Time-Resolved, Nonequilibrium Carrier and Coherent Acoustic Phonon Dynamics in (Cd,Mg)Te Single Crystals for Radiation Detectors

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Volume-type radiation detectors are devices that collect charged particles, such as electrons, produced by photon interaction with the detector material, typically a single crystal. In the case of highly energetic x-ray radiation photons, they interact with matter through three main mechanisms: the photoelectric effect, the Compton effect, and electron–positron pair production. The photoelectric effect is by far the most dominant effect among them because Compton-scattered photons, as well as high-energy gammas from electron–pair annihilations, typically escape from the detection volume and their energy cannot be collected. Photoconductive devices are, in fact, the most popular solid-state radiation detectors since they can often operate at room temperature, cover the spectral range up to hard x rays and even γ rays, and are easy to design and fabricate.

There is a high demand for solid-state x-ray detectors in applications ranging from medical imaging to homeland security (portable screening units) and astrophysics. Currently, cadmium zinc telluride [(Cd,Zn)Te or CZT] is the accepted material of choice; however, because of its large Zn segregation constant, it has poor crystal-growth yield, making it costly to fabricate in large volumes.[1,2] Proposed alternatives to CZT are cadmium manganese telluride [(Cd,Mn)Te or CMnT] single crystals[3,4] and most recently developed cadmium magnesium telluride [(Cd,Mg)Te or CMgT] (Ref. 5). All of the above materials are ternary alloys that contain tellurium, which ensures their very high stopping power and 100% absorption efficiency for x-ray photons with energies up to above 100 keV (Ref. 4). However, CMgT also possesses all other necessary qualities for an optimal radiation detector, i.e., high density (5.83 g/cm^3), high electron effective mass (49.5), ultrahigh resistivity (~10^{10} Ω-cm), and a good electron mobility lifetime ($\mu\tau_e$) product (>10^{-4} cm^2/V) (Ref. 5). In addition, the CMgT "parent" crystals, CdTe and MgTe, exhibit very similar lattice constants, namely, 6.48 Å and 6.42 Å, respectively,[6] resulting in a high crystallinity yield of the CMgT material.

This summary focuses on the ultrafast optical properties of the latest member of the above-mentioned ternary materials, namely, the CMgT single crystal. We present comprehensive femtosecond pump–probe spectroscopy studies where we measure time-resolved carrier dynamics and, subsequently, analyze the data within a coupled rate-equation model, developed to reveal both the carrier recombination and trapping components of the relaxation process. In addition, we time-resolve long-lived coherent acoustic phonons (CAP's) in a manner similar to a method earlier implemented for CMnT crystals.[7]

In our experiments, we test CMgT crystals with optimal composition and ultrahigh resistivity for x-ray detection applications,[8] namely $Cd_{0.92}Mg_{0.08}Te$, and with two different dopants: indium (CMgT:In) and germanium (CMgT:Ge). In both cases, doping ensures that the resulting crystals exhibit ultrahigh resistivity; however, the used dopants act very differently. Indium doping is intended to simply compensate the native concentration of holes (CdTe-based crystals are naturally *p*-type semiconductors), while

Ge is a deep impurity, introduced to "pin down" the Fermi level at the middle of the band gap. The impurities obviously affect the crystalline quality of the resulting materials, negatively affecting the $\mu\tau_e$ product that needs to be as large as possible for a sensitive detector. Therefore, typically, the single crystals intended for x-ray detectors are annealed after their growth in order to improve their crystalline structure. In our work, we compare time-resolved dynamics of photoexcited carriers in CMgT:In and CMgT:Ge, both as-grown and annealed crystals, in order to determine what material exhibits the best transport properties, i.e., minimal trap concentration and the longest electron lifetime, required for the optimal photoelectric radiation detector.

To understand the physics of the nonequilibrium relaxation dynamics of photoexcited carriers in our CMgT crystals (see Fig. 1 as an example), we fitted our experimental probe-normalized, transient-reflectivity $\Delta R/R$ waveforms, measured in both one- and two-color setups, to the trapping and relaxation model that we developed and presented in Ref. 9. We note that in Fig. 1, as well as all other cases, the numerical total concentration of photoexcited carriers' dependence (solid red line) fit perfectly with the experimental $\Delta R/R$ transient (black circles). The initial fast relaxation time τ_1 is ascribed as the direct trapping of excited electrons, while the subsequent relatively much slower relaxation time τ_2 can be interpreted as the Shockley–Read–Hall recombination. The values of the fitting parameters, in the case of Fig. 1, are $\tau_1 = 0.21$ ps and $\tau_2 = 4.77$ ps. The subpicosecond value of τ_1 indicates that, in this sample, carrier trapping is the dominant mechanism of the photoresponse. The latter also explains the negative dip at the relaxation part of the $\Delta R/R$ signal. The value of τ_2 is also very short, showing that traps are also very effective in nonradiative electron-hole recombination. Therefore, our approach provides a detailed description of the physical processes governing both the carrier excitation and subsequent relaxation dynamics in our as-grown and annealed CMgT:In and CMgT:Ge samples.

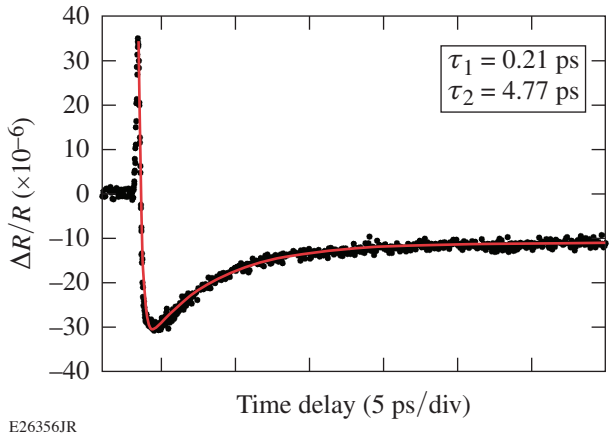

Figure 1
Normalized reflectivity transient change $\Delta R/R$ (black circles) measured in a one-color, pump–probe spectroscopy setup for an as-grown CMgT:In sample. The solid red line is the best fit to the trapping and relaxation model and the corresponding fitting parameters are listed in the inset.

During the course of our two-color pump–probe studies performed on both CMgT:In and CMgT:Ge samples, we consistently observed the presence of weak but very regular oscillations on the relaxation part of $\Delta R/R$ waveforms when traced in a time-delay range of the order of a nanosecond. These oscillations can be satisfactorily interpreted within the propagating strain-pulse model, introduced by Thomsen *et al.*[10] and Wu *et al.*[11] The high-energy and high-fluence femtosecond pump pulses incident on the crystal surface introduce electronic as well as thermal stress and generate a strain transient (lattice discontinuity) that propagates with a velocity of sound v_s into the sample at the direction orthogonal to the surface, locally altering its optical properties; namely, the refractive index of the crystal. The time-delayed probe beam penetrating the crystal is partially reflected from this traveling discontinuity and interferes with the part of the probe light reflected from the crystal surface, resulting in the regular oscillations observed on top of the $\Delta R/R$ photoresponse signal, interpreted as CAP's. CAP propagation was dispersionless with the constant propagation velocity corresponding to the speed of sound in our CMgT, (111) oriented crystal. The intrinsic lifetime of CAP's was estimated to be as long as 10 ns or more.

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