Charge transfer in photorefractive CdTe:Ge at different wavelengths

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Abstract

The charge transfer processes in photorefractive CdTe:Ge were modeled using the data of optical absorption, magnetic circular dichroism (MCD) and electron paramagnetic resonance (EPR) spectroscopies. Within the developed model the variations in the photorefractive properties of different CdTe:Ge samples are explained by differences in the relative concentrations of donor and trap centers. The existence of two different centers of comparable concentrations, each in two charge states, allows charge redistribution between them and gives rise to optical sensitization of some CdTe:Ge samples for photorefractive recording under an auxiliary illumination. In the present article we follow the proposal of pseudo-3D presentation of light-induced absorption to distinguish the main charge transfer processes at different excitation energies and explain the sensitization of CdTe:Ge for photorefractive recording at 1.06, 1.32 and 1.55 μm by light with appropriate wavelength. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

Cadmium telluride is promising photorefractive material because of the largest electrooptic constant among all photorefractive semiconductors. The best CdTe:Ge samples ensure the largest photorefractive gain factor among all semiconductors without applied electric field [1]. However, the electron–hole competition does not allow to reach the theoretical limit of the gain factor. Recent photorefractive, spectroscopic, magnetooptic and electron paramagnetic resonance (EPR) studies in CdTe:Ge [2,3] showed that at least two centers each in two possible charge states contribute to the charge transfer. The variation of the photorefractive properties of different samples are explained by the difference in the relative concentrations of these donor and trap centers. On the other hand, the charge redistribution between different centers makes possible the optical sensitization of CdTe:Ge for photorefractive recording. The light with appropriate wavelength may redistribute charge carriers in such a way that the effective trap density becomes larger while the electron–hole competition becomes inhibited at the recording light wavelength. However, such

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sensitization is revealed not for all CdTe:Ge samples. In the present article we use the technique of pseudo-3D presentation of light-induced absorption proposed and developed in [4] to define the charge transfer processes at different wavelengths and to find the difference in charge transport for samples which exhibit the optical sensitization and others which do not allow it (considered typical examples are samples N12 and PM4, respectively).

2. Charge transfer processes at different excitations

The results of previous investigation of photorefractive CdTe:Ge [2,3,5] can be summarized as follows: (1) Four absorption bands (1–4) are observed in the absorption and light-induced absorption spectra with maxima around 0.94, 1.1, 1.22, and 1.35 eV, respectively. (2) Magnetic circular dichroism (MCD) spectra prove that bands 1 and 2 are associated to paramagnetic centers. (3) ODMR and photo-EPR show in agreement with previous studies that band 1 at 0.94 eV corresponds to the hole photoexcitation from the Ge^+; band 2 at 1.1 eV is associated to an unknown center $X^{0/-}$ in a paramagnetic state (X^0). (4) The pronounced photoabsorption detected in all samples indicates that the two considered centers (Ge and X) interchange charge carriers under illumination, e.g., the electron released from X^- can be trapped either by X^0 or by Ge^+. (5) EPR shows that Ge^+ is present in all samples at thermal

equilibrium (without any illumination); its amount is weaker in the N12 sample as compared to PM4. MCD provides a precise monitoring of the relative amounts of Ge^+ in different samples, as it is demonstrated in the accompanying article [5]. (6) The electron acceptor center in the N12 sample has been identified by EPR as the center previously attributed to the Te vacancy [6], the so-called F-center. This center is not present in the PM4 sample, where instead an A-center has been observed. The absolute defect concentrations of Ge^+ and A^- have been determined by EPR.

Let us analyze the absorption data measured at $T = 4.3$ K with the technique of pseudo-3D plots [4] starting from the sample PM4 (Fig. 1(a)). Analysis of the relevant photoabsorption will help us later to understand charge transfers in sample N12, the most interesting from the photorefractive point of view because, in contrast to PM4, it demonstrates a two-beam coupling gain factor close to the theoretical limit without external electric field under optical sensitization.

2.1. Low photon energy excitation (0.8–1 eV)

Obviously there is a deep around 0.94 eV (probe light) under a pump light in the same energy range. This minimum corresponds to the self-induced bleaching and indicates that a so-called primary process implies the excitation of a hole from Ge^+ to the valence band. There are two weak maxima corresponding to the same charge transfer process
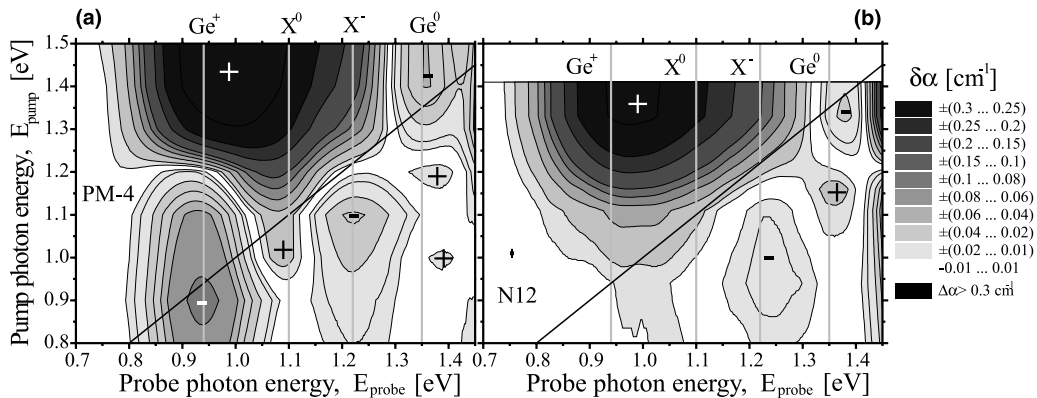

Fig. 1. Pseudo-3D plots of light-induced absorption measured in CdTe:Ge samples PM4 (a) and N12 (b).

around 1.35 eV (band 4) and 1.1 eV (band 2). The former indicates the increase of the Ge^0 concentration as a result of the decrease of Ge^+. The latter is assigned to increase of X^0 amount because part of X^- transforms to X^0 capturing the free hole liberated by Ge^+. The second minimum located at 1.22 eV corresponds to the decrease of X^- when the center captures free holes. Summarizing, the charge transfers at low energy excitation may be presented as: $Ge^+ + hv = Ge^0 + h$; $X^- + h = X^0$.

2.2. High photon energy excitation (1.25–1.5 eV)

There is a negative light-induced absorption at high energy (around 1.35 eV) close to the bisect. This absorption change corresponds to the electron excitation from Ge^0. Obviously there is a complex maximum (around 1.0 eV) associated to this primary process. It is difficult to distinguish different unresolved close maxima and perhaps additional minima. It is certain however that the absorption corresponding to Ge^+ increases. The electron excited from Ge^0 may be captured by X^0. This results in an increase of the absorption at 1.22 eV (the density of X^- enlarges) and a decrease at 1.1 eV (part of X^0 transforms into X^- capturing holes). The dominant charge transfer at high energy is most probably: $Ge^0 + hv = Ge^+ + e$; $X^0 + e = X$.

2.3. Intermediate pump energy excitation (1–1.25 eV)

The minimum of the negative light-induced absorption at 1.22 eV is shifted down from the bisect because of the band overlap with the strong broad maxima of the light-induced absorption at 1.1 and 0.94 eV. Taking this into account we can claim that minimum at 1.22 eV indicates the primary process at intermediate energy excitation. This primary process is the electron excitation from the X^-. The density of the X^0 increases due to this process, as indicated by the absorption increase at 1.1 eV. The light-induced electrons are captured by Ge^+ which in turn transform into Ge^0. The maximum at 1.35 eV indicates the increase of the Ge^0 concentration while the tail of the minimum at 0.94 eV above the bisect confirms that Ge^+ decreases. For this case the dominant charge transfer is: $X^- + hv = X^0 + e$; $Ge^+ + e = Ge^0$.

The charge transfer is shown schematically in Fig. 2 for different excitations. Arrows near the labels of the centers in different states indicate whether the corresponding concentration increases or decreases. The pseudo-3D presentation illustrates elegantly and clearly the rather spectacular photo-induced charge redistribution in the PM4 sample. Meanwhile, the usual presentation [2,3,5] is useful to analyze the details and quantitative features in the photoabsorption spectra.

The pronounced difference in photoabsorption spectra of the sample N12 (Fig. 1(b)) is the absence of the primary minimum at low energy excitation corresponding to the hole excitation from Ge^+. This can be understood from the photo-EPR results which show that only a small fraction of the Ge donor (about 4×10^{15} cm^3, as evaluated from EPR experiments) is ionized under thermal equilibrium conditions. Eventually the satellite minimum at 1.22 eV indicates that the same charge
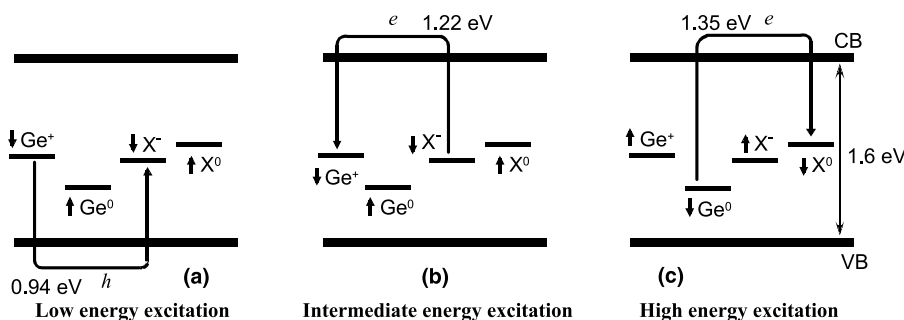

Fig. 2. Charge transfer in CdTe:Ge shown schematically for different excitation energies.

transfer processes take place in N12 sample at low energy excitation as in PM4, but with much lower efficiency. It is evident from photoabsorption data that the light with lower photon energy (1 eV) increases noticeably the Ge^+ concentration in N12 sample as compared to PM4 (1.2 eV). This is confirmed also by photo-EPR and MCD. That is why the charge transfer observed in PM4 at high energy only (see Fig. 2(c)) is already important for N12 sample at intermediate energy. The illumination of N12 sample at 1.064 μm (1.165 eV) results in concentration changes of both states of Ge and X centers opposite to that produced by 1.32 μm (0.94 eV) illumination.

Let us discuss the optical sensitization of photorefractive response using the last conclusion. Photorefractive measurements show that the grating is recorded in N12 sample at 1.55 m (0.8 eV) and 1.32 μm (0.94 eV) by carriers of a different sign as compared to the 1.064 μm (1.165 eV) recording. At 1.55 and 1.32 μm the grating is formed by the holes excited from Ge^+ and trapped by Ge^0 and X^- (see Fig. 2(a)). Light at 1.064 μm improves recording since it increases the Ge^+ concentration. For 1.064 μm recording the charge transfer shown in Fig. 2(c) becomes important for sample N12. The grating is recorded by electrons excited from Ge^0 and trapped by Ge^+ and X^0. The positive

effect of 1.32 μm illumination is in decrease of the X^- concentration which instantly leads to the increase of the X^0. The same carriers are mainly responsible for the grating recording in PM4 sample at 1.064 μm, 1.32 μm and 1.55 μm. Photoabsorption data suggest that the main carriers are holes. The optical sensitization is not observed in this case as concentrations of all species changes in the same directions within considered wavelength range. It is not excluded that the illumination with photon energy larger than 1.3 eV may improve the photorefractive response of PM4 sample.

References

[1] K. Shcherbin, A. Shumeljuk, S. Odoulov, P. Fochuk, G. Brost, SPIE Proc. 2797 (1996) 236.
[2] B. Briat, K. Shcherbin, B. Farid, F. Ramaz, Opt. Commun. 156 (1998) 337.
[3] B. Briat, F. Ramaz, B. Farid, K. Shcherbin, H.J. von Bardeleben, J. Cryst. Growth 197 (1999) 724.
[4] K. Kröse, R. Schwarfschwerdt, A. Mazur, O. Schirmer, Appl. Phys. B 67 (1998) 79.
[5] F. Ramaz, B. Farid, O. Panchuk, P. Fochuk, T. Arnoux, J.C. Launay, B. Briat, Optical Materials 18 (2001) 7 (this meeting).
[6] B. Meyer, P. Omling, E. Weigel, G. MüllerVogt, Phys. Rev. B 46 (1992) 15135.