field broadenings and shifts exceed the atomic transition widths.

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Burning out a complex-shaped hole by a coherent series of picosecond pulses

A. K. Rebane, R. K. Kaarli, and P. M. Saari

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Photochemical hole-burning in inhomogeneously broadened spectra of admixture molecules at low temperatures[1,2] is a highly promising method both for deeper investigation of the physics of admixture molecules in solids[3,4] and for solving a number of important practical problems (for example, creating optical memory elements of increased density for data storage[5,6]).

In experimental investigations of photochemical hole-burning published up to the present time a stationary regime of burning and recording the holes was assumed. The present work reports on hole-burning by a series of picosecond pulses, the shape of which reflects the phase relations between pulses nonoverlapping in time. Use of nonstationary pulsed excitation can broaden considerably the possibilities of the hole-burning method[7] to include storage and subsequent readout of the time profile of the pulses.

Let us assume that in the standard hole-burning scheme continuous emission is replaced by a train of N short laser pulses, nonoverlapping in time, and having definite phase relations between them. Neglecting the dependence on the spatial coordinates, this pulse train can be written

$$E(t) = \mathrm{Re} \sum_{k=1}^{N} E_k(t-\tau_k) \exp[i\Phi_k(t-\tau_k)],$$

where $E_k(t)$ and $\Phi_k(t)$ determine the time dependence of the amplitude and phase of pulse number k, while τ_k is its delay relative to the beginning of the time reckoning.

If in an irradiated substance sensitive to hole-burning the phase relaxation time T_2 is considerably smaller than the time dividing the separate pulses from each other, the resulting total spectral distribution of the intensity of the burning radiation $S(\omega)$ will be determined simply by the summed intensities of the separate pulses of the train

$$S(\omega) = \sum_{k=1}^{N} S_k(\omega),$$

where $S_k(\omega)$ is the spectral distribution of the intensity of pulse k. This means that in this limit ($T_2 \approx \min|\tau_k - \tau_{k+1}|$) the character of the pulsed hole-burning will be identical with burning under the action of stationary radiation having the same spectral distribution of intensity. If the phase relaxation time T_2 substantially exceeds the duration of the pulse train, the intensity distribution of the burning radiation, formally determined by the Fourier transform of the autocorrelation function of the radiation, will also depend on the phase relations between the separate pulses of the train, which leads to the appearance of additional modulation of the shape of the hole.

In the experimentally simplest case realized by us of pulse burning by two identical pulses having the delay τ between them, obtained by splitting a beam from a picosecond dye laser in a Michelson interferometer, the burning intensity may be written

$$2S(\omega)[1+\cos(\omega\tau+\Phi_0)],$$

where $S(\omega)$ is the intensity spectrum of a single pulse and Φ_0 the initial phase shift.

In the pulse-burning process examined, the shape of the hole is determined by interference between a neighboring pulse and the excited state of the substance left by the preceding pulses. Interference is possible because of the phase memory of the medium, which preserves the memory of the phases of past excitation pulses. There will be no interference if the time between the pulses exceeds the phase relaxation time T_2 of the substance.

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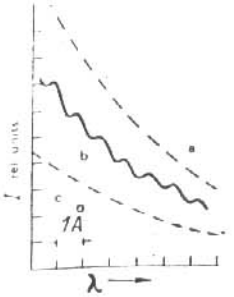

FIG. 1. Part of excitation spectrum of cryptocyanine in region of modulated hole: a—signal level before burning; b—level of maximum modulation; c—disappearance of modulation upon further burning due to saturation of hole.

The remarkable property of such burning is the possibility of fixing for an unlimited time the fine spectral structure of the separate ultrashort pulses and of a coherent series of them without disrupting their time structure, which in the case of direct measurement of a spectrum of ultrashort pulses is an exceedingly difficult task (on the problem of optimal measurement of the spectral composition of ultrashort pulses).[8]

In Fig. 1 the region of a hole in the excitation spectrum of cryptocyanine molecules in an alcohol matrix at 1.8 K is depicted, obtained by burning by the above-mentioned paired series of pulses from a picosecond dye (oxazine-1) laser, a synchronously pumped krypton laser with active Q modulation (duration of dye laser pulses, 2–3 psec; halfwidth of spectrum, 3 Å at $\lambda = 7100$ Å). The period of spectral modulation, 1.8 cm^{-1}, arising due to interference of excited states, corresponds to a spatial delay between pulses of 0.57 cm.

The maximum relative amplitude of the modulation of the measured excitation spectrum was attained at a hole depth comprising at the maximum 30% of the initial level. Upon further burning to 60% the relative amplitude of the modulation decreases, which is connected, naturally, with the finite value of the Debye-Waller factor (see, for example, Ref. 3).

The effect of a sinusoidal modulation in the excitation spectrum upon analogous burning with a 2-cm delay leads to an estimate of the upper limit of the homogeneous width of the purely electronic line $\Gamma_{00} \leq 0.1$ cm^{-1}.[4]

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Multiplex with a spherical interferometer

I. Ch. Mashek and M. P. Chaika

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The Fabry-Perot interferometer plays an essential role among high-resolution optical apparatus. But at high resolution, which is easy to achieve by increasing the thickness, its speed and free spectral range are low. The free spectral range can be increased by the use of a multiplex—two sequentially arranged Fabry–Perot interferometers.[1] On the other hand, in 1956 Connes proposed a spherical interferometer,[2] the speed of which at large thicknesses is greater than that of a plane interferometer at the same resolution.[3]

A complex interferometer–multiplex consisting of a relatively thin plane interferometer and a spherical interferometer several times thicker permits attaining high resolution with significantly greater speed than the above-mentioned multiplex.

The optical scheme of the multiplex examined is given in Fig. 1. The source can be placed either at point A (when the receiver is at point B) or at point B when the receiver is at point A. Lens L is not essential; one can omit it if the receiver or the source has an area not less than the area of diaphragm D_2, and one can place them in immediate proximity to D_2. Objective O serves to couple the two interferometers. The angle 2φ, as also the maximum size of the diaphragm $D_1 = D_2$, is determined by the resolving power of the spherical interferometer and the angle $2i$ by that of the plane. If one

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