecond structure in the laser pulses was not revealed. The spectrometer was able to process each pulse signal without accumulation. The data-averaging facilities were implemented in the software.

3. Results and discussion

When a glass sample is irradiated with a focused IR laser pulse, the HR is clearly visible to the naked eye as a homogeneous white track. When the same sample is irradiated with a second-harmonic laser pulse with the same energy and beam diameter, the HR is completely masked by intense scattered light. In both of these cases the HR is reliably detectable with a photomultiplier through a single-grating monochromator (with the appropriate cut-off filter). As the experiments show, the HR intensity is of the same order of magnitude for first-harmonic and second-harmonic laser excitation. When the sample is excited with plane-polarized laser radiation, the emission investigated is isotropic and unpolarized.

No correlation was found between the glass composition and the HR intensity. For glass samples of the same composition, the relative efficiency of the HR excitation varies from one sample to another. All of the data presented in this paper correspond to the $Na_2O{\cdot}6B_2O_3$ glass composition and 1066 nm excitation.

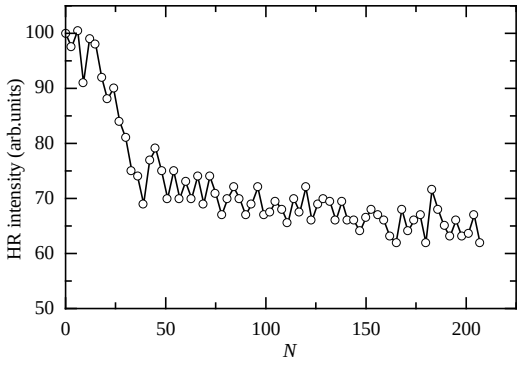

Figure 1. The exposure dependence of the HR intensity.

Detectable HR appears at laser intensities of about 100–200 MW cm^{-2}. No visible signs of damage are observed in the glass samples after laser irradiation. However, certain stable changes of the glass properties are detected after the laser irradiation. Figure 1 represents

the exposure dependence of the HR intensity, J, at $\lambda = 600$ nm. The number of laser pulses is denoted by N. The irradiation conditions were as follows: $F = 200$ MW cm^{-2}; pulse energy: 15 mJ; repetition rate: one pulse per second. Under such conditions, the heating of the irradiated glass volume is negligible. As seen from figure 1, when N exceeds 50 the J versus N curve becomes almost flat. This fact was taken into account when performing the experiments. All of the data represented in this paper were obtained after preliminary irradiation of the glass sample with $N = 100$–200 pulses. The nature of the observed partial fading is not clear from the available data. It should also be mentioned that visual inspection reveals no distinction between the irradiated and non-irradiated volumes of the glass sample.

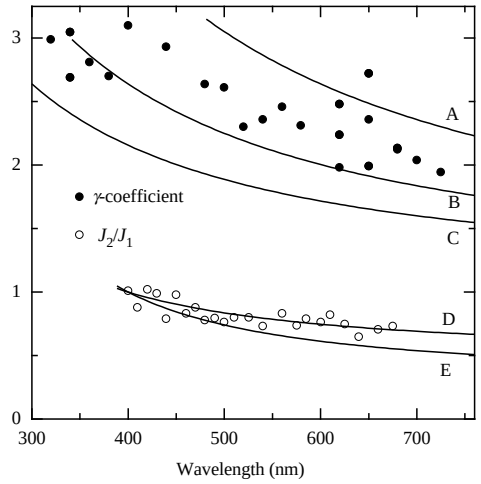

Figure 2. The spectral dependence of J_2/J_1 (open circles) and γ (filled circles). Solid curves: calculated for particle temperatures of 10 000 K (A), 15 000 K (B), 20 000 K (C), 30 000 K/15 000 K (D), 20 000 K/10 000 K (E).

The HR emission spectrum has no visible structure. When the sample is excited with 100 MW cm^{-2} radiation, the spectral distribution of the emitted light is approximately uniform over the range 400–700 nm (the spectral non-uniformity of the spectrometer sensitivity was corrected). The results of the spectral measurements are given in figure 2, where J_1 and J_2 are the HR intensities for 100 and 200 MW cm^{-2} excitation respectively. To ensure readability, the J_2/J_1 ratio is set to 1 at $\lambda = 400$ nm. The plot shows that the relative contribution of the 'blue' spectrum portion increases by approximately 25% when the excitation intensity is doubled.

Figure 3 represents the HR intensity J at a fixed wavelength ($\lambda = 450$ nm) as a function of the excitation intensity F. The J–F dependence is essentially non-linear. Increase of F by a factor of 3 results in increase of J by a factor of about 50. The filled circles in this plot represent the data obtained at the glass temperature, 450 K, while the open circles represent the data obtained at 300 K. The absence of thermal quenching may be taken as additional evidence that the emission investigated is not luminescence.

As one can conclude from figure 3, the relationship between J and F for a moderate deviation of F can be approximated as follows:

$$\lg J = \text{constant} + \gamma \lg F. \tag{1}$$

The γ-coefficient can be easily calculated from figure 3 as the slope of the curve. As is seen from figure 3, γ is not constant: it decreases with increase of F. Furthermore, the experiments show that γ depends on the HR wavelength. The spectral dependence of γ at $F = 400$ MW cm^{-2} is represented in figure 2.

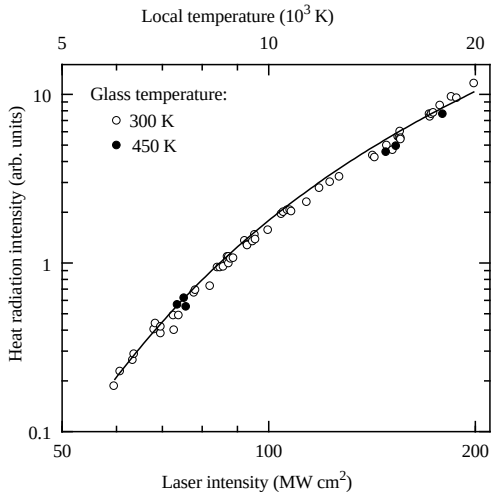

Figure 3. The HR intensity as a function of the excitation intensity. Solid curve: calculation; circles: experiment.

The HR kinetics measurements were performed using an oscillograph and a photomultiplier with 2 ns resolution. It is found that the shape of the HR signal is similar to the shape of the laser pulse. This is an important result. It indicates that the heat relaxation processes are relatively quick as compared with the laser pulse duration. That is why the heating up and cooling down of the inhomogeneities should be considered as quasi-stationary processes. In other words, one should not consider the laser action as the ballistic energy input and should use the appropriate energy-balance equations.

The above-mentioned optical properties of the laser-induced HR can be described by a simple model in which the inhomogeneities are considered as spherical particles of the same radius. Using the stationary approximation, the following heat-balance equation can be obtained:

$$FS_1 = -k\frac{\Delta T}{\Delta R}S_2 + \sigma T^4 S_3 \tag{2}$$

where k is the thermal conductivity coefficient, σ is the Stefan–Boltzmann constant, S_1 is the absorption cross-section, S_2, S_3 are the areas through which the particle loses energy by thermal conduction (S_2) and by heat emission (S_3), $\Delta T/\Delta R \simeq (T_0 - T)/\Delta R$ is the temperature gradient, and T is the particle temperature, $T_0 = 300$ K is the glass temperature. The proposed model does not take into consideration other possible mechanisms of particle energy dissipation, such as phase transitions in the absorption centre and in its neighbourhood, and generation of acoustic waves due to thermal expansion.

Some considerations should be noted concerning the phenomena investigated. The particle temperature is expected to reach thousands of degrees Kelvin. A high local pulsed pressure is also probable. The ultraviolet radiation emitted by the incandescent particles can produce photoionization in the glass. Such difficult conditions make it impossible to use the ordinary values of the material parameters in the calculations. That is why the thermodynamical parameters used in this paper are to be considered only as estimated values. It is also assumed that they are independent of temperature. As regards the particle radius, a plausible supposition is that the particles are small enough to form the observed homogeneous track. It can also be presumed that $S_2 = S_3 = 4S_1$.

The numerical evaluation shows that at particle temperatures below 25 000 K the energy supply (FS_1) provided by the laser beam with $F > 50$ MW cm^{-2} exceeds the radiative

energy loss ($\sigma T^4 S_3$) by a factor of more than 5. Therefore, for $T_0 \ll T < 25\,000$ K and for $F > 50$ MW cm^{-2}, the following relation can be derived from (2):

$$T = \text{constant} \times F. \tag{3}$$

That is, the particle temperature T is proportional to the excitation intensity F. So it becomes possible to calculate the J versus F relationship by using the well-known Planck formula for the black-body radiation density. The results of appropriate calculations are represented in figures 2 and 3 by solid curves. The agreement between the calculated and experimental data seems to be noteworthy.

The above-given results provide the possibility of estimating the particle temperature. As is seen from figure 2 (curve D) and figure 3, for $F = 100$ MW cm^{-2} the particle temperature is about 10 000–15 000 K. For $F = 400$ MW cm^{-2}, as seen from figure 2 (curves A and B), the local temperature is the same as for $F = 100$ MW cm^{-2}. This fact indicates the failure of the expression (3). It is apparent that, for laser intensities over 100 MW cm^{-2}, the proposed model becomes ill-founded and needs to be improved.

In addition, the particle temperature can be estimated by assuming that the total absorbed energy, $FS_1\tau_i$, where τ_i is the laser pulse duration, effects heating of the particle according to the formula $cm(T - T_0)$, where c is the specific heat and m is the particle mass. Then, for $F = 100$ MW cm^{-2}, the particle temperature is approximately $T = 10\,000$–$12\,500$ K for $R = 1$ μm and $T = 20\,000$–$25\,000$ K for $R = 0.5$ μm, which is in agreement with the above-given temperature estimate.

According to the estimated temperature, the maximum black-body wavelength is located in the UV spectral region (290 nm at 10 000 K), which confirms the observed decrease in intensity of the HR spectrum with λ.

As was shown by the kinetics measurements, the heat emission lifetime, τ, does not exceed the laser pulse duration. First, consider the temperature relaxation inside a spherical particle due to the thermal conduction. According to the classical problem, solved in mathematical physics, the time required for the temperature relaxation can be estimated [4] as follows:

$$\theta = cR^2\rho k^{-1}\pi^{-2} \tag{4}$$

where R is the particle radius, c is the specific heat, and ρ is the density. The calculated values of the HR lifetime τ are given in table 1 for various values of k and R. To transform θ into the corresponding τ, the above-mentioned Planck formula was employed. The data represented in table 1 correspond to the platinum particles. The τ-values for aluminium particles are 30% shorter. As is seen from the table, the quick relaxation (with a lifetime

Table 1. The lifetime of the HR due to the thermal conduction energy loss.

Thermal conductivity coefficient (W m^{-1} K^{-1})	HR lifetime (ns)				
	$R = 0.5$ μm	1 μm	2 μm	5 μm	10 μm
1	23.8	95.3	381	2380	9530
2	11.9	47.6	191	1190	4760
5	4.76	19.1	76.2	476	1910
10	2.38	9.53	38.1	238	953
20	1.19	4.76	19.1	119	477
30	0.79	3.17	12.7	79.4	317
50	0.48	1.91	7.63	47.7	191

shorter than the 30 ns laser pulse) is possible for certain combinations of R- and k-values. The thermal conductivity coefficient of 1 W m^{-1} K^{-1} (of the order of magnitude for glass) can explain the quick relaxation for particles with $R < 0.5$ μm. For larger particles ($R > 1$ μm), increased k-values should be assumed or the model used should be modified.

Second, consider the lifetime of the HR for a spherical particle due to the thermal relaxation to the environment. The overheated inhomogeneity can be considered as a spherical entity with increasing volume. The radius of the effectively heated volume can be estimated as $R + \sqrt{Dt}$, where D is the thermal diffusivity, and R is the particle radius for 'cold' conditions. Assuming conservation of energy with increasing volume, the relaxation time of the HR signal at the wavelength 450 nm is estimated to be approximately 15 ns for $R = 0.5$ μm.

Finally, consider the temperature relaxation of the incandescent particle due to the radiative energy loss. The energy dissipation can be described by the following simple equation:

$$cm\frac{\mathrm{d}T}{\mathrm{d}t} = -\sigma T^4 4\pi R^2 \tag{5}$$

where m is the particle mass. Integration of equation (5) leads to the following values of the heat emission lifetime (for platinum particles): $R = 0.5$ μm; $\tau = 1.4$ μs; $R = 5$ μm; $\tau = 14$ μs. Such lifetime values were not observed experimentally. This is an additional substantiation of our assertion of a secondary role of the radiative mechanism of particle energy loss.

4. Concluding remarks

The proposed model may seem to be oversimplified. It contains a number of assumptions, both hidden and specified in the text. Naturally, the model should be considered as a first order of approximation. However, it provides a real possibility of explaining most of the phenomenological properties of laser-induced incandescence investigated for borate glass.

The experiments described in this paper show that the heat radiation of the inhomogeneities in glass can be considered as a significant source of errors in experimental laser spectroscopy. If special care is not taken, the chances are that heat radiation can significantly distort the data obtained in some experiments with pulsed lasers. For example, a problematic situation arises when one investigates the quenching of luminescence by a powerful laser beam. In such experiments the laser-induced heat radiation can be of the same order of magnitude as the luminescence being quenched, and therefore errors can be expected.

Acknowledgments

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