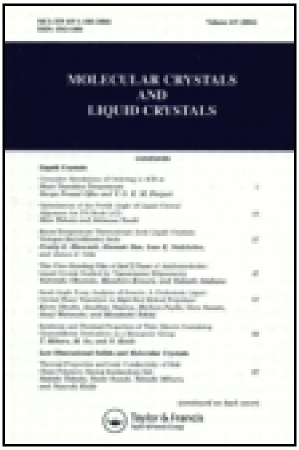

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# Persistent Spectral Hole Burning Applications in Distributed Data Storage and Processing

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# PERSISTENT SPECTRAL HOLE BURNING APPLICATIONS IN DISTRIBUTED DATA STORAGE AND PROCESSING

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Abstract Brief review of zero-phonon lines, persistent spectral hole burning (PSHB) and its applications is given. The provided by PSHB additional dimension - the spectral one in optical data storage allows in principle to get densities up to $10^{12}$ bit $cm^{-2}$ or $10^{16}$ bit $cm^{-3}$. In both cases an $10^{-16}$ $cm^{-3}$ cell comprising on average only 100 impurity molecules is involved to store a bit. It is a two small amount because of fluctuations in the distribution of impurities over the cells. Parallel processing and distributed data storage, realized in PSHB holography and optical modelling of neural networks, are prospective to utilize the full capacity of PSHB storage and processing.

## 1. INTRODUCTION

Persistent spectral hole burning (PSHB) was found independently by two groups (at Troitsk, near Moscow and Tartu, Estonia) twenty years ago [1,2]. Fast growth of studies started ten years ago when several well equipped laboratories of solid state laser spectroscopy in USA, Germany, Switzerland, and France got also Involved. A series of international conferences has started (The third one - Ascona, Switzerland, September 1992), several hundreds of papers and first monographs have been published [3,4].

High resolution PSHB matrix isolation spectroscopy of molecules has been developed, including studies of chlorophylls and its relatives [5], and even photosynthetic units [6]. A number of applications have been proposed among them as one of the most attractive potential applications - PSHB data storage and processing.

## 2. ZERO-PHONON LINE

Zero-phonon line (ZPL) - a characteristic feature of the low temperature impurity spectra of solids[7,4,3] - is the foundation stone of high-resolution persistent spectral hole burning.

Theory tells us that the ZPL's linewidth at zero temperature is given by the lifetime of the excited electronic state [7]. ZPL is accompanied by a broad phonon sideband (PS). The Debye-Waller factor (DWF) shows how much of the bulk intensity of a vibronic transition belongs to ZPL. When temperature increases, DWF decreases rather rapidly. The increasing temperature broadens also to some extent the ZPL. In most molecular systems there are no ZPLs at temperature above 20-40 K.

The temperatures used in PSHB studies are usually 1.8-2 K and the typical spectral characteristics are roughly the following: 1. ZPLs linewidths are $10^{-3}$-$10^{-4}$ $cm^{-1}$; 2. The most important for spectral resolution parameter - ratio of the transition frequency to the linewidth is up tp $10^{7}$-$10^{8}$; 3. Broadening of ZPL, caused by scattering of phonons at the excited impurity, is approximately the same as linewidth determined by the radiative lifetime of the excited electronic state; 4. Relation of the ZPLs peak intensity to that of the PS is about $10^{5}$-$10^{6}$, (the absolute value of absorption cross section see Section 7).

## 3. INHOMOGENEOUS BROADENING. PERSISTENT SPECTRAL HOLE BURNING

ZPL of a single molecule is so narrow and therefore sensitive that the irregularities of a real solid influence its frequency very strongly compared to the linewidth. A body of impurities has an inhomogeneously broadened ZPL whose width may be from one $cm^{-1}$ (single crystals) up to a thousand $cm^{-1}$ (glassy metrices). A low temperature impurity spectrum comprises two parts: a really continuous PS and a pseudo-continuous body of sharp and intense ZPLs. The latter part is actually a broad band of thousands of sharp resonences and provides a wonderful device to perform fine physics.

Under monochromatic laser excitation these impurities, whose ZPLs are in resonance with it, undergo excitation-deexcitation cycles with high repetition rate and chemical changes in them or in their environment may be created. ZPLs of the changed impurities are shifted far away from their initial positions and a hole (dip) is formed at the laser frequency. The changes are fixed by rearrangements of the positions of the atoms and molecules and can have very long lifetimes.

PSHB is a tool to control by illumination (of decent intensity, because the doses matter in the first place) the optical properties - the absorption coefficient and related to it refraction index. PSHB turns inhomogeneous broadening of ZPLs into a useful feature.

## 4. PSHB FILTER OF TRANSPARENCY. DATA STORAGE

The broad absorption band with a sharp hole is a narrow-line ($10^{-3}$-$10^{-4}$ $cm^{-1}$) wide-apperture spectral filter. The spectrum of transparency may be more complicated - comprise two or more lines or have various continuous profiles. An illumination subject to be filtered can burn for itself a profile of transparency, matching its spectrum of intensity.

PSHB optical memory is actually a transparency filter of specific profile. We can attach to a diffraction limited $10^{-8}$ $cm^2$ spatial spot (the wavelength of light being $10^{-4}$ cm) up to $10^4$ bits of information defining the content of a bit by absence or presence of a hole at a certain frequency in the absorption band. The total density becomes really high: $10^8 \times 10^4 = 10^{12}$ bit $cm^{-2}$ and $10^{12} \times 10^4 = 10^{16}$ bit $cm^{-3}$ for a possible fourdimensional ($x$, $y$, $z$, $\nu$) memory. A booklet of ten 10 cm x 10 cm memory sheets for the threedimensional case ($x$, $y$, $\nu$) comprises $10^{15}$ bits. To write in and read such tremendous amounts of information in a bit-by-bit way becomes quite a time consuming task. To burn a hole usually takes seconds. The shortest time reported (for a very shallow one) is about 30-100 nanoseconds [8] and even such a speed leaves the writing-in time unreasonably long.

There is a principal limit for both - write-in and read-out: to fix or detect a hole with the accuracy $\delta\nu$ in the frequency domain we have to illuminate at least as long as $\delta\nu^{-1}$.

The message is that for the high capacity PSHB memories parallel processing becomes a must.

Another point is that simply extending the bit-by-bit memory to the frequency domain we are not making use of the new quality of the new physical dimension - the frequency. We have to search for and employ new ways for data processing. This is the path or thinking that leads us to the time-and-space domain holography.

Two ways of fully parallel processing based on PSHB have been realized: holography (stationary spectral holography [9] and time-and-space domain holography [10]) and models of neural networks [11]. In both approaches parallel processing is accompanied by distributed data storage. The latter becomes also a must when the mentioned above storage densities are used.

Focussing with the diffraction limited accuracy means that the depth of the focal cannot be smaller than wavelength and the volume of a PSHB pixel ($\Delta x \Delta x \Delta \nu$) is $10^{-16}$ $cm^3$. Impurity concentration of $10^{18}$ $cm^{-3}$ is the maximum value at which we can still suppose that there is no interaction between impurities destroying the spectral selectivity. It means that there is on average only 100 impurity molecules to fix a bit. This is too small amount to avoid errors in distinguishing between the changes caused by hole burning and simply by fluctuations of the initial distribution of impurities. Distributed data storage used in holography and models of neural networks is much less sensitive to errors and probably allows better utilize the high capacity of PSHB memories.

## 5. PERSISTENT SPECTRAL HOLE BURNING HOLOGRAPHY

Powerful PSHB applications for data storage and processing are space-and-frequency domain [9], space-and-time domain [10], space-time-and-frequency domain [11] holographies. These advanced approaches are based on very high spectral resolution of PSHB materials and methods to control their optical properties by illumination, also on distributed data storage and fully parallel coherent write-in and read-out procedures. Results are impressive: storage and playback of thousands of holograms written at different frequencies in a small piece of PSHB material [9,12], storage and playback of all the classical features of a light pulse (i.e. three-dimensional image in colour, including the polarization characteristics) and its dependence on time ("holographic movie of events") [10], conjugation of wavefronts [10], imaging through scattering media [13], shaping of light pulses [14], associative recall of events [15]. All holographic approaches are rather insensitive to errors in storage and processing. Associative properties of the time-and-space domain holography is a fine demonstration of it: a fraction of the event can recall the whole one (in space and time!)

## 6. ERROR-CORRECTIVE PSHB OPTICAL MODELLING OF NEURAL NETWORKS

Capability of PSHB to provide frequency as a new dimension and enlarge (in principle up to $10^4$-$10^5$ times) the number of possible interconnections has been shown[11]. Four models and various ways of PSHB experimental realization have been considered. E.g. in the third case quadratic auto-associative memory is coded in three dimensions (coordinates x, y and $\nu$) and materializes 32 x 32 x 32 = 32768 optical interconnections; the probe vector is taken as 32 x 32 spatial matrix (coordinates x, y); the output is one-demensional, consists of 32 bits along the frequency axis $\nu$ and corrects 4 erroneous bits. Experimental details are described in [11].

In the first three experiments the read-out of the frequency-domain bits is accomplished sequentially by tuning the wavelength of the read-out laser. Obviously it makes the procedure long without any real reason to be long.

In the fourth case the simultaneous parallel read-out not only for the spatial locations but also for all the frequency-domain read-out bits has been realized experimentally. All the output information is comprised in the only one output pulse and coded in frequency, what sould be convenient to transfer it along optical waveguides. Actually here the picosecond read-out pulse accomplishes optically paralell tensor-matrix multiplication, the lens performing the summation part of the calculation.

## 7. SINGLE IMPURITY MOLECULE DETECTION AND SPECTROSCOPY. NEW HORIZONS FOR DATA STORAGE?

The peak value of the absorption cross section of ZPL in absence of non-radiative quenching of excited electronic energy differs from the universal value for a free atom $I_0^{at} = \lambda^2/(2\pi)$ by two factors:

$$I_0(T) = I_0^{at}(T)\ \alpha\ (T)\ \beta\ (T) = \lambda^2/(2\pi)\ \alpha\ (T)\ \beta\ (T),$$

where $\alpha$ (T) stays for the DWF and $\beta$ (T) - for the temperature broadening in the units of ZPL's radiative width. As an convenient estimate for molecular impurities in organic molecular matrices at liquid helium temperatures we can take $\alpha(T) = \beta(T) = 0.5$. The resonance excitation wavelength via ZPL is $\lambda = 1000$ d, where d is the geometrical size of the impurity molecule. So the ZPL's absorption cross section exceeds the geometrical size of an impurity molecule by a factor about $10^5$. That is why ZPL is also the foundation stone for the spectroscopy of a single impurity molecule spectroscopy - a new prospective field of matrix spectroscopy of molecules [16].

Does the possibility to detect a single molecule open new routes for optical data storage and processing? A reliable way to detect a single molecule is multiple resonance excitation and collecting a sufficiently large number of fluorescence photons emitted by the impurity. The advantage is that statistical fluctuations involved in the measurement may be transferred from the number of impurities per pixel $(\Delta x \Delta y \cdot \Delta \nu)$ to the number of photons counted and the latter may be made large. It opens the possibility to clearly distinguish between a pixel, in which is one or more molecules (1 written in), and a pixel, where none is present (0 written in). So in principle it is possible to read a given information stored with a density about one bit per one molecule. Long time required to collect a large number of photons emitted from a single molecule is certainly a disadvantage, creating difficulties for coherent processing and parallel read-out. The main task to be solved is how to write in the information. Who and how could provide the PSHB material with very smooth distribution of about N molecules over N pixels?

## REFERENCES


1. B.M.Kharlamov, R.I.Personov, L.A.Bykovskaja, Opt. Commun. 12, 191 (1974).
2. A.A.Gorokhovskii, R.K.Kaarli, L.A.Rebane, JETP Lett. 20, 216 (1974); Opt. Commun. 16, 282 (1976). L.A.Rebane, A.A.Gorokhovskii, J.V.Kikas, Appl. Phys. B29, 235 (1982).
3. W.E.Moerner, ed., Persistent Spectral Hole Burning; Science and Applications (Springer-Verlag, Berlin, Heidelberg, 1988).
4. O.Sild, K.Haller, eds. Zero-Phonon Lines and Spectral Hole Burning in Spectroscopy and Photochemistry (Springel-Verlag, Berlin, Heidelberg, 1988).
5. R.A.Avarmaa and K.K.Rebane, Spectrochimica Acta, 41A, 1365 (1985).
6. Technical Digest on Persistent Spectral Hole-Burning: Science and Applications, Conference 26-28 Sept. 1991, Monterey, California (Optical Society of America, Washington D.C., 1991, Vol. 16 ).
7. Karl K.Rebane, Impurity Spectra of Solids (Plenum Press, New York, 1970).
8. M.Romagnoli, W.E.Moerner, S.M.Schellenberg, M.D.Levenson, G.C.Bjorklund, J. Opt. Soc. Am. B1, 341 (1984); A.Winnacker, R.M.Shelby, R.M.Macfarlane, Opt. Lett. 10, 350 (1985).
9. A.Meixner, A.Renn, U.Wild, J. Chem. Phys. 91, 6728 (1989).
10. T.W.Mossberg, Opt. Lett. 7, 7 (1982). A.Rebane, R.Kaarli, P.Saari, A.Anijalg, K.Timpmann, Opt. Commun. 47, 173 (1983). P.M.Saari, R.K.Kaarli, and A.K.Rebane, JOSA B3, 527 (1986).
11. A.Rebane and O.Ollikainen, Opt. Commun. 83, 246 (1991). O.Ollikainen and A.Rebane, in Optical Memory and Neural Networks, SPIE, 1621, 351 (1991). O.Ollikainen, A.Rebane, K.Rebane, J. Optical and Quantum Electronics, submitted.
12. A.Renn, A.J.Meixner and U.P.Wild, J. Chem. Phys. 92, 2748 (1990). A.Rebane, S.Bernet, A.Renn and U.P.Wild, Opt. Commun. 86, 7 (1991).

13. A.Rebane, J.Feinberg, Nature, 351, 378 (1991).
14. H.Sõnajalg, A.Gorokhovskii, R.Kaarli, V.Palm, M.Rätsep, P.Saari, Opt. Commun. 71, 377 (1989).
R.Kaarli, P.Saari, H.Sõnajalg, in /6/ p. 32.
15. A.Rebane, Opt. Commun. 65, 175 (1988).
16. W.E.Moerner and L.Kador, Phys. Rev. Lett., 62, 2535 (1989);
W.P.Ambrose and W.E.Moerner, Nature, 349, (1991);
M.Orrit and J.Bernard, Phys. Rev. Lett. 65, 2716 (1990).