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HOW TO DESIGN A HIGH-TEMPERATURE PERSISTENT SPECTRAL HOLE BURNING OPTICAL MEMORY

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Persistent spectral hole burning (PSHB) [1, 2] has opened a novel field of science and applications (see e.g. [3–5]). PSHB, based on zero-phonon lines (ZPL) [4–6], which are very narrow and of high peak intensity only at liquid helium temperature, is up to now actually confined to temperature below about 10 K. On the other hand, low-temperature PSHB has demonstrated its very high capacity in optical data storage and processing in a number of fascinating ways (see [3] and review papers [7–10]): frequency domain holography [3, 5, 11], time domain holography [5], space-and-time domain holography [3–5, 12], optical modelling of neural networks [13]. The PSHB applications are well implanted in laboratories, equipped with liquid helium facilities. For commercial applications the liquid helium is still not popular and PSHB at high temperature (room or at least liquid nitrogen temperatures) is required. The problem of high-temperature ZPLs and founded on them spectrally selective PSHB is interesting also for solid state physics.

Straightforward attempts to find bulk materials with high storage capacity $c = \Gamma_{inh} : \Gamma_{hom}$ (Γ_{inh} – inhomogeneous and Γ_{hom} – homogeneous ZPL widths) have not been really successful, firstly, because the high-temperature ZPLs are usually several orders of magnitude broader or less intense or both than the liquid helium ones. Further, the conditions for a narrow ZPL and large inhomogeneous broadening are to a certain extent contradictory. ZPLs are intense and narrow when the difference between the adiabatic potentials (potential energy surfaces) of excited and ground electronic states is small. On the other hand, the same condition leads to small inhomogeneous broadening [14]. Thus it is difficult to get large the decisive for data storage parameter – the information storage capacity c. However, in principle it is not impossible to find a high-temperature high-c material. The efforts have to be continued but parallel attention should be paid to ways of designing devices which use materials with quite modest c values.

The objective of this short note is to discuss the outlooks for that kind of devices.

One possibility is to use an impurity activated solid having in spectrum a relatively narrow homogeneous high-temperature ZPL, e.g. 1 cm^{-1} at room or 0.1 cm^{-1} at liquid nitrogen temperatures. Inhomogeneous broadening can be magnified to some extent (e.g. tenfold) manipulating the structure and composition of the host solid. Up to a hundred spectrally distinguishable holes can be burned in at room temperature (see papers by Jaaniso and Bill in [5]). Make an optical fibre of this material and put the fibres together into a bundle of fibres, and form thus a data storage and processing device (as proposed in [15]) in which each fibre can be addressed in hole burning write-in and optical read-out as an independent spatial pixel. Thus a bundle of 1 cm^2 cross section comprising 1000 fibres provides about 10^5 space-and-frequency domain pixels. The length of fibres is not important. In the case of long fibres different sections of them can be activated with different impurities and thus enhance the capacity by a factor of 10.

Another possibility is to utilize better the frequency dimension finding ways to get narrower ZPLs. If we place the PSHB material in the near field of the evanescent waves, e.g. activate with impurities the coating of a fibre (or the surface of a waveguide plate as in [16]), we can utilize the stronger gradients of the electric field compared to those of the far field and get thus two benefits: enhancement of inhomogeneous broadening and softening of selection rules for forbidden transitions.

A proposal to try a high-temperature PSHB data storage device could be formulated in the following six points.

(1) Find a ZPL, narrow at high temperature. Perhaps the best ZPLs could be found for some forbidden transitions having very small oscillatory strength. Note that at $T \to 0$, because of the very small radiative linewidth, the homogeneous peak absorption cross section (PACS) can be rather large, e.g. about 10^{-11} cm^2, if other transitions beside radiative ones are excluded [17]. At high temperatures the situation is different: nonradiative processes are strongly prevailing. Nevertheless, the small oscillatory strength is not the principal obstacle: the very large

radiative PACS, exceeding the geometrical size of an impurity by a factor of 100 000, opens some prospects.

(2) Try to distort the symmetry at the site of impurity and thus gain two effects. First, soften the selection rules and thus enhance the oscillatory strength. Second, enlarge the inhomogeneous broadening.

(3) Place the impurities in the near field of the exciting light. A possible way to do this is to cover a waveguide (optical fibre or a planar waveguide; see [16]) with the impurity-activated solid as a coating so that the evanescent part of the laser light propagating along the fibre will perform the excitation.

(4) Make the interface between the fibre and coating rich of tiny roughnesses having sizes smaller than the exciting wavelength (e.g. implanting grains smaller than the wavelength of a proper optical material into the coating). The exciting evanescent waves will be distorted and forced to have faster spatial changes (higher spatial harmonics enhanced) and thus the gradients in the exciting field can be large compared to those in the freely propagating light in the far field of the same frequency. This will mean enhancement of the higher multipole (beyond the dipole) transition probabilities (which are not actually forbidden, but very small because the wavelength of the conventional exciting (freely propagating) light exceeds the size of the impurity centre by about three orders of magnitude).

(5) Hope that the dose of irradiation required to burn holes is not hopelessly large and the holes are of good shape and stable enough even at high temperature. For a host disordered in the vicinity of the impurity photophysical mechanisms of hole burning can be effective. To implant a co-activator to enhance the hole burning is attractive, but the energy transfer from the principal activator must not destroy the spectral selectivity of the hole burning.

(6) Compose the fibres coated as described above in a device proposed in [15].

A more detailed description of the expected characteristics and possible difficulties will be published elsewhere in the context of high-temperature ZPLs.

Some remarks.

(1) The statement in the second sentence of requirement 5 is to a certain extent contradictory to a stable hole: the disordered host usually opens also easy pathways for diffusion and spectral diffusion, especially when there is a contribution to disorder by vacancies and other "empty places". The first part of this requirement is also not easy to match: if the excitation energy is absorbed in a zero-phonon transition, which ends in a well screened excited electronic state, Stokes losses are small and the transfer of its energy to environment is hindered and creation of photophysical hole-burning processes is difficult.

(2) The really narrow at low temperatures homogeneous ZPL widths of forbidden transitions can be measured via the dephasing times by photon-echo (and other time domain) methods. If a very narrow spectral hole is

burnt, the frequency domain measurements based on Doppler shifts [18] can also be used.

(3) Note that the situation reminds us to some extent of the surface enhanced Raman scattering [19], which provides the enhancement of the scattered light (or site selective fluorescence from a broad inhomogeneous body of impurities?) up to six orders of magnitude.

(4) The use of different dopants which provide not completely overlapping inhomogeneous bands to activate different sections of the fibre can be a favourable approach to get a summary inhomogeneous band quite a few times broader than that for a single dopant and thus enhance *c*. The other option, already shown experimentally to be prospective, is to put together a "sandwich" of layers of different host materials (ionic mixed crystals), activated with the same activator (Sm^{2+}) [20]. Different activators in one host have been realized with organic hole burning materials [13].

(5) Up to now the best results for room-temperature PSHB have been obtained with narrow f–f transition's ZPL in rare earth ions (see [20] and references therein). In these papers inhomogeneous broadening is magnified by using the disordered composition of ionic host. Seven holes for a single sample are demonstrated and the possibility of burning a few tens of holes using composite materials is shown. In [21] room-temperature PSHB in a neutron-irradiated diamond is reported.

(6) To have a very rough estimate in numbers, it is supposed (for bulk PSHB solids) in [14] that the decisive parameter, capacity *c*, decreases by a factor of 10 with each step in the sequence of increasing temperatures $2\ K \rightarrow 10\ K \rightarrow 77\ K \rightarrow 300\ K$. Thus, if at 2 K 100 000 holes are available, at room temperature 100 holes have to be considered as a roughly estimated limit for the best high-temperature PSHB bulk materials. A few tens of holes have already been reported [20]. To continue the line of intuitive rough estimates, I would say that the coated fibre with a number of different activators in the near field design could enhance the number of high-temperature holes by a factor of 10, i.e. up to 1000 holes in one fibre at room temperature should be possible. Thus, an optical cable comprising 1000 fibres [21] can process one million bits; taking into account the possibility of utilizing also the differences in the depth of holes, an additional factor of 2^3–2^5 can be gained. This estimate looks too optimistic but it does not contradict the laws of nature.

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