PECULIARITIES OF HOLOGRAPHIC RECORDING IN a-$As_{40}S_{15}Se_{45}$ FILMS AT MILLISECOND EXPOSURES[1]

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Millisecond impulse hologram recording in amorphous chalcogenide films is performed for the first time and experimentally studied. The maximum diffraction efficiency in a-As-S-Se films was 0.32% which is about five times lower than for the corresponding continuous recording. However, about 500 times lower exposure is needed to reach the same diffraction efficiency in the former case. The millisecond recording is non-permanent. The dark relaxation time of millisecond holograms is 107 minutes. A photoinduced recharging of localized states in the bandgap is found to be responsible for the millisecond recording. It can be applied for non-permanent optical storage and optical information processing.

Introduction

Photoinduced effects in amorphous materials are of a special interest because of the disordered state peculiarities. For this reason and also because of the numerous applications, amorphous chalcogenide films have been actively studied since 1970s [1—3]. Real time holographic information storage is one of the most important applications. Usually the mechanism of photoinduced structural changes (PSC) [1—3] is used for holographic recording. It enables the recording of efficient and practically stable holograms. However, three other hologram recording mechanisms are known as well. They include photoinduced D-center orientation [3, 4], photostimulated relaxational structural changes [3, 4], and photoinduced recharging of localized states in the bandgap [3, 6]. D-center orientation has been used both for scalar [4] and vector [5] hologram recording in a-As_2S_3 and a-As-S-Se films, respectively. As far as we know, photostimulated relaxational structural changes have not been used for hologram recording. So far, the localized state recharging mechanism has not been directly used for hologram recording as well. Indirectly, A.V.Tyurin et al. [6] have used the recharging of D-centers at elevated temperature to initiate PSC and stable hologram recording in As-S glass.

In this paper, we report on the first (to our knowledge) room temperature hologram recording in amorphous chalcogenide films directly based on transient photoinduced recharging of localized states by the bandgap light. This phenomenon was first reported by M.Iijima and Sh.Kurita in 1980 [7]. They have found a broadband transient absorption in a-As-S films induced by pulsed 514.5 nm light but did not use this mechanism for holographic recording. We have recorded elementary holograms — holographic gratings in a-$As_{40}S_{15}Se_{45}$ films basing on this mechanism by millisecond impulses of a bandgap light. The properties of these holograms are different from those recorded by PSC. Millisecond recording can be used for non-permanent optical memory and optical information processing.

1. Experiments and their Results

Elementary holograms — transmission holographic gratings with periods Λ=1-7 μm were recorded in 4.7 μm thick fresh non-annealed amorphous $As_{40}S_{15}Se_{45}$ films (thermally evapourated onto the K-8 glass substrates) by two symmetrically incident He—Ne laser beams (λ=632.8 nm) of equal intensity. Three recording modes were used. First, unfocused beams with the $1/e^2$ diameter of 2.10 mm were used for recording. It was quickly found that millisecond impulse recording is impossible in this case because the necessary exposures are too small. Further, we used continuous wave (CW) recording by unfocused beams only for comparison purposes. Second, laser beams focused by a $F = 5$ cm lens with $1/e^2$ diameter of 35 μm were used in CW mode to record holograms whose parameters can be directly compared to those of impulse holograms. Third, the same focused laser beams with the 35μm diameter but interrupted by a millisecond photographic switch (discretely adjustable from 4 to 125 milliseconds) have been used to record single millisecond impulse

[1]This article is dedicated to Professor Marat Soskin on the occasion of his 75th birthday.

holograms. Several pulses were used for the exposure times $t > 125$ ms.

The first order diffraction efficiency (DE) η and the corresponding specific recording energy $W = I_0 t/\eta$ have been measured for all three recording modes and compared (I_0 being the average recording light intensity). I_0 varied from 0.19 to 0.39 W/cm^2 for the unfocused mode and from 1.04 to 1.62 kW/cm^2 for focused recording modes. The DE of millisecond holograms was measured just after the end of the recording pulse. (The readout time delay up to several minutes did not change the results). In the case of CW modes, we used the maximal DE $\eta_{\max}$ and the corresponding $W_{\max}$=I_0t/$\eta_{\max}$ As η and W (and $\eta_{\max}$ and $W_{\max}$) varied depending on the recording site we repeated measurements several times and took the average values. The variations of η and W due to the inhomogeneity of samples are estimated to be less than 30 and 50%, respectively.

It was experimentally found that millisecond recording in a-As-S-Se films is non-permanent. Hologram DE $1/e$ dark relaxation time was found to be 107 minutes. This enabled the recording by several millisecond pulses up to the saturation. In contrast, the CW holograms were practically stable within several years. However, the sample storage in the dark already for eight months resulted in the dramatic decrease of the millisecond recording efficiency so that it was practically impossible. The CW recording efficiency also decreases but at tolerable extent [3].

Millisecond holograms can be optically erased within several seconds by one of the 0.72 kW/cm^2 recording beams at $\Lambda \approx 2$ μm. The erasure time increases when the grating period is increased.

Average holographic parameters of the three recording modes are compared in Table. One can see that the highest DE can be reached in the focused CW mode, the lowest recording energy — in unfocused CW mode. However, the millisecond impulse recording mode gives about a 500-fold reduction of the specific recording energy compared to the focused CW mode. The highest DE in the unfocused CW mode was $\eta_{\max}$=0.62% (I_0=0.39 W/cm^2, Λ=1.4 μm, $W_{\max}$=105 J/(cm^2%)). In the focused CW mode the highest $\eta_{\max}$=1.61% (I_0=1.38 kW/cm^2, Λ=2.3 μm, $W_{\max}$=0.593 MJ/(cm^2%)) whereas, in the millisecond impulse recording mode, we have obtained the highest η=0.32% (I_0=1.26 kW/cm^2, t = 250 ms, Λ=2.0 μm, $W_{\max}$=3.9 kJ/(cm^2%)). There exists the optimum exposure $I_0 t$ = 300 J/cm^2 and the minimum exposure $I_0 t \approx 6$ J/cm^2 for millisecond recording. The DE grating period dependence of the millisecond recording is qualitatively the same as for the CW PSC based recording with the maximum at Λ=1.3 μm which is somewhat shorter than for the CW PSC case ($\Lambda \approx 2$ μm).

Discussion

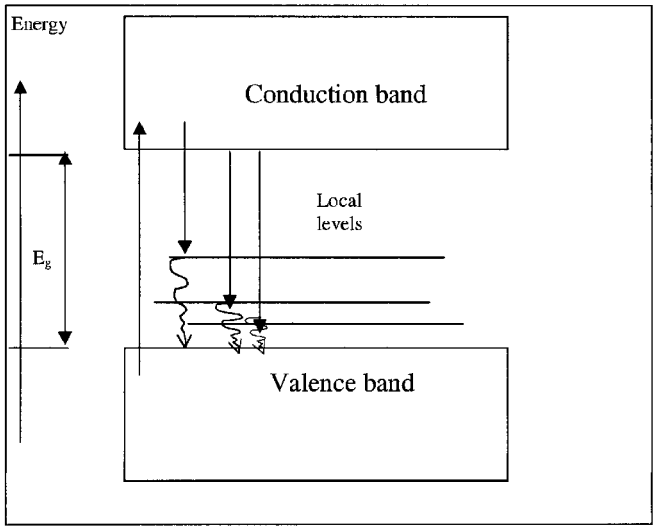
Non-permanent nature of millisecond holograms and completely different recording parameters (Table) rule out the PSC mechanism. In principle, thermal recording at $I_0 t$ ≈1 kW/cm^2 is possible. However, this is not consistent with the 107 minutes hologram dark relaxation time because the maximal heat diffusion time between the hologram maxima and minima τ_Q = $\Lambda^2/4\kappa$ (κ=5.4×10^{-7} m^2/s [8] being the coefficient of temperature conductance of the film) in our experiments did not exceed 23 μs. On the other hand, the (calculated) photoinduced temperature increase was about 70 °C which is much less than the glass transition temperature T_g=180°C [1]. Of course, thermal effects cannot be excluded but they are not dominating.

Photostimulated relaxational structural changes are too slow to occur within milliseconds [3]. D-center orientation [4, 5, 9] can be excluded because of the different hologram lifetime (2 days) and different grating period dependence (DE maximum at 10 μm).

Therefore, we conclude that a photoinduced recharging of localized defect states in the band gap of a-As-S-Se films (as proposed in [7]) is responsible for millisecond recording (Figure 1). Such localized states located at 0.6—1.0 eV above the valence band are indeed found by electrophotographic spectroscopy [10]. Bandgap light excites electrons to the conduction band. After that electrons are captured at empty localized levels thus temporarily changing the absorption and refractive indices. The electron lifetime at these local levels determines the millisecond hologram dark relaxation time of 107 minutes which corresponds to 0.97 eV activation energy at room temperature. The localized states in the bandgap are, most probably, due to the structural disorder of amorphous films (dangling bonds, etc.) [1].

Comparison of three hologram recording modes in a-As$_{40}$S$_{15}$Se$_{45}$ films at $\Lambda \approx 2\mu$m

	Unfocused CW recording	Focused CW recording	Millisecond impulse recording
I_0, kW/cm^2	0.00029	1.04	1.27
$\eta_{\max}$, %	0.48	0.74	0.115
$W_{\max}$, kJ/(cm^2%)	0.20	1470	2.715



Proposed mechanism for millisecond impulse hologram recording — photoinduced recharging of local levels in the band gap

The necessary minimum exposure of about 6 J/cm^2 for millisecond recording, most probably, is determined by the sensitivity of the used photodetector (Spectra Physics 404 Power Meter, the minimum detectable power 1 μW). The absence of PSC at such exposures during milliseconds shows the cooperative nature of the PSC effect that needs $I_0 >> 10^3$ W/cm^2 at millisecond exposure times. As known, PSC are possible even at nanosecond exposures for $I_0 \approx 10^6$ W/cm^2 [1] and at several seconds exposures at 0.3 W/cm^2 [2].

In principle, the millisecond holographic recording can be used for the non-permanent optical information storage and for optical information processing (e.g., for optically driven Van der Lugt filters). For this to be practical, it is necessary to optimize the millisecond recording in order to increase the photosensitivity by choosing optimal chemical compositions, recording and readout wavelengths, etc. This is the task for further studies.

Conclusions

Millisecond impulse hologram recording is carried out for the first time to our knowledge and experimentally studied in amorphous chalcogenide films. It is possible only with focused laser beams. The maximal DE is 0.32%. Comparison with focused and unfocused CW recording modes is made. DE in 4.7 μm thick a-As-S-Se films is about six times lower in comparison with the focused CW recording whereas specific recording energy is more than 500 times lower. The dark relaxation time of millisecond holograms is 107 minutes. The photoinduced recharging of localized states in the band gap is found to be responsible for the millisecond recording. Millisecond impulse holograms in amorphous chalcogenides can be used for non-permanent optical storage and optical information processing.

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ОСОБЛИВОСТІ ГОЛОГРАФІЧНОГО ЗАПИСУ В ПЛІВКАХ a-$As_{40}S_{15}Se_{45}$ ПРИ МІЛІСЕКУНДНІЙ ЕКСПОЗИЦІЇ

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Р е з ю м е

Вперше виконано і експериментально вивчено запис голограм мілісекундним імпульсом в аморфних галогенідних плівках. Максимальна дифракційна ефективність в плівках a-$As_{40}S_{15}Se_{45}$ становить 0,32%, що менше ніж при відповідному неперервному записі приблизно в п'ять разів. Однак необхідна для досягнення такої дифракційної ефективності експозиція в 500 разів менша. Мілісекундний запис не є перманентним. Час релаксації такої голограми в темноті становить 107 хв. Як виявилось, фотоіндукована зміна заряду локалізованих станів у забороненій зоні відповідає за мілісекундний запис. Можливе застосування останнього для тимчасового оптичного запису інформації, а також для її оптичної обробки.