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RESEARCH LAB SAN JOSE CA W E MOERNER 15 MAR 85 TR-8

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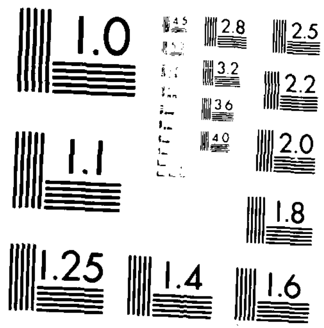

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Persistent spectral hole-burning (PHB) is a photoinduced process in low temperature solids that may lead to a possible future application, frequency domain optical storage. The feasibility of such a data storage device depends critically upon having recording materials that undergo spectral hole-burning with certain well-defined characteristics. It is a stimulating challenge for the laser spectroscopist, photochemist, and physicist to find suitable materials and to devise detection techniques that make this application possible.

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Laser-Light-Induced Physical Processes in Optical Materials: Persistent Spectral Hole-Burning

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LASER-LIGHT-INDUCED PHYSICAL PROCESSES IN OPTICAL MATERIALS: PERSISTENT SPECTRAL HOLE-BURNING

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Abstract: Persistent spectral hole-burning (PHB) is a photoinduced process in low temperature solids that may lead to a possible future application, frequency domain optical storage. The feasibility of such a data storage device depends critically upon having recording materials that undergo spectral hole-burning with certain well-defined characteristics. It is a stimulating challenge for the laser spectroscopist, photochemist, and physicist to find suitable materials and to devise detection techniques that make this application possible.

Introduction

One potentially important application of laser spectroscopy of molecular and ionic defects in solids at low temperatures is the use of persistent spectral hole-burning to form a frequency domain optical storage system [1] [2] [3] [4] . PHB (also called photochemical hole-burning although photophysical hole-burning also occurs) has the interesting property of allowing as many as 1000 or more bits of information to be stored in the volume irradiated by a single focused laser beam. PHB utilizes an additional dimension beyond x-y spatial dimensions to achieve this dramatic increase in areal storage density. In effect, various bits are addressed by the laser frequency or wavelength at which they are stored, hence the name "frequency domain optical storage" means the use of PHB for optical storage of digital data. Of course, the feasibility of such a data storage scheme depends critically upon having recording materials that undergo spectral hole-burning with certain well-defined characteristics. It is a stimulating and interdisciplinary challenge for the solid-state spectroscopist, photochemist, and laser physicist to find suitable materials and to devise detection techniques that make this application possible.

The first requirement for spectral hole-burning is that molecules (or other absorbing centers[5]) must be dispersed in a suitable transparent matrix and cooled to liquid helium temperatures. The molecules must be sufficiently rigid and sufficiently uncoupled from the host lattice that the lowest energy optical absorption is a "zero-phonon" absorption, i. e., a purely electronic or vibrational transition. If the host matrix were a perfect crystal, the sample absorption would appear as in the upper half of Figure 1: all molecules would absorb at the same frequency with the same width, Γ_H, called the homogeneous width. However, in a real crystal, glass, or polymer (see the lower half of Figure 1), slight differences in the environment around each molecule caused by local strains or nearby defects cause the various molecules absorb at slightly different wavelengths. For this case, the way in which the sample absorbs light in the lowest energy absorption can be viewed as a superposition of narrow absorption lines of width Γ_H from the various molecules distributed throughout the sample. The result is a broad, smooth absorption line of width Γ_I that is said to be inhomogeneously broadened. To give a crude analogy, one may say that the ensemble of identical molecules absorbs many different colors, but each molecule only absorbs a given, well-defined color. It is the fact that the local environments of the various molecules are different that makes the various molecules absorb slightly different laser frequencies.

The basic process of spectral hole formation

If a laser beam with a well-defined frequency ω_l irradiates such a sample, only those molecules that are in resonance with the laser can be excited (see Figure 2 (a) and (b)). If the laser linewidth is less than Γ_I or better yet less than Γ_H as is usually the case, one can selectively excite different classes of molecules simply by tuning the laser. The number of different classes of molecules that can be accessed in this way is on the order of the ratio of the inhomogeneous width to the homogeneous width, Γ_I/Γ_H, a factor which can range from 100 to 10^4. (It is precisely because we want this factor to be large that liquid helium temperatures are required. At higher temperatures, Γ_H becomes larger than Γ_I due to phonon broadening effects.) If the laser is focused to a small spot, the size of that spot is limited by the wavelength of the light itself to be greater than roughly one micron in diameter. Even within the small volume irradiated by a one micron diameter laser beam, a narrowband tunable laser can probe different groups of molecules simply by changing the laser wavelength.

Now if the molecules undergo a photo-induced change when light is absorbed (see Figure 2(b)) such that the product does not absorb at the laser wavelength, the optical absorption at ω_1 decreases. Such light-induced changes may involve a photochemical change in the molecule itself where a product is formed [7] [8] or a light-induced change in the local environment near the molecule [9] [10] . The resulting decrease or "dip" at ω_1 in the absorption line is called a spectral hole. If the photoinduced change in the molecule is persistent on time scales of months or years at low temperatures, the spectral holes at various locations within the line can be used to encode binary information, where, for instance, the presence of a hole at a particular frequency within the inhomogeneous line might correspond to a binary "1" and the absence of a hole might correspond to a binary "0". Figure 3 gives an example of a 19 bit hole pattern written in the absorption line of free-base phthalocyanine molecules in a poly(methylmethacrylate) (PMMA) host[4]. Since in one laser spot many groups of molecules are available, 1000 or perhaps 10,000 bits can, in principle, be stored in the frequency domain in the volume illuminated by a focused laser beam, resulting in potential areal storage densities as large as 10^{11} bits/cm^2. This type of data storage system can feature fast random access and high data rates as well as high areal density, and is called a frequency domain optical storage system. As one can see, PHB utilizes many of the unique properties of laser radiation: narrow linewidth, spatial coherence, and tunability. The ability to use PHB in a future optical storage application depends heavily upon the dynamics of the photo-induced change that occurs in the absorbing centers such as hole width, quantum efficiency, presence of bottlenecks, etc.

General systems and materials issues

It is to be emphasized that even though the frequency domain optical storage concept sounds simple, this idea can hardly be regarded as a fully practical technology at the present time. Indeed, no single material has been found that possesses all the required properties. In the rest of this paper, several crucial materials and engineering issues and their partial solutions will be described. Table 1 lists some of the properties that are required of the reading and writing system engineering and Table 2 list some of the required materials properties in order to produce practical data storage and retrieval. The middle column lists the material or technique with which the property has been demonstrated. For each entry in the table, the reader is urged to consult the references for more detail.

Table 1: Engine[illegible]ng Requirements

Property	Material or Technique	Reference
Hole detection	FM spectroscopy, λ modulation, HUMPH, FREMPOLSPECT	[11] [12] [13] [14]
Reading and writing with diode lasers	current and temperature tuning	[15] [16]
Focus/servo in liquid He	Ronchi grating, dither	[17]
No crosstalk between adjacent spots	measured: R' in LiF	[18] [19]

In general, most of the engineering or systems requirements have been shown to be solvable within the current state of the art, although single-mode diode lasers with wider tuning ranges would be desirable. In particular, recent research on FM spectroscopy has demonstrated the shot-noise-limited sensitivity of this method for detection of unmodulated absorptions[20].

Any reasonable system design also places demands on the properties of the photoactive hole-burning material itself, and Table 2 summarizes some of the principal requirements.

Table 2: Material Requirements

Property	Material or Technique	Reference
$\Delta\nu_{hole}$ = 100-500 MHz	color centers, H_2Pc-PE,...	[6] [21]
GaAlAs compatibility	R' in LiF(8330 Å) H_2Pc-PE in H_2SO_4	[22] [23]
Reversible burning	H_2Pc, H_2P, ...	[8] [24]
Long hole lifetime at low temperatures	Quinizarin in glasses	[25]
Fast burning (≃30 ns/bit)	H_2Pc-PE	[26]
Fast burning, high SNR, fast reading, focused spot	difficult for single-photon mechanisms	[27]
Gated hole-burning mechanism	Sm^{2+}-BaClF, carbazole in boric acid	[28] [29]
Room temperature cycling	Sm^{2+}-BaClF	[28]

Most of these requirements involve fairly obvious considerations. For instance, the material must be active at GaAlAs laser wavelengths, because these lasers are tunable, inexpensive, compact, and readily available commercially. To give another example, holes must be burned in times on the order of 30 ns in order to handle the high data rates expected for a large data base. Recently, detectable holes have been burned in less than 100 nsec using a high sensitivity organic system and FM spectroscopy detection[26]. The existence of most of the required properties in the upper half of the table has been separately demonstrated in a number of inorganic and organic systems, which attests to the high rate of recent progress in this area.

Single-photon processes

Let us now focus on the entry in Table 2 that is third from the bottom: a material in order to be useful in a practical storage system must simultaneously show all the required properties: the ability to form deep holes in short burning times and yet allow fast reading at high signal-to-noise ratios with focused beams. For example, a material with low hole-burning efficiency would be quite easy to read without serious destruction of the written holes, but such a system would be difficult to write with high contrast in short burning times. Conversely, a system that shows fast burning due to a high quantum efficiency for hole production would be difficult to read without burning of the unwritten centers by the tightly focused reading beam.

To understand this problem more fully, a thorough analysis of the coupled reading-writing problem in small spots for materials with single-photon or monophotonic hole-burning mechanisms has been recently completed[27]. Figure 4 schematically shows the energy level structure appropriate to single-photon mechanisms. An incident photon flux F is absorbed with an absorption cross section σ. A given center that has absorbed a photon can either decay to the ground state with rate Γ_1 or undergo the transformation leading to hole-burning with probability or quantum efficiency η[30]. The essential problem with single-photon processes is that there is no threshold in the hole formation mechanism. The process of hole detection requires the absorption of photons by the remaining unburned centers, and if high powers are necessary to detect the dip in the absorption line with adequate signal-to-noise, the hole pattern will be destroyed by the reading laser beam (i.e., a "trench" will be formed over the spectral region probed by the reading laser). This optimization problem has been analyzed in detail to determine whether any combination of single-photon materials parameters would yield acceptable reading performance [27]. Figure 5 shows the results of this materials analysis. The two fundamental materials parameters are naturally the hole-burning quantum efficiency η and the absorption cross section σ. A material with specific values of η and σ would be represented by a point on the η-σ plane shown in the figure. Furthermore, since the analysis requires that the absorption coefficient α_0 of the sample be fixed at a constant value that optimizes the signal-to-noise ratio, the concentration of centers necessary to keep α_0 fixed (top axis) must decrease as the cross section increases. In other words, for given η and σ, the concentration listed on the top axis must be achieved in order to optimize signal-to-noise ratio. The lower right triangular region represents the class of materials parameters that would not yield acceptable signal-to-noise ratios for any number of reads. In other words, a useful single-photon material must have <u>low</u> absorption cross section and high quantum efficiency, with sufficient solubility in the host to yield the concentrations shown on the upper axis of the figure. Within the upper left allowed region, the lines represent contours of constant numbers of reads. The achievable number of reads varies from 1 to greater than 10^4; again superior performance results from low cross sections and high quantum efficiencies. (η values greater than 0.1 are not allowed, because hole broadening would occur due to excessive photochemistry, and σ values less than 10^{-15} cm^2 would require prohibitively high densities of centers.)

The results in Figure 5 show that a new challenge exists for workers in the field of PHB: find single-photon materials that have values of quantum efficiency and cross section that fall within the allowed region, as well as solubilities that allow concentrations shown along the top axis. One might look for hole-burning in partially allowed transitions, such as n-π^* transitions of organic molecules and d-f transitions of divalent rare earth systems. However, the parameter space of useful η-σ values is somewhat small.

Gated PHB

One way around this problem would be to consider those materials that do not suffer from the intrinsic limitations of single-photon, monophotonic processes with no threshold. One such class of materials are those with two-step PHB mechanisms, called gated mechanisms in the second to the last entry in Table 2. Figure 6 shows the general idea of gating. The wavelength λ_1 excites a homogeneous packet within an inhomogeneously broadened line. If no external field is present, the center returns to the original ground state without forming a spectral hole. However, in the presence of λ_1 and some external field, the center undergoes the transformation leading to hole-burning. This is the origin of the term "gating": the external field acts as a gate on the hole-burning process. The hole may be detected using λ_1 alone, since hole detection is merely probing the ground state distribution of those centers that did not react to form the spectral hole. Since the external field is not present during the reading process, hole detection may then be nondestructive. In effect, gated mechanisms add a "threshold" to the writing process, which uncouples the reading and writing processes. The external field may be a second photon of a different wavelength or the gating could perhaps be achieved by any other external field, such as electric field, magnetic field, stress field, and the like.

Recent materials research at IBM has been devoted to a search for gated or two-color or photon-gated PHB mechanisms [31] in inorganic as well as organic materials. In photon-gated PHB, two photons (of different wavelengths) are required for the photoinduced change leading to hole-burning (writing). In recent experiments, photon gated PHB has been observed for the first time for Sm^{+2} ions in BaClF crystals[28]. The mechanism is thought to be that shown schematically in Figure 7. The first wavelength, λ_1, excites the system from the ground state to a an intermediate level. Extended irradiation at λ_1 produces essentially no hole production, but brief periods of simultaneous irradiation with λ_2 allows deep holes to be burned at λ_1. The second photon is thought to excite the ion from level 2 to the conduction band or to an autoionizing level and the photoejected electron is subsequently trapped in the host matrix.

This material has a further exciting and unexpected property: a pattern of holes burned at low temperature persists even after cycling up to room temperature and back down to helium temperatures. Apparently, the electrons are trapped at sites with very high barriers, and the relaxation of strains upon cycling is small enough to prevent loss of the site selection. This discovery shows that materials exist that can relieve one of the most serious concerns with frequency domain optical storage: volatility of the stored data. Further experiments are in progress to understand this novel process, and the reader is urged to consult the references for more detail[28].

Moreover, two-color photon gated PHB has also been observed in an organic material composed of carbazole molecules in boric acid glasses[29]. This important results proves that gated mechanisms exist in organics as well as inorganics. The presumed mechanism is depicted schematically in Figure 8. Upon excitation in the singlet-singlet origin with λ_1= 335 nm, the molecule undergoes intersystem crossing with a high yield to form triplets. From level 3 (T_1), the molecules return to the original ground state if λ_2 is not present, and no hole is formed. However, in the presence of λ_2=360-405 nm, holes are formed at the singlet excitation wavelength, λ_1, presumably due to photoionization of the molecule and trapping of the ejected electron in the boric acid glass matrix. Work is in progress to fully understand this novel process[32].

Areas for future research

These two new examples of gated spectral hole-burning have opened up a new class of materials for PHB, and considering the limitations on single-photon materials, the search for gated mechanisms should be an important area for future studies. Indeed, the observations of gated PHB can only be regarded as proofs-of-principle, because the materials showing gating do not possess all the other required properties for frequency domain optical storage. Gated PHB should be observable in a variety of other systems that may offer improved properties. The search must now concentrate on finding the largest number of useful properties in one material active at diode laser wavelengths. For instance, photoionization of ions and molecules with zero-phonon lines near 8000 Å should be considered. Considering that 8-10 years of basic research was necessary on single-photon processes in order to attain the present level of understanding, gated processes deserve an equal amount of effort and attention.

Nevertheless, single photon processes with η and σ values in the upper left corner of Figure 5 would satisfy many of the requirements for frequency domain optical storage applications. Since years of experience has already been gained in single-photon materials, and considering that gated PHB materials are extremely new, a parallel effort to find optimal single-photon processes would also be useful. For any material, the issue of erasability must be considered to see if a sufficient number of erase cycles can be achieved. Furthermore, the possible existence of deleterious heating effects must be ascertained to assure that reading and writing power levels are not so high that stored holes are broadened or erased. The future feasibility of a frequency domain optical storage system rests on the discovery of single-photon materials with the required values of cross section and quantum efficiency, or upon optimizing gated mechanisms for spectral hole-burning.

Acknowledgement

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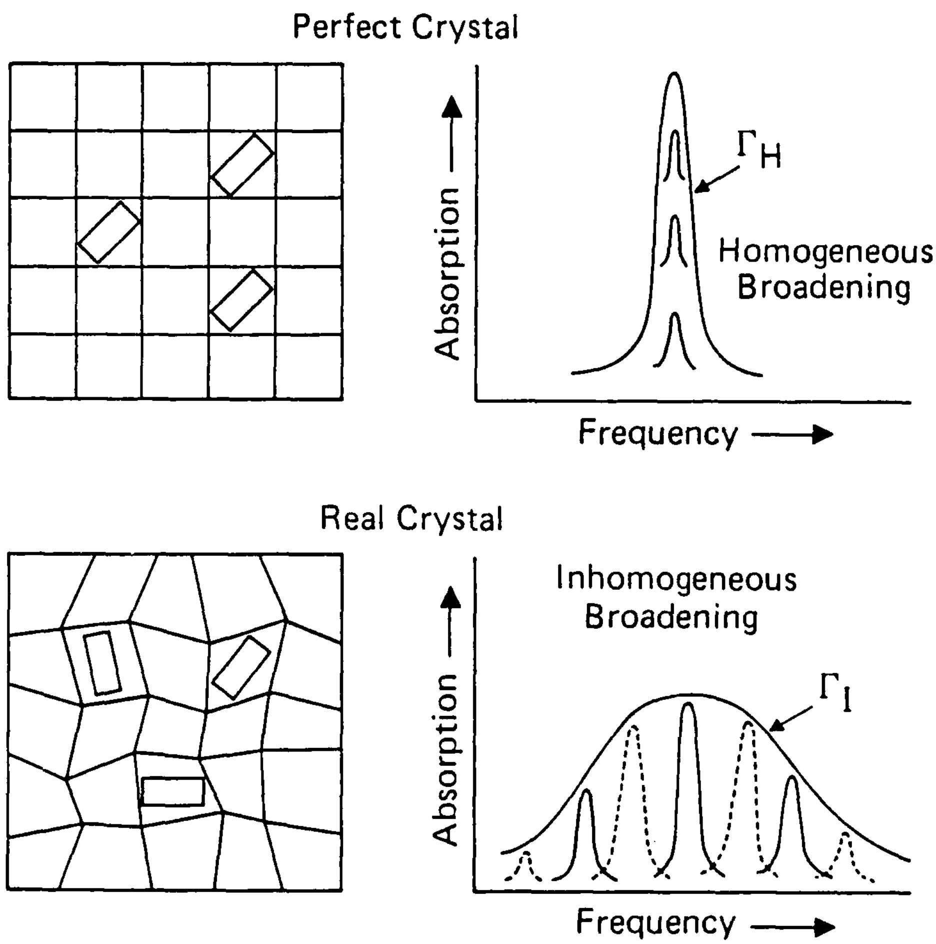

Figure 1. (Upper half) Schematic of absorbers dispersed in a perfect crystal. At low temperatures, the absorption line is homogeneously broadened with width Γ_H. (Lower half) Illustration of one source of inhomogeneous broadening in real solid matrices. The distribution of local environments leads to a distribution of center frequencies of absorption. The resulting lineshape has width Γ_I.

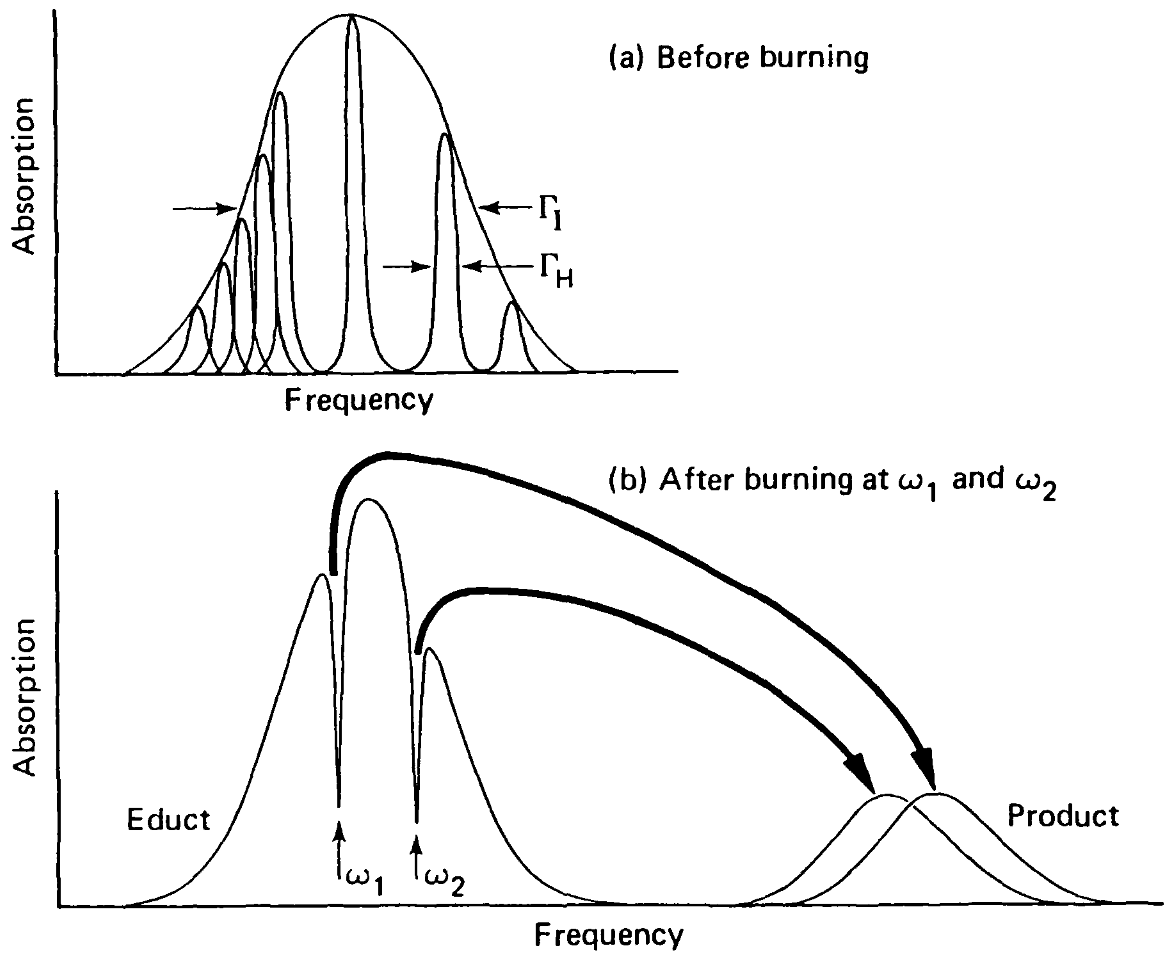

Figure 2. (a) Schematic of an inhomogeneously broadened absorption line at low temperatures. (b) Illustration of the formation of a spectral hole due to laser irradiation at ω_1 and ω_2.

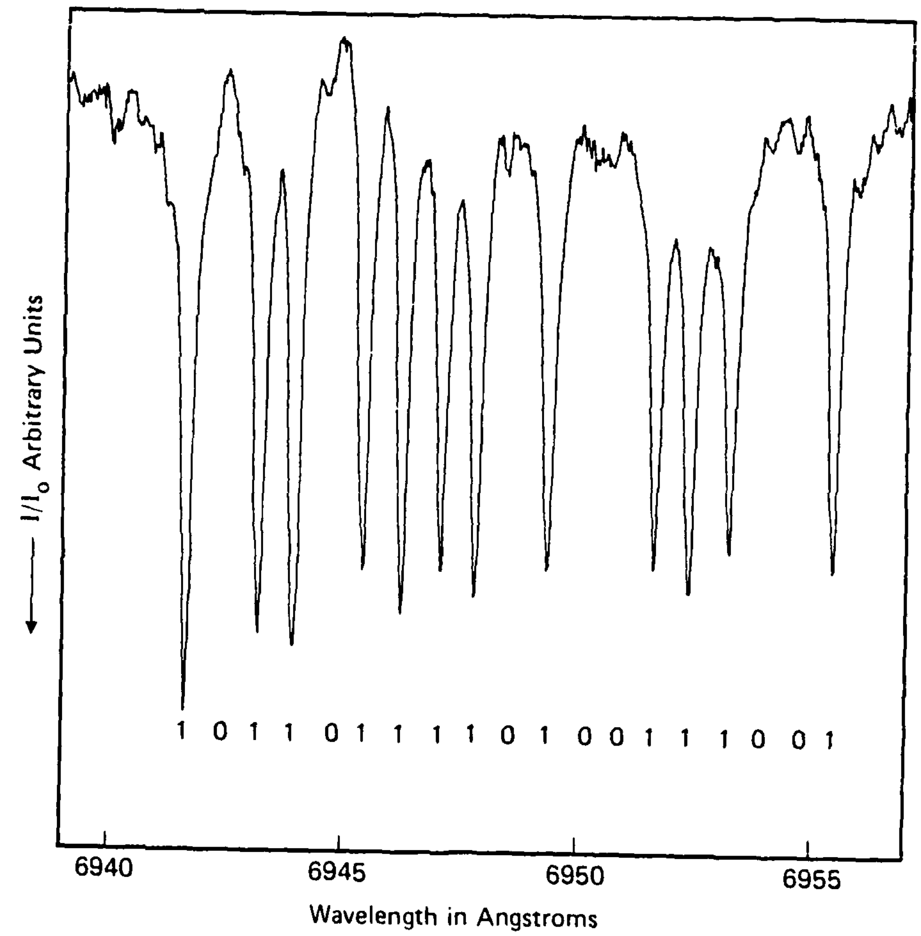

Figure 3. Portion of the absorption spectrum for free-base phthalocyanine molecules in PMMA showing a sequence of holes burned in the absorption line (after Reference [4], by permission).

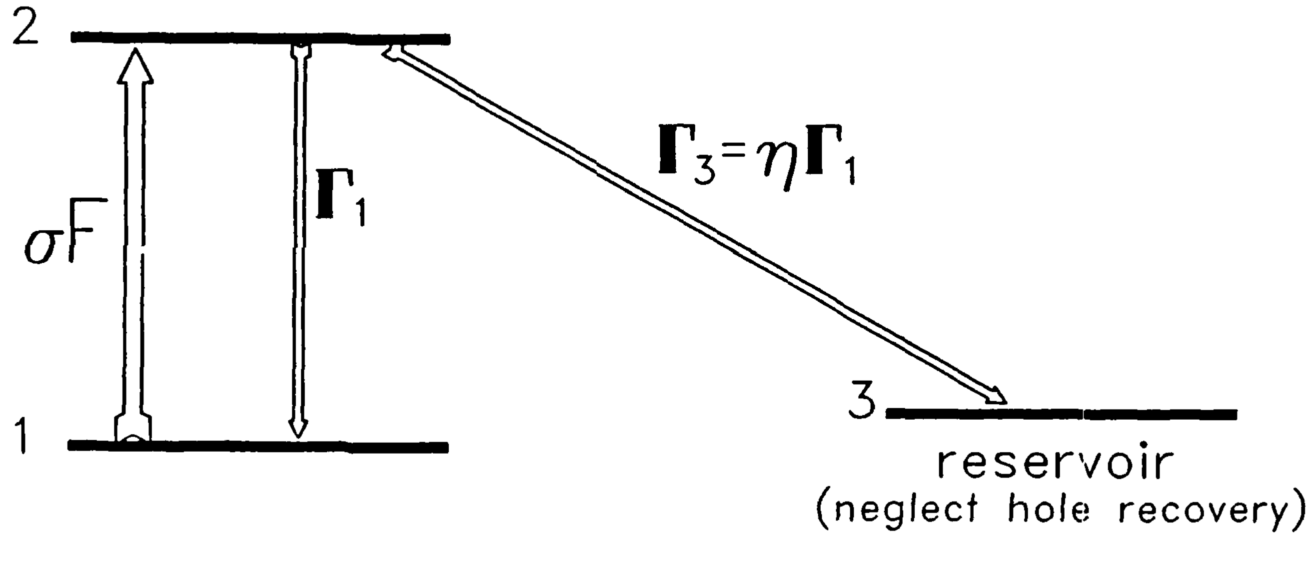

Figure 4. Schematic energy level diagram for absorbing centers with a single-photon hole-burning mechanism. Level 1 is the ground state, level 2 is the excited state, and level 3 is a permanent reservoir ground state that schematically depicts the hole formation process. The various pumping and decay rates are defined in the text.

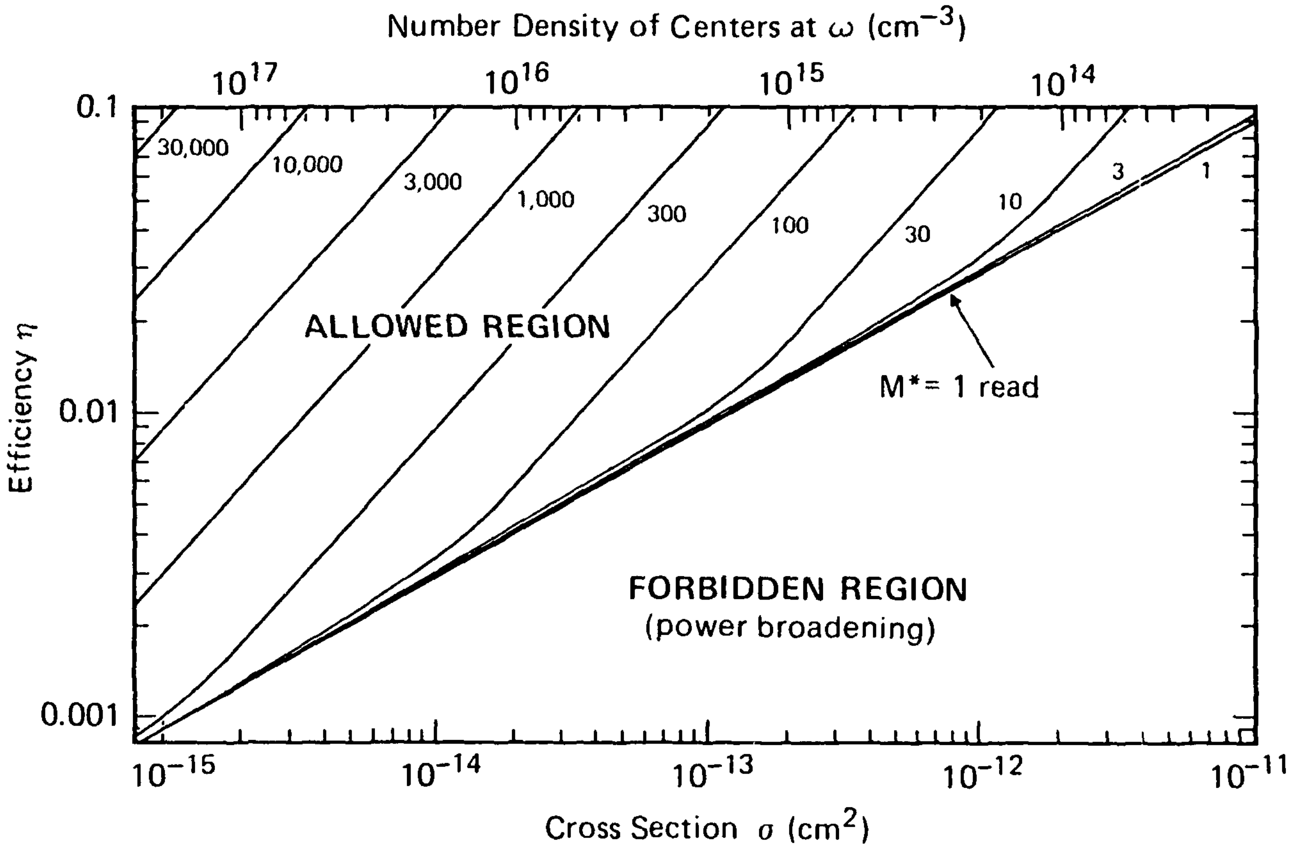

Figure 5. Allowed region of efficiency η, cross section σ, and number density of centers at ω, in order for the first read to yield acceptable signal-to-noise ratio. M^* is the number of reads achievable for a given material. The contours in the upper left portion of the figure represent contours of constant M^*.

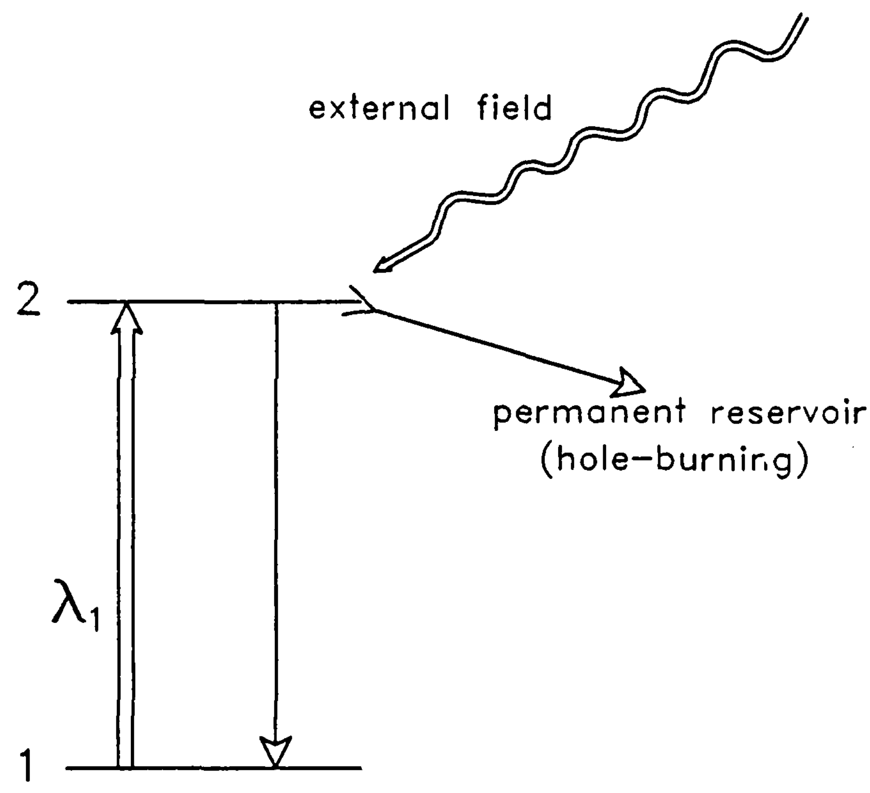

Figure 6. Illustration of basic scheme for gated, two-step PHB.

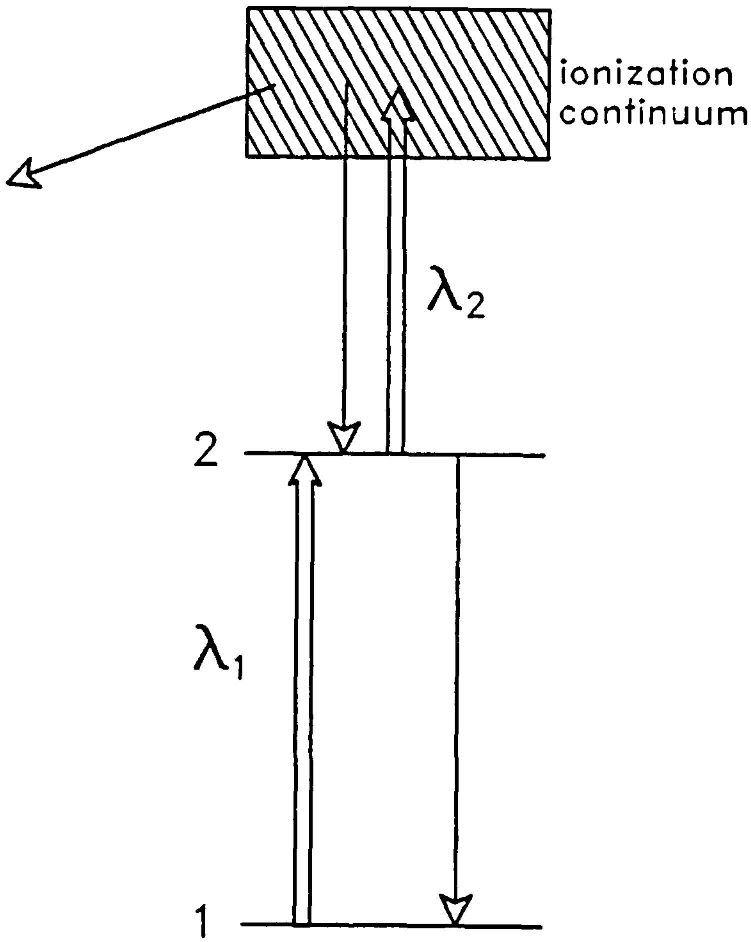

Figure 7. Illustration of gated PHB for Sm^{2+} in BaClF.

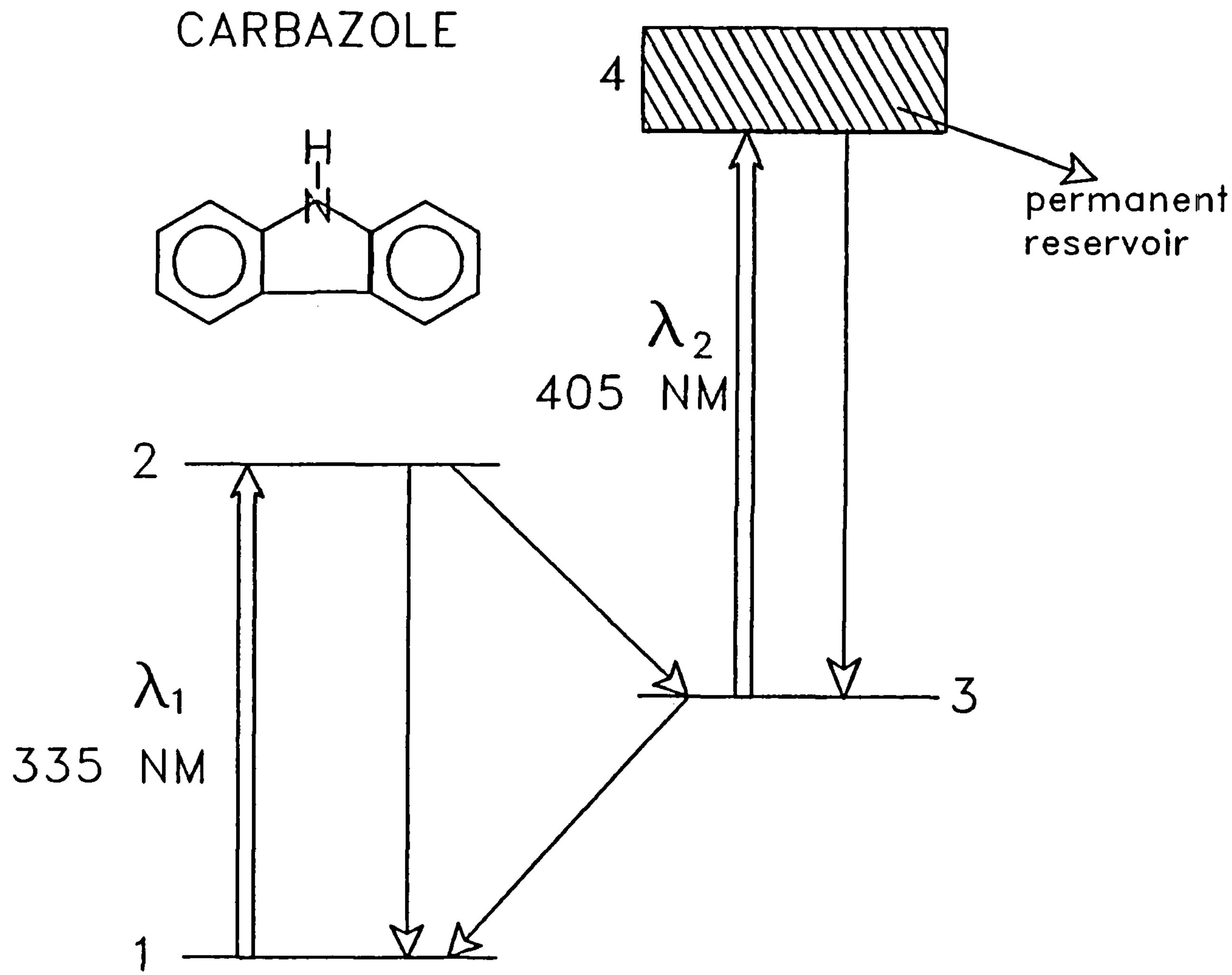

Figure 8. Illustration of gated PHB mechanism for carbazole molecules in boric acid glass. Level 1 is the singlet ground state S_0, level 2 is the singlet excited state S_1, level 3 is the lowest triplet state T_1, and the levels labeled 4 represent conduction band or autoionizing states that lead to photoionization.

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