Charge storage and persistent photoconductivity in a $CdS_{0.5}Se_{0.5}$ semiconductor alloy

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Relaxation of stored charge carriers in the persistent-photoconductivity (PPC) mode in a $CdS_{0.5}Se_{0.5}$ semiconductor alloy has been investigated. The relaxation time constant of PPC is systematically measured as a function of temperature, from which the recombination barrier height E_{rec} has been determined. Low-temperature exciton luminescence has also been investigated and the exciton transition linewidth, which is broadened due to the presence of compositional fluctuations, is measured. The values of E_{rec} deduced from the measured exciton linewidth and the localized-to-delocalized transition temperature in the PPC mode are consistent with the results obtained from the PPC-decay measurements. These experimental results are consistent with our previous interpretation that PPC in $CdS_{0.5}Se_{0.5}$ semiconductor alloys is caused by random local potential fluctuations induced by compositional fluctuations.

I. INTRODUCTION

Persistent photoconductivity (PPC), photoinduced conductivity that persists for a very long period of time after the termination of the photoexcitation, has been studied for more than two decades. It is one of the most interesting yet not well understood phenomena in semiconductors. PPC has been observed at low temperatures in bulk semiconductors and in various other configurations.[1-4] Recently, a relaxation time constant as long as 10^{13} s at 10 K has been observed in $Al_{0.3}Ga_{0.7}As$.[5] In contrast, a decay-time constant on the order of 10^3 s has been found in II-VI semiconductor alloys. Nevertheless, PPC exists up to room temperature and can be optically quenched by infrared radiation in II-VI semiconductor alloys, which makes them very important materials for device applications, such as infrared detectors.[6]

Several mechanisms have been proposed to explain the origin of PPC. PPC could be caused by interface barriers in heterojunction materials.[7,8] In $Al_xGa_{1-x}As$ semiconductors, the DX center is believed to be the origin of PPC.[9,10] In this model description, PPC results because recapture of electrons by the DX centers is prevented by a thermal barrier at low temperatures. The nature of the DX center is under intensive investigation,[11-14] with key questions focusing on whether it is a deep center with large lattice relaxation (LLR) (Refs. 13 and 15) or simply an effective-mass state impurity of the L conduction band with small lattice relaxation (SLR).[14,16-18] A description of the DX center as being D^- with negative-U character has been proposed,[19,20] and is supported by many experiments. Although the PPC effect has been extensively studied in $Al_xGa_{1-x}As$, the understanding of such an effect in II-VI materials is still at a very early stage.

Recently, we have observed that the stored charge carriers in PPC mode in a $Zn_{0.3}Cd_{0.7}Se$ semiconductor alloy experience a phase transition from a hopping-conduction to a percolation-transport state at the critical temperature T_c and their relaxation behavior was found to follow the stretched-exponential function.[21-23] There are three pieces of experimental evidence for this transition: (a) PPC buildup level as a function of temperature,[22] (b) PPC decay time constant and decay exponent as functions of temperature,[22] and (c) PPC noise level as a function of temperature.[21] All of these depict a transition of percolation type near T_c. From these experimental results, we proposed that random local potential fluctuations (RLPF) induced by compositional fluctuations may be responsible for PPC in II-VI semiconductor alloys. In fact, it has been proposed previously that inhomogeneities caused by impurity doping in impurity-doped and compensated semiconductors are the most likely reason for PPC in II-VI materials.[1] It is known that in II-VI semiconductor alloys, fluctuations of local composition produce concomitant fluctuations in the band edges, which cause energy-band tails in these materials.[24,25] Below a low-temperature limit T_L ($T_L = 70$ K at a bias of 1.5 V for $Zn_{0.3}Cd_{0.7}Se$, for example), the electrons are highly localized and the transport becomes negligible; as temperature increases, PPC is induced by electron hopping between localized states; as temperature increases to above T_c, some localized electrons become delocalized (activated to levels above the mobility edge) and the charge transport occurs via the electrons percolating through the conduction network, while holes remain localized because of their heavier mass. Electrons are localized in the sites or percolate through the network of the potential minima and are spatially separated from the localized holes because the potential minima for electrons are the potential maxima for holes. This leads photoexcited charge carriers to have long recombination lifetimes and leads to the effect of PPC. Further increase in tempera-

ture would give electrons sufficient thermal energy to transfer to the maximum barrier height, E_{rec}, to recombine with holes soon after photoexcitation and cause a thermally activated PPC decay behavior. As a consequence, in addition to the transition temperature T_c, there exists an upper-temperature limit T_U and a low-temperature limit T_L for the observation of PPC in II-VI semiconductor alloys.

In this paper, we study the PPC decay process in a $CdS_{0.5}Se_{0.5}$ semiconductor alloy. The decay-time constants are measured systematically in the entire PPC temperature region, and from these we obtained the recombination barrier height, E_{rec}, which represents the root mean square of the energy differences between the potential minima and maxima of the fluctuating potentials in the conduction-band edge. Low-temperature exciton luminescence is also investigated for $CdS_{0.5}Se_{0.5}$. The exciton transition linewidth predominantly caused by compositional fluctuations in the sample is measured and used to deduce the energy fluctuation parameter for electrons of the excitons. The result is compared with that obtained from PPC data. Our experimental results are consistent with our previous interpretation that PPC in $CdS_{0.5}Se_{0.5}$ is caused by RLPF induced by compositional fluctuations.

II. EXPERIMENT

The sample used for this study was a high-purity and nominally undoped $CdS_{0.5}Se_{0.5}$ semiconductor alloy supplied by Cleveland Crystals, Inc. of size about $5 \times 10 \times 1$ mm^3 with dark-room temperature resistivity of about 10^9 Ω cm. Gold leads were attached to the sample using indium solder, and the junctions of 1 mm in diameter about 5 mm apart on the sample surface were carefully tested for Ohmic character. The c axis is perpendicular to the sample surface. The sample was attached to a copper sample holder, which is inside a closed-cycle He refrigerator, with care taken to ensure good thermal contact yet electrical isolation. A mercury lamp was used along with appropriate filters, so two lines at 435.8 and 546.1 nm dominated the output of the excitation source. The data obtained at different conditions were taken in such a way that the system was always allowed to warm up to 300 K and relax to equilibrium after each measurement, then cool down in darkness to the desired temperature of measurements. This is to ensure that each set of data has the same initial condition. Measurements of PPC buildup and decay at different photon-dose conditions are taken by illuminating the sample for different time intervals while keeping the same intensity at different temperatures. The PPC buildup levels depend on excitation photon dose. The typical excitation photon flux used for the measurements is on the order of 10^{13} photons/cm^2 s. The current is measured by a Keithley digital electrometer (model 617), which is interfaced with a computer. A 1.5-V bias is supplied by a battery.

Photoluminescence data were collected in a reflecting mode. Excitation pulses of about 7 ps in duration at a repetition rate of 1 MHz were provided by a cavity-dumped ultrafast dye laser (Coherent 702-2CD), which was

pumped by a yttrium-aluminum-garnet (YAG) laser (Quantronix 416) with a frequency doubler. The lasing photon energy was 2.125 eV with spectral width of about 2 meV corresponding to a band-to-band excitation. The average power density is about 100 mW/cm^2. The detection system consists of a time-correlated single-photon counting system and a double monochromator with a spectral resolution of 1 meV.

III. RESULTS AND DISCUSSIONS

PPC decay curves for $CdS_{0.5}Se_{0.5}$ at two representative temperatures, $T = 8.6$ and 40 K, are shown in Fig. 1. The PPC curves are normalized to unity at $t = 0$, the moment of excitation light being terminated. The dark current has been subtracted. These two curves show that PPC decays faster at $T = 40$ than at $T = 8.6$ K. At $T = 8.6$ K, the PPC level is about 6% of its initial level after 1000 s decay. In this sample, PPC is observable down to 8 K, and T_L at a bias of 1.5 V has not yet been determined because our low-temperature system can achieve a lowest temperature of about 8 K. Just as in $Zn_{0.3}Cd_{0.7}Se$, the decay of PPC in $CdS_{0.5}Se_{0.5}$ can be approximated by a stretched-exponential function, a behavior frequently observed in disordered systems,

$$I_{PPC}(t) = I_{PPC}(0) \exp[-(t/\tau)^\beta] , \qquad (1)$$

where τ is the decay-time constant and β is the decay exponent. This has been demonstrated by the plot of $\ln[\ln I_{PPC}(0) - \ln I_{PPC}(t)]$ as a function of $\ln(t)$ for PPC decay curves obtained at these two temperatures. An approximately linear behavior can be observed in Fig. 2. From these curves we can obtain τ and β. The stretched-exponential-decay approximation shown in Fig. 2 for PPC relaxation in $CdS_{0.5}Se_{0.5}$ is not as good as in
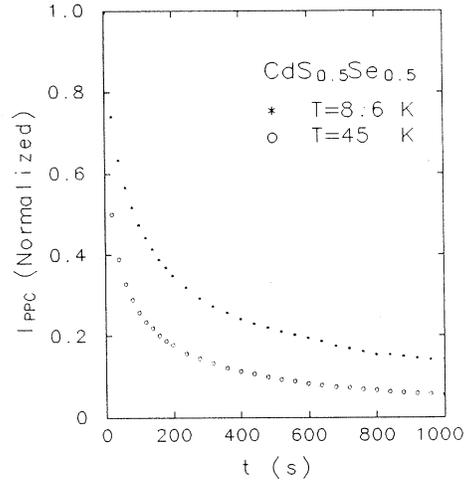
FIG. 1. Normalized PPC decay curve of $CdS_{0.5}Se_{0.5}$ at two representative temperatures, $T = 8.6$ and 40 K, after 500 s of light illumination. The dark current has been subtracted out. The excitation light flux used is on the order of 10^{13} photons/cm^2 s.
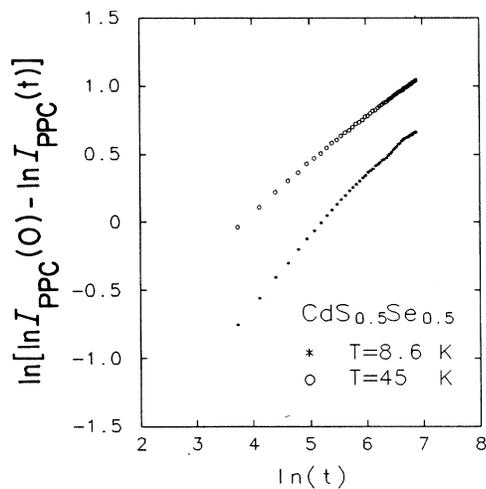
FIG. 2. Plot of $\ln[\ln I_{PPC}(0)-\ln I_{PPC}(t)]$ as a function of $\ln(t)$ for two selective temperatures, $T=8.6$ K. Approximately linear behavior indicates that the PPC decays according to the stretched-exponential function, $I_{PPC}(t)=I_{PPC}(0)\exp[-(t/\tau)^{\beta}]$.

$Zn_{0.3}Cd_{0.7}Se$.[22] This is believed to be due to the fact that the degree of the fluctuations in $CdS_{0.5}Se_{0.5}$ is not as large as that in $Zn_{0.3}Cd_{0.7}Se$, which is consistent with the experimental observation that the transition temperature T_c is 120 K for $Zn_{0.3}Cd_{0.7}Se$ and 15 K for $CdS_{0.5}Se_{0.5}$.[21] In any case, such an approximation enables us to measure the effective relaxation time constant τ systematically as a function of temperature, which can provide important information.

Figure 3 is the plot of the decay-time constant τ as a function of temperature for $CdS_{0.5}Se_{0.5}$. We can see that PPC decays slower near 15 K than at temperatures lower than 15 K, which is very similar to the behavior exhibited

FIG. 4. Semilogarithmic plot of the decay-time constant as a function of $1/T$ for $CdS_{0.5}Se_{0.5}$.

by $Zn_{0.3}Cd_{0.7}Se$. The Arrhenius plot $\ln\tau$ vs $1/T$ in Fig. 4 shows two distinctive temperature regions. At high temperatures ($T>20$ K), PPC decay is caused by thermally activating electrons into the maximum-potential regions to recombine with holes; therefore, τ is thermally activated and a linear behavior in this temperature region ($T>20$ K) is evident. At low temperatures, τ is weakly dependent on temperature, and PPC decay may be caused by wave-function overlap between electrons and holes in real space.

Dependence of the PPC relaxation behavior on excitation photon dose is also investigated for $CdS_{0.5}Se_{0.5}$. Figure 5 shows the decay curves obtained at 8.4 K after excitation with three different light-illumination times. Similar to the behavior exhibited by $Zn_{0.3}Cd_{0.7}Se$, at a con-

FIG. 3. Plot of the PPC decay-time constant τ as a function of temperature for $CdS_{0.5}Se_{0.5}$. Experimental conditions are the same as those in Fig. 1.
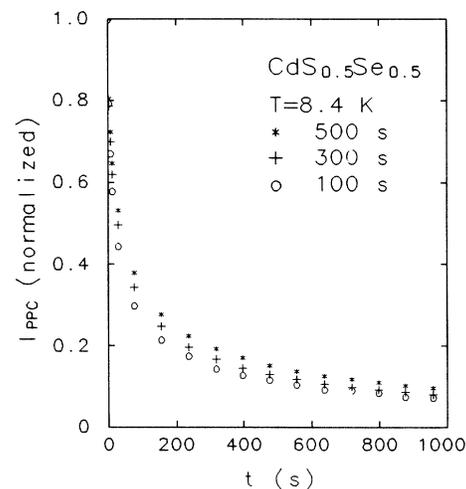
FIG. 5. I_{PPC} (normalized) vs decay time t for different illumination times.

stant temperature and prior to the PPC saturation, the relaxation of PPC proceeds faster with a decrease of excitation photon dose as depicted in Fig. 5. This behavior can also be explained qualitatively in terms of our model description. As the electron quasi-Fermi-level (or carrier concentration) increases, PPC decays slower for carriers in the delocalized state (above mobility edge) than in the hopping transport state due to the electron-redistribution effect discussed previously,[22] thus τ increases with an increase of the quasi-Fermi-level or excitation photon dose. In the very-low-excitation photon-dose region, different behavior is observed. Further experimental and theoretical investigations are required to explain the temperature and excitation photon-dose-dependent PPC relaxation behavior for carriers in percolation and hopping transport regimes quantitatively.

At the thermally activated region, the decay-time constant τ can be expressed as

$$\tau = \tau_0 \exp(E_{\rm rec}/kT) , \qquad (2)$$

where $E_{\rm rec}$ is the recombination barrier height and k is the Boltzmann constant. The potential barrier $E_{\rm rec}$ obtained from the high-temperature region shown in Fig. 4 is 6.6 meV. This value indicates a small degree of potential fluctuations of the $CdS_{0.5}Se_{0.5}$ sample. This is direct experimental evidence that a small recombination barrier can lead to the PPC effect.

We believe that the recombination barrier height in different samples, $E_{\rm rec}$, is uniquely determined by the compositional fluctuations in the samples. On the other hand, the transition temperature T_c observed in the PPC mode as well as T_U and T_L for different samples are also correlated to the compositional fluctuations in the sample. In order to establish their relation, we assume that for II-VI semiconductor alloys $A_x B_{1-x} C$, the conduction-band edge varies with the local values of x and acts as a potential energy for electrons.[24] Since the growth of the sample is a random process, the composition of lattice sites containing the type-A atoms should fluctuate around the "nominal" composition value x_0. For the sake of simplicity, we suppose that there is no correlation in the atom distribution in the lattice; the probability of obtaining a composition x may be assumed to follow a Gaussian distribution, which can be written as

$$P(x) = P_0 \exp[-(x-x_0)^2/2\sigma_x^2] , \qquad (3)$$

where P_0 is determined by normalization condition $\int P(x)dx = 1$ and σ_x is the compositional-fluctuation parameter. For semiconductor alloys with different compositions and growth conditions, the values of σ_x are different. The energy at the conduction-band edge, $E_c(x)$, is a function of the local composition x. For small fluctuations Δx, this function may be written as[24]

$$E_c(x) = E_c(x_0) + \alpha_c \Delta x , \qquad (4)$$

where α_c is the rate of variation of the conduction-band edge with composition x, $\alpha_c = (dE_c/dx)_{\Delta x=0}$. We further assume that after photoexcitation the low-potential lattice sites are filled by electrons and that the transition

occurs when the ratio of the filled low-potential sites to the total available sites approaches a critical concentration value, P_c, and the average energy of an electron equals the energy at the percolation threshold (or mobility edge). After these considerations, a relation between T_c and σ_x has been obtained.[23] For $CdS_{0.5}Se_{0.5}$, the observed T_c is 15 K,[21] from which we deduced a value for σ_x of about 0.0035. the fluctuations in the local composition may be assumed to be on the order of $\sqrt{2}\sigma_x$. In reality, the structure of the random fluctuating potentials of the band edges caused by the fluctuations of local composition may be very complicated and is unknown at this stage. However, the root mean square of the fluctuating potentials, which represents the average recombination barrier height, $E_{\rm rec}$, may be approximated by

$$E_{\rm rec} \approx E_c(x_0 + \sqrt{2}\sigma_x) - E_c(x_0 - \sqrt{2}\sigma_x)$$
$$\approx 2\sqrt{2}\alpha_c\sigma_x . \qquad (5)$$

We also assume that $\alpha_c = a\alpha$ with $\alpha = (dE_g/dx)_{\Delta x=0}$, the rate of variation of the band gap with composition x and that $E_g(x)$ varies linearly with x,[26] where $\alpha = 740$ meV for $CdS_x Se_{1-x}$.[27] Here, a indicates the fraction of α contributing to the conduction band. This does not imply that the band gap fluctuates with the local values of composition x in the sample. In fact, the model assumes that the band edges vary with local composition; nonetheless the band gap remains approximately constant through the space. The estimated values of σ_x and α_c give $E_{\rm rec}$ to be about 5.9 meV with assumption of $a = 0.8$, which is in reasonable agreement with the result obtained from the plot of $\ln\tau$ vs $1/T$ shown in Fig. 4. Notice that the actual value of a has not yet been determined for this material. Furthermore, the recombination barrier height $E_{\rm rec}$ measured from the PPC decay-time constant also depends on the stored charge carrier concentration because it is measured from the quasi-Fermi-levels. These experimental results suggest that PPC in $CdS_{0.5}Se_{0.5}$ is caused by conduction of charge carriers stored in random local potential wells induced by compositional fluctuations.

It is also well known that the properties of photogenerated electron-hole pairs (or excitons) are strongly influenced by compositional fluctuations.[28] In semiconductor alloys, excitons exhibit an important feature, i.e., the exciton localization in the random fluctuating potentials induced by compositional fluctuations. Recently, by studying the time-resolved photoluminescence of exciton transition in the same sample,[29] we found the Mott-Anderson type of localization just below the mobility edge for excitons from their lifetime measurements, which is consistent with the characteristics exhibited in the PPC mode. Figure 6 shows the low-temperature (8.4 K) photoluminescence of the localized-exciton transition line in $CdS_{0.5}Se_{0.5}$. Two peaks at 2.075 and 2.041 eV are ascribed, respectively, to the radiative recombination of a localized exciton and its one-phonon replica. The measured full width at half maximum (FWHM) of the localized exciton line is about 11.3 meV compared with about 0.2 meV in a good-quality binary crystal of CdS or CdSe. These results clearly indicate the existence of composi-
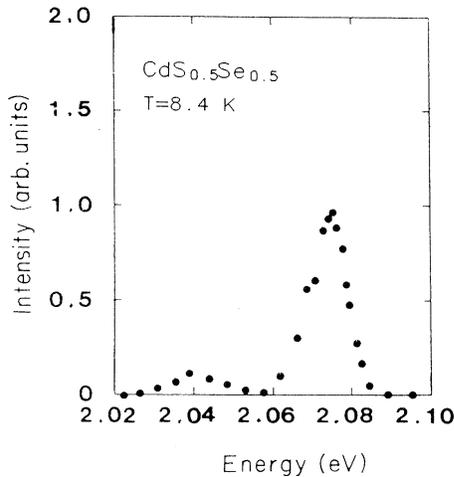
FIG. 6. The low-temperature ($T=8.4$ K) exciton photo-luminescence of $CdS_{0.5}Se_{0.5}$. The excitation energy was 2.125 eV with an average power density of about 100 mW/cm^2.

tional fluctuations in this sample. The fluctuations of local composition cause fluctuations in the band edges leading to the broadening of the exciton luminescence band. Because the linewidth of the exciton transition is caused by energy fluctuations in both the conduction and the valence bands, we can assume that the energy fluctuations for the electrons of the excitons in the conduction band is about half of 11.3 meV. Therefore, the exciton transition linewidth measurement gives a value for energy fluctuations in the conduction band to be 5.7 meV, which is a bit smaller than the value of E_{rec} obtained from the PPC measurements. This is what we expected. For localized excitons, their electrons cannot distribute in those sites with energies as high as the potential maximum (see Fig. 3 of Ref. 6). Thus half of the FWHM of the localized exciton transition line should be smaller than E_{rec}. These results further explore the alloy-disorder effect and again support our interpretation for PPC in II-VI semiconductor alloys.

The behavior of the decay-time constant as a function of temperature shown in Fig. 4 resembles that of a similar plot for $Al_{0.3}Ga_{0.7}As$.[5] The decay of PPC in $Al_xGa_{1-x}As$ was also found to follow stretched-exponential functions.[5,30] Recently, it has been proposed that self-compensation of As and P impurities in ZnSe results primarily from a large lattice relaxation, which leads to formation of a positively charged A^+ defect center and consequently the effect of PPC.[31] However, as the sample studied here is high-purity and nominally undoped $CdS_{0.5}Se_{0.5}$, the effect of the foreign impurities is not involved here. Nonetheless, in undoped II-VI semi-

conductors, an intrinsic lattice defect may act as a donor or acceptor, similar to the case of a foreign impurity atom, and the problem of contamination may also have to be considered. One might ask whether these defects which may have a thermal-capture barrier due to lattice relaxation could cause the PPC effect observed here. Experimentally, we may not have enough evidence for a conclusive answer to this, but some of the different PPC behaviors resulting from the DX centers and random local potential fluctuations have been distinguished previously.[21] These early results obtained for $Zn_{0.3}Cd_{0.7}Se$ and $CdS_{0.5}Se_{0.5}$ show that there is a low-temperature limit for the observation of PPC and a phase transition from localized to delocalized states for charge carriers in the PPC mode, which are not expected from the DX center model. Furthermore, experimental results reported here are fully consistent with our previous interpretation for PPC in nominally undoped II-VI semiconductor alloys. In particular, it is impossible to account for 6-meV capture barrier in $CdS_{0.5}Se_{0.5}$ by DX centers. In contrast, the capture barrier obtained for $Al_xGa_{1-x}As$ has the smallest value on the order of 200 meV at $x=0.3$.[32] Additionally, the PPC buildup behavior in II-VI and III-V is also very different.[33] Finally, PPC in II-VI semiconductor alloys can be quenched by far-infrared radiation,[6] while in $Al_xGa_{1-x}As$ it cannot be optically quenched as proved recently.[34] At this stage, based on these experimental results, we believe therefore that a different mechanism from the DX center is involved in nominally undoped II-VI semiconductor alloys; namely, the random local potential fluctuations induced by compositional fluctuations may be responsible for PPC observed in these materials.

IV. CONCLUSIONS

Charge-storage and PPC phenomena in a $CdS_{0.5}Se_{0.5}$ semiconductor alloy have been studied. The observed behavior of the PPC decay-time constant as a function of temperature shows that the decay of the stored charge carriers is caused predominantly by thermal activation capture at temperatures $T > 20$ K. The recombination barrier height obtained from the PPC-decay-time-constant measurements is consistent with the results obtained from the PPC transition temperature and the exciton transition linewidth measurements. These experimental results again suggest that the random local potential fluctuations (RLPF) induced by the compositional fluctuations are responsible for PPC in $CdS_{0.5}Se_{0.5}$.

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