

Electronic transitions in CdTe under pressure

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We present a photoluminescence study of CdTe:Sb at 5 and 15 K under hydrostatic pressures of 0 to 32 kbar. We determine the pressure coefficients of several electronic transitions: the neutral-acceptor-bound exciton A^0X , its phonon replica A^0X -LO, the electron-to-acceptor transition $e-A^0$, its phonon replica $(e-A^0)$ -LO, and four donor-acceptor peaks. A nonlinear pressure behavior is found for the main exciton peak. The linear term is 7.6 ± 0.2 meV/kbar. We find that the binding energies of the A^0X and the bare acceptor change with pressure. This change influences acceptor-bound magnetic polaron binding energies in the $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ alloys.

INTRODUCTION

The growth of II-VI semiconductor heterostructures by molecular-beam epitaxy has produced unusual combinations of strained-layer superlattices that can be tailored to specific device applications. Among these are CdTe/ZnTe, CdTe/ $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$,¹ and CdTe/ $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$.² Due to lattice mismatch, the valence-band discontinuities depend on the interlayer strain as well as on the bulk energy gaps. The strain in the layers causes a splitting of the light- and heavy-hole bands, and the alternation of compressive and tensile stresses causes different well depths for light and heavy holes. It is therefore vitally important to know accurate hydrostatic and uniaxial deformation potentials of the bulk constituents in order to calculate strain-induced shifts in the valence bands.

In our present experiment, performed using photoluminescence (PL) at 5 and 15 K, we observe A^0X , its phonon replica (A^0X -LO), $e-A^0$, its LO-phonon replica [$(e-A^0)$ -LO], and four deeper levels due to donor-acceptor (DA) recombination. We have obtained accurate linear and sublinear pressure coefficients for the appropriate transitions. Early measurements³⁻⁵ of the pressure coefficient of CdTe were limited to the band gap, and were carried out at room temperature and 77 K with use of absorption and reflectivity. A recent work reports the pressure coefficients of the exciton and a DA peak at 2 K using PL.⁶ The sample these authors used exhibited intense DA recombination, with the exciton appearing as a weak peak on the high-energy DA tail. This feature caused signal-to-noise problems, allowing them to obtain only a linear pressure coefficient. We will show that their inability to determine the second-order coefficient led to a linear term that is 15% smaller than our value, and consequently to errors of similar magnitude in their renormalization of previous uniaxial stress data. The errors are significant when these deformation potentials are used in calculations involving related strained-layer heterostructures.

Further impetus for studying bulk CdTe comes from recent interest in the diluted magnetic semiconductor (DMS) alloy $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$. The DMS alloys display novel spin-dependent phenomena arising from the large $sp-d$ exchange interaction. Among these novel phenomena are bound magnetic polarons (BMP's), which are ferromagnetic spin clusters caused by the exchange interaction between the spin on the magnetic ion and a carrier spin localized at an impurity. The formation of BMP's gives rise to a magnetic binding energy (BE) in addition to the usual Coulombic term in acceptor-related transitions. In $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$, BMP effects have been observed for both the neutral-acceptor-bound exciton,⁷ A^0X , and the bare acceptor,⁸ via the electron-to-acceptor transition $e-A^0$. Just as the Coulombic BE's for $e-A^0$ are larger than that of A^0X , the BMP BE's are larger, causing $e-A^0$ to shift rapidly away from A^0X with increasing Mn concentration up to $x \sim 0.25$.

We have recently investigated the tuning of the magnetic interactions in $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ with pressure.⁹ We have found that the bare acceptor does not follow A^0X : the energy separation between $e-A^0$ and A^0X decreases with pressure for small Mn compositions ($x=0.05$), while it increases with pressure for larger compositions ($x=0.15$). Since the BE consists of Coulombic and magnetic terms, it is critical to disentangle the two. The pressure dependence of the Coulombic part is measured in the present experiment. We find that the Coulombic BE of A^0X increases faster with pressure than the BE of the bare acceptor, causing a net decrease in their separation. This allows us to determine the Coulombic "base line" and therefore the changes in the magnetic BE's in $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$.

EXPERIMENTAL DETAILS

CdTe doped with $\sim 10^{17}$ Sb/cm³ was grown using the vertical Bridgman technique. The sample was cleaved into a piece about $100 \times 100 \times 30$ μm^3 and loaded in a Merrill Bassett diamond-anvil cell. Argon, loaded cryo-

generally, was used as the pressure-transmitting medium. Fluorescence from the R_1 - R_2 ruby lines was used to calibrate the pressure. A 0.85-m-focal-length double-grating monochromator or a 1-m focal-length single-grating monochromator with a GaAs photomultiplier and photon-counting electronics were used. Data were taken at 5 K (using a continuous-flow cryostat) or at 15 K (using a helium refrigerator). The pressure was measured accurate to ~ 0.25 kbar, and the pressure homogeneity, determined from the linewidth of the exciton, was better than ± 0.5 kbar at the highest pressures. Data were obtained in the pressure range of 0–32 kbar, beyond which CdTe undergoes a phase transition to a metallic rocksalt phase and luminescence is quenched. The luminescence does not recover upon reducing the pressure: the $\sim 20\%$ decrease in volume¹⁰ produces a large number of defects and microcrystallites that quench the radiative transitions.

RESULTS AND DISCUSSION

Photoluminescence (PL) spectra were excited with 0.5–2 mW of 5145-Å radiation from an argon-ion laser. The luminescence was intense. For example, the 1-bar spectrum shown in Fig. 1 was excited with 1 mW on a spot about 30 μm in diameter, and the intensity of the A^0X peak was 3×10^5 counts/sec at 15 K. The relative intensities of A^0X , $e-A^0$, and the DA peaks were found to vary with excitation intensity. We were therefore care-

ful to choose a laser intensity that was convenient for the measurement and maintain it throughout the experiment.

As seen in Fig. 1, at 15 K we observe the A^0X , $e-A^0$, its LO-phonon replica ($e-A^0$)-LO, and deeper levels due to donor-acceptor (DA) recombination,¹¹ labeled DA1 through DA4. The zero-phonon DA is observed as a weak shoulder at 5K. DA1 through DA4 are separated by LO-phonon energies, and are assigned to overtone LO-phonon replicas. Apart from A^0X -LO, which was seen only at 5 K, there were no significant differences between the 5- and 15-K data. The spectrum in the vicinity of the A^0X at 5 K and ambient pressure is shown in the inset of Fig. 1. In contrast to the spectra in Ref. 6, the intensity of the exciton peak remains high through the entire pressure range, allowing an accurate determination of the pressure coefficients.

The energies of the peaks as a function of pressure are plotted in Fig. 2. We fit the energies of all eight peaks to linear and nonlinear functions,

$$E(P) = E(0) + \alpha P, \quad (1)$$

and

$$E(P) = E(0) + \alpha P + \beta P^2, \quad (2)$$

where P is the pressure in kbar. We found that the residual sum of squares for A^0X , A^0X -LO, $e-A^0$, and ($e-A^0$)-LO transitions was 3–6 times lower when we used the nonlinear function, Eq. (2), than when we used the linear

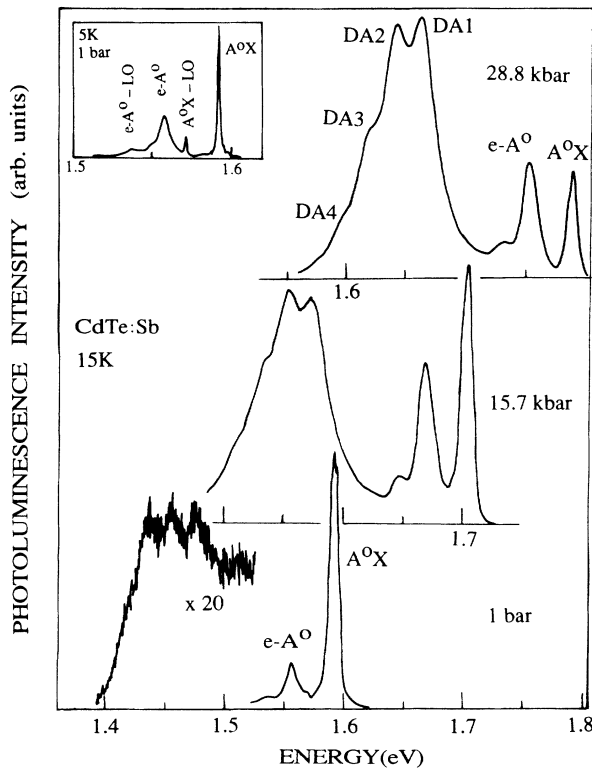


FIG. 1. PL spectra of CdTe:Sb at 15 K for several pressures. The A^0X , $e-A^0$, ($e-A^0$)-LO, and four DA transitions are observed. Inset is a 1-bar spectrum at 5 K, where A^0X -LO is seen.

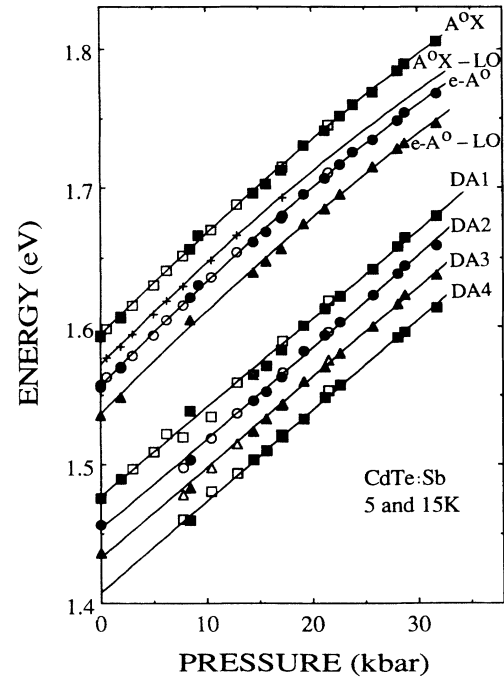


FIG. 2. Energies of the peaks observed in CdTe(Sb) as a function of hydrostatic pressure. The solid lines are least-squares fits to a nonlinear function [Eq. (2)] for A^0X , A^0X -LO, $e-A^0$, and ($e-A^0$)-LO, and to a linear function [Eq. (1)] for DA1 through DA4. The parameters obtained are listed in Table I. The open (solid) symbols are 5-K (15-K) data. The fits are for the 15-K data except in the case of A^0X -LO.

TABLE I. Pressure coefficients of observed transitions.

Transition	$E(0)$ (eV)	α (meV/kbar)	β (meV/kbar ²)	Comments
Present work (CdTe, 15 K unless indicated otherwise)				
A^0X	1.594 ± 0.002	7.59 ± 0.19	-0.029 ± 0.007	
A^0X -LO (5 K)	1.572 ± 0.002	7.74 ± 0.20	-0.038 ± 0.007	
$e-A^0$	1.556 ± 0.002	7.91 ± 0.18	-0.038 ± 0.005	
$(e-A^0)$ -LO	1.536 ± 0.002	7.78 ± 0.24	-0.034 ± 0.007	
DA1	1.477 ± 0.002	6.39 ± 0.09		
DA2	1.452 ± 0.002	6.60 ± 0.08		
DA3	1.432 ± 0.002	6.52 ± 0.07		
DA4	1.407 ± 0.002	6.57 ± 0.06		
Previous works (CdTe)				
Band edge (Ref. 5)		8.0 ± 0.2		77 K, absorption, reflectivity
Band edge (Ref. 3)		7.9 ± 0.2		300 K, reflectivity
Band edge (Ref. 4)	1.483	8.3	-0.004	300 K, absorption
Exciton (Ref. 6)		6.5 ± 0.2		2 K, PL
Other related alloys				
A^0X (Ref. 9)	1.663 ± 0.001	7.91 ± 0.17	-0.036 ± 0.005	15 K, $\text{Cd}_{0.95}\text{Mn}_{0.05}\text{Te}$
A^0X (Ref. 9)	1.826 ± 0.001	7.66 ± 0.2	-0.032 ± 0.007	15 K, $\text{Cd}_{0.85}\text{Mn}_{0.15}\text{Te}$

function, Eq. (1). In contrast, the deeper levels did not show a significant nonlinearity when Eq. (2) was used, and the residual sum of squares did not differ much between Eq. (1) and Eq. (2). We therefore conclude that the shallow effective-mass-like states have nonlinear shifts with pressure, while the DA peaks shift linearly with pressure.

The pressure coefficients are listed in Table I. α and β are consistent with the values for A^0X that we have obtained for the $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ alloys.⁹ Table I also lists pressure coefficients obtained from previous works. The 300- and 77-K measurements of the band edge give α 's close to our α for A^0X (Refs. 3–5). In contrast, there is a large (15%) disagreement between our work and that of Dunstan *et al.*⁶ who use PL at 2 K, and obtain a much smaller α . One would expect the pressure coefficients of Ref. 6 to be in much better agreement with our measurements, since both were performed at low temperatures. The discrepancy lies in the fact that Dunstan *et al.*⁶ used a linear fit, and their lack of data at higher pressures prevented them from obtaining β . If we fit our A^0X energies to a linear function, Eq. (1), we obtain $\alpha = 6.62 \pm 0.11$ meV/kbar, close to the value of 6.5 meV/kbar obtained in Ref. 6. However, the residual sum of squares is 3 times larger than with a nonlinear fit. The linear and nonlinear fits to A^0X energies are shown in Fig. 3. The linear function misses the data points at both high and low pressures. The residuals (Fig. 3, inset) of the linear fit are large at both high and low pressures, and fall on a parabola (dashed curve), while those of the linear fit scatter more or less evenly about an almost flat line (solid curve). It is therefore clear that the nonlinear fit is superior. Since the nonlinear term β is fairly large,¹² forcing a straight line fit to the data can lead to a significantly smaller α .

The smaller-pressure coefficient of Ref. 6 is particularly

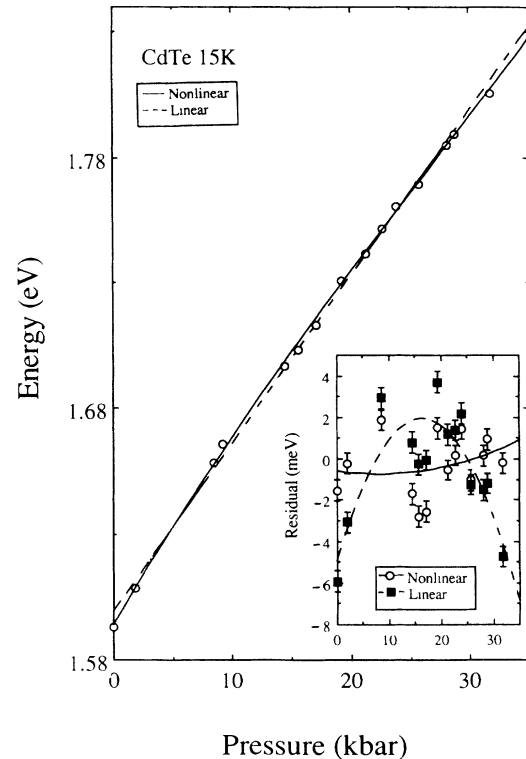


FIG. 3. A^0X energies as a function of pressure fit to linear (dashed line) and nonlinear (solid curve) functions with the parameters described in the text. Note that the linear function clearly misses data points at both high and low pressures. This is illustrated in the inset, where the residual of each fit is shown. The residuals of the linear fit fall on a parabola, with large residuals at low and high pressures (dashed curve), while the residuals of the linear fit scatter more or less evenly about an almost flat line (solid curve).

misleading in the low-pressure range, where the contribution of β is small and $\alpha=7.6$ should be used instead of 6.5 meV/kbar. Reference 6 used this smaller value to correct Thomas's shear deformation potentials,¹³ where there were inconsistencies in the hydrostatic deformation potentials obtained from $\langle 100 \rangle$ and $\langle 111 \rangle$ uniaxial stress data. Corrected with our new α , the shear deformation potentials¹⁴ are $b=1.4\pm 0.4$ eV and $d=3.4\pm 0.6$ eV. The error bars arise principally from the scatter in the uniaxial stress data.

Theoretical calculations of α 's have ranged from 2.8 (empirical pseudopotential)¹⁵ to 5.5 (linear muffin-tin orbitals)¹⁶ and 8 meV/kbar (Phillips-van Vechten).¹⁷ The last two values are in reasonable agreement with our results.

The effect of pressure on excitonic binding energies is fairly small, and is usually not noticed in PL experiments due to the large changes in the band gap with pressure. In the present experiment, however, we find that the energy separation between A^0X and $e-A^0$ changes with pressure, indicating that their Coulombic binding energies change. This energy separation, $\delta=E(A^0X)-E(e-A^0)$, is plotted as a function of pressure in Fig. 4. Despite the scatter, the trend toward a decreasing separation with pressure is clear, decreasing by ~ 2.5 meV over 30 kbar. The changing separation is largely due to the fact that the A^0X BE, which involves the increasing electron effective mass, increases more rapidly than the BE of $e-A^0$, whose heavier hole mass barely changes.

A rough calculation can be made using a simple hydro-

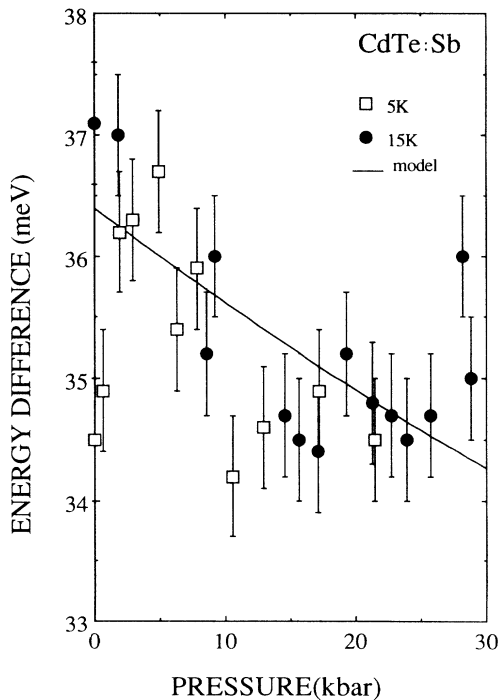


FIG. 4. The energy difference between A^0X and $e-A^0$ as a function of pressure. The solid line is a fit to Eq. (6), as described in the text.

genic picture.¹⁸ The effective Rydberg for A^0X is

$$\mathcal{R}_{A^0X}^* = \frac{e^4 \mu^*}{2\hbar^2 \epsilon_0^2}, \quad (3)$$

where the reduced mass μ^* is given by

$$\frac{1}{\mu^*} = \frac{1}{m_e^*} + \frac{1}{m_{\text{DOS } h}^*}, \quad (4)$$

where $m_{\text{DOS } h}^*$ is the density-of-states (DOS) effective mass for holes; $m_{\text{DOS } h}^* = [(m_{\text{lh}}^*)^{3/2} + (m_{\text{hh}}^*)^{3/2}]^{2/3}$; m_e^* , m_{lh}^* , and m_{hh}^* are the effective masses¹⁹ for electrons ($0.096m_e$), light holes ($0.1m_e$), and heavy holes ($1.09m_e$), respectively; and ϵ_0 is the static dielectric constant. The effective Rydberg for the bare acceptor, \mathcal{R}_A^* is different from Eq. (3) only in that μ^* is replaced by $m_{\text{DOS } h}^*$.

The effective masses and ϵ_0 are affected by pressure. We express the pressure dependence of the rydberg ($\mathcal{R}_{A^0X}^*$ or \mathcal{R}_A^*) as in Ref. 20,

$$\mathcal{R}^*(P) = \mathcal{R}^*(0) \exp(-2\kappa P) \frac{m(P)}{m(0)}, \quad (5)$$

where $\kappa = (1/\epsilon_0)(d\epsilon_0/dP)$, and $m(P)$ is the appropriate effective mass at a pressure P . The energy separation $\delta(P)$ then is

$$\delta(P) = \left[\mathcal{R}_{A^0X}^* \left(\frac{m_{\text{DOS } h}^*(P)}{m_{\text{DOS } h}^*(0)} - \frac{\mu^*(P)}{\mu^*(0)} \right) + \delta(0) \frac{m_{\text{DOS } h}^*(P)}{m_{\text{DOS } h}^*(0)} \right] \exp(-2\kappa P). \quad (6)$$

With the exception of κ , all the quantities in Eq. (6) are fairly well known. To our knowledge, there have been no experiments that measure ϵ_0 as a function of pressure in CdTe. The scatter in our data makes it difficult to use it to determine κ . We therefore use the following approach. We calculate the changes in the masses with pressure using the Kane three-band model²¹ and use $\delta(0) = 36.4$ meV, the zero-pressure energy separation between A^0X and $e-A^0$. Since ϵ_0 is expected to decrease with pressure,²² we choose values of κ that give us values of $\mathcal{R}_{A^0X}^*$ that are typical for A^0X (25 ± 5 meV).²³ We fit our data to Eq. (6) using $\kappa = -10^{-4}$ to -5×10^{-4} , using $\mathcal{R}_{A^0X}^*$ as an adjustable parameter, and obtain $\mathcal{R}_{A^0X}^*$ to be 24 to 32 meV, respectively, which are reasonable values for the BE's. These fits (which are indistinguishable for the above-mentioned κ 's) are shown in Fig. 4 as the solid line.

The κ 's used above are consistent with what one expects. An order-of-magnitude estimate can be obtained from the Penn formula²⁴

$$\epsilon_0 \approx 1 + (\hbar\omega_p / E_g)^2, \quad (7)$$

where $\hbar\omega_p$ is the plasma frequency, and E_g is the Penn gap. Since the Penn gap is close to the E_2 gap, corresponding to a maximum in the reflectivity,²⁵ it is reasonable to assume that pressure-induced changes in E_2 are followed by E_g . A theoretical calculation has obtained²⁶ $\alpha(E_2) = 3$ meV/kbar. Using theoretical values at ambient pressure of $E_g = 5.79$ and $\epsilon_0 = 10.3$ (Ref. 27), and assum-

ing that $\alpha(E_2)=\alpha(E_g)$, we calculate ϵ_0 at 30 kbar to be 10.0, giving $\kappa=-9\times 10^{-4}$, close to the order of magnitude of κ that gives reasonable values for $\mathcal{R}_{A^0X}^*$. Incidentally, holding $\kappa=0$ gives $\mathcal{R}_{A^0X}^*=22$ meV, indicating that a large fraction of the changes in $\delta(P)$ arise from m_e^* (and therefore A^0X).

The above is an order-of-magnitude calculation that shows the origins of the trends in $\delta(P)$. A full calculation would require refinements in $\epsilon_0(P)$ and in calculating the effective masses, as well as a more sensitive measurement of $\mathcal{R}_{A^0X}^*(P)$.

These measurements are of relevance in studies of $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$, where the $(A^0X)-(e-A^0)$ separation has both Coulombic and magnetic contributions. The pressure dependence of the Coulombic part is established in this experiment. The changes in the separation are about 3 times larger⁹ in $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ than in CdTe , implying that the magnetic binding energies change more than the Coulombic part.

We have also observed systematic changes in the relative intensities of the A^0X , $e-A^0$, and DA peaks as a function of pressure. As mentioned before, the relative intensities are sensitive to the incident laser intensity and pressure. The spectra shown in Fig. 1 show how the DA

intensity rises relative to A^0X at high pressures. All spectra were taken with a laser power of 1 mW. The pressure dependence suggests a resonance due to a higher band.

CONCLUSIONS

We have obtained accurate linear and sublinear pressure coefficients for the A^0X , $A^0X\text{-LO}$, $e-A^0$, and $(e-A^0)\text{-LO}$, and linear pressure coefficients for the four deeper levels due to donor-acceptor (DA) recombination. We find that the A^0X and $e-A^0$ binding energies increase with pressure, causing a net decrease in the separation. This change is of importance in studies on the bound magnetic polaron in $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$.

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and $D_{u'}=2.9\pm 0.5$ eV. The numbers calculated in Ref. 6 were $D_u=1.83$ eV and $D_{u'}=2.57$ eV.

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