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DYNAMICS AND SPATIAL DISTRIBUTION OF EDGE LUMINESCENCE GENERATORS IN CDS FROM TIME-RESOLVED EXCITATION SPECTROSCOPY

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The decay rate of D°-A° recombination radiation is shown to increase for pairs in regions of strong surface electric field. Time-resolved spectral features appear at impurity-bound exciton resonances.

INTRODUCTION

Although donor-acceptor (D-A) recombination emission in semiconductors is reasonably well understood[1], the processes contributing to the photoproduction of the D^0-A^0 generators have not been adequately explored. The interplay among exciton and charge carrier roles results in different spatial and neutral-pair-separation distributions which are manifested in the subsequent emission. Excitation spectroscopy of D^0-A^0 emission[2] is the best means to address this problem, because of resonant exciton (complexes) photoexcitations and a rapidly varying absorption coefficient with excitation-photon energy. In CdS, several D-A pair continuous-excitation spectroscopy studies have been reported[3,4]. We present here results of our studies at liquid helium temperatures of sub-microsecond time-resolved excitation spectroscopy of D-A emission carried out on pure, un-doped n-type CdS crystals of very different compensation, providing different electric field strengths near the surfaces.

EXPERIMENTAL

A flashlamp-pumped dye laser provides 0.2 nm spectral width, 150 ns photoexcitation pulses of intensities ranging from about 10^3 to 10^5 w/cm^2. The transient D-A luminescence response, spectrally filtered through a monochromator, is recorded at 100 ns intervals. One sample, designated EP-A, is highly compensated(10^{10}ohm-cm resistivity at 300K), and displays persistent photoconductivity below 240K. The other, designated UHP, is relatively uncompensated(2 ohm-cm). We estimate E fields of $\sim 10^3$ and $\sim 10^4$ V/cm over surface regions of the order of 1 μm and 100 nm, respectively, for these samples due to surface state filling from neutral donors.

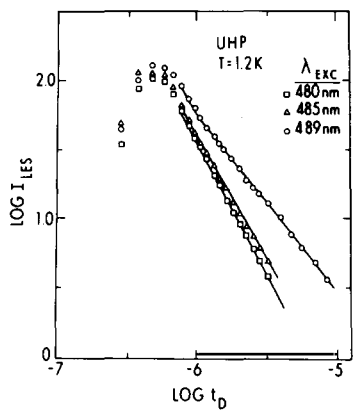

FIG. 1. Decay of D^0-A^0 recombination radiation at 518 nm. I_{LES} in arb. units; t_D in seconds.

RESULTS

The LES (low energy series) emission intensity at 518 nm for the UHP sample is shown in Fig. 1 as a function of t_D, the time delay after firing of the laser pulse. We define a decay rate parameter α as the negative of the decay curve slope; it is fairly constant (power law decay) in the limited time domain shown by the solid lines. The decay is faster in the strongly absorbing exciton-region, λ <485.5 nm. Fig. 2 shows excitation spectra, with t_D as a parameter. At particular absorption resonances, for example the A(n=1) free exciton at 485.5 nm and the bound-exciton I_{1A} at 488.8 nm, we note emission intensity dips, also observed in continuous-excitation spectroscopy studies[4]. Our time-resolved study further shows the growth of the *relative amplitudes* of these dips with increasing

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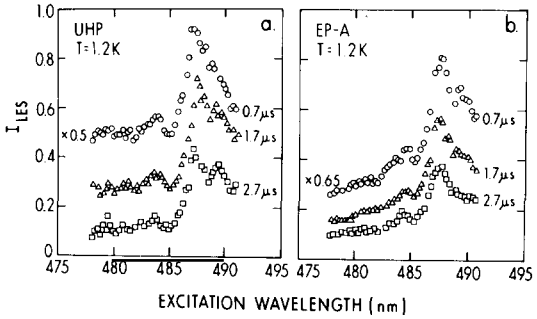

FIG. 2. Excitation spectra of D^0-A^0 recombination radiation at 518 nm. I in a and b in a.u.

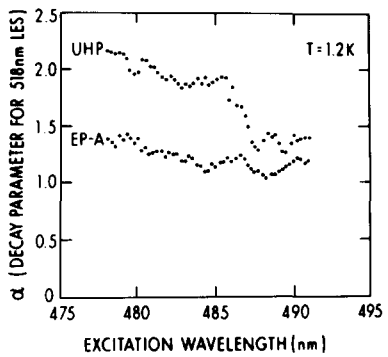

FIG. 3. Excitation spectra of decay parameter α.

t_D for the UHP, but not for the EP-A sample. In Fig. 3, the spectral dependence of the decay parameter α is plotted for both samples. Of special interest is the abrupt decrease for the UHP sample in α as λ increases above the exciton-region boundary.

DISCUSSION

Since exciton lifetimes are short compared with the time scale of these experiments, interpretation must be based on the status of D^0-A^0 pairs at about 10 ns after cessation of excitation, and subsequent processes occurring on a time scale of microseconds, such as D^0-A^0 recombination, and redistribution by hopping or tunnelling of inhomogeneously produced A^0 and D^0 density. Dips at excitation absorption peaks are expected simply because fewer D^0-A^0 pairs at saturating light intensity are generated in the reduced absorption-region. Furthermore, sufficiently large surface electric fields can preclude stable shallow D^0 formation. From such considerations alone, the *relative* size of these dips should not change with increasing t_D. However, with an increased decay rate of pairs associated with a given emission λ in the near-surface region, compared with those in the interior, such as may occur with a large near-surface E field, *relative* dips which grow with increasing t_D result. This explains most features in Figs. 2 and 3, if one ascribes a larger surface E field to the UHP sample. The I_{1A} dips are partially accounted for by the above arguments, but other mechanisms must also be considered. In effect, I_{1A} resonance absorption removes A^0 pair recombination capability. Since every I_{1A} break-up does not necessarily result in an A^0, dips can result. If I_{1A} formation or break-up probabilities select for distant A^0-$D^{+,0}$ pairs, relative dips which increase with t_D could occur, since distant pairs contribute more importantly at longer t_D. Non-saturation of close pairs due to weaker absorbing I_{1A} excitation radiation can also skew the D^0-A^0 pair separation distribution, especially at low light intensities.

A variational method calculation was made of the dependence of α, at particular emission λ, on E. The principal contribution comes from the final state D^+-A^- electric dipole interaction with E, and we found that E ~10^4 V/cm yields a decay rate increase consistent with our experiments. A quantitative comparison would require account taken of changes in the LES spectrum with electric field[5]. Experiments at higher temperatures, low excitation intensities, other emission wavelengths, and on samples with mixed (etch-polish) front and rear surface treatments[6], support our interpretation.

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