Motion-dependent magnetic properties of excitons in CdTe

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Very large changes in the Zeeman splittings and in the diamagnetism of excitons as they acquire kinetic energy in wide quantum wells of CdTe are reported. The changes are found to be functions of the translational wave vector K_z of the exciton in the growth direction of the well, irrespective of the width of the well, and are also found to be strong functions of the direction of the magnetic field. The behavior is accounted for by a model in which mixing occurs between the hydrogenic states which describe the exciton in the center of mass or adiabatic approximation. The mixing is ascribed to terms which arise in the Luttinger Hamiltonian when it is extended to describe excitons. Excellent quantitative agreement with experiment, including the results of changing the strain in the wells, is obtained by using Luttinger parameters close to those previously reported. The model is applicable to wide quantum wells made from any zinc-blende semiconductor and confirms that the huge motion induced changes in magnetic properties, observed here for CdTe and previously also for ZnSe and GaAs, should be universal for such materials.

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I. INTRODUCTION

In semiconductors, neutral excitons are formed from electron-hole pairs bound together by the Coulomb interaction. In most bulk semiconductors they have energy-level structures which are analogous to that of a hydrogen atom. Excitons interact strongly with light, so that much of what is known about them comes from studies of their optical properties, especially in magnetic fields. Because of the need for wave-vector conservation during the interaction between excitons and photons, most magneto-optical studies have concerned excitons with wave vectors which were comparable with those of the photons and therefore small, so that the excitons were essentially at rest. Relatively little is known, therefore, about how the properties of excitons change when they acquire kinetic energy. It is such changes that form the subject of the present paper.

The essential feature of our study is the use of quantum wells (QWs) that are very wide in comparison to the Bohr radius of the exciton and in which the changes in energy which result from quantum confinement of the exciton are smaller than the binding energy due to the Coulomb interaction. The two-particle motion can then be considered within the "adiabatic" approximation (e.g., Refs. 1–5), in which the exciton is treated as a (hydrogenlike) composite particle formed by the electron and hole mutually orbiting each other (the internal motion) plus a translational motion of their center of mass. The translational motion can be considered in terms of a particle in a box model and, if the quantum well is sufficiently deep, the component of the exciton wave vector perpendicular to the plane of the well (i.e., parallel to the growth direction, taken to be the z axis) has a magnitude K_z which is quantized according to $K_z=N\pi/L$, where N is a nonzero integer and L is the width of the well (corresponding to there being N half-wavelengths of the particle wave function within the well). States with different values of N have slightly different energies, so that when the electron and hole recombine, the resulting photon energies are functions of N and can be resolved with a high-resolution spectrometer (e.g., Refs. 6–8). These different emission lines correspond to different values of the kinetic energy of the exciton and the behavior of each under an applied magnetic field can individually be studied. In this way we have found that the magnetic properties, and hence the electronic structure of the excitons, are strong functions of the kinetic energy.

The work reported here concerns quantum wells of the binary II–VI semiconductor CdTe. This material is particularly appropriate because of the relatively high exciton binding energy (11 meV) (Ref. 9) and because quantum wells of large width can be produced with uniform strain in which the linewidths in the optical spectra are sufficiently narrow. Preliminary studies of the phenomenon have been reported previously,[8] not only for CdTe but also (with limited data) for GaAs and ZnSe, and a tentative model was proposed in which the magneto-optical properties were accounted for by motion induced mixing between the $1S$ hydrogenlike ground state and higher lying states such as $2P$. In the present paper we provide a full description of the CdTe magneto-optical properties for a greater range of well widths and for a full range of field orientations. We consider not only the motion induced changes in the exciton magnetic moments (as characterized by the gyromagnetic ratios or g values) but also the

TABLE I. Details of the CdTe wide quantum well samples. For those on (001) substrates, S is the strain-induced splitting between the light-hole and heavy-hole states (the latter lying lower) at $K_z=0$. The first four rows refer to Grenoble specimens and the fifth to those from Warsaw.

Substrate	Barrier	Barrier height (meV)	Well widths (Å)	S (meV)
$Cd_{0.96}Zn_{0.04}Te$ (001)	$Cd_{0.90}Mn_{0.10}Te$	150	1442, 1671	~12
$Cd_{0.96}Zn_{0.04}Te$ (001)	$Cd_{0.92}Zn_{0.08}Te$	43	660	~10
$Cd_{0.88}Zn_{0.12}Te$ (001)	$Cd_{0.92}Zn_{0.08}Te$	43	370	~36
$Cd_{0.97}Zn_{0.03}Te$ (110)	$Cd_{0.94}Zn_{0.06}Te$	30	660	
CdTe/GaAs (001)	$Cd_{0.7}Mg_{0.3}Te$	460	400, 800, 1100, 1500	~15

changes in the diamagnetism. We examine the mixing process in greater detail and, as a result of the additional data, obtain an improved theoretical model.

The plan of the paper is as follows. Following a description of the experimental details, we present data for a series of quantum wells with a wide range of widths grown on (001) planes. The spectra obtained for states of different N are characterized in terms of their Zeeman splittings (g values) and diamagnetic shifts under magnetic fields both normal to and inclined to the growth axis. The effects of changing the strain in the well are then discussed. Experimental data for a (110) quantum well are then described. A simple model is considered in which the mixing between the hydrogenic states is caused by terms in the well-known Luttinger Hamiltonian[10] for electrons and holes in semiconductors, as extended by later workers[1–3] to describe the properties of excitons. In particular, we invoke mixing between the $1S$ ground state of the exciton, which is of heavy-hole (HH) character, and excited nP light-hole (LH) states. The model accounts not only qualitatively but also quantitatively for the observed magneto-optical properties.

II. EXPERIMENTAL DETAILS

A. Details of the specimens

The specimens were produced by molecular-beam epitaxy and were from two sources, thus providing a series of different CdTe well widths, different barrier materials, and different values of strain.

(a) *Specimens grown at Grenoble.* These were of three types. (i) The specimens with largest CdTe well widths (1442 and 1671 Å) had barriers of $Cd_{0.90}Mn_{0.10}Te$. They were grown on $Cd_{0.94}Mn_{0.06}Te$ buffer layers (typically 4 μm thick) on (001) $Cd_{1-x}Zn_xTe$ substrates with $x\approx 0.04$. In these quantum wells the strain was such as to cause the HH exciton states at $K_z\approx 0$ to lie lower than the LH states by an amount $S\approx 12$ meV. In zero field, the barrier height for heavy-hole excitons is of order 150 meV (see Sec. III H for the effects of magnetic fields on these barrier heights). (ii) Other specimens had CdTe wells but with barriers of $Cd_{0.92}Zn_{0.08}Te$ and were grown on (001) $Cd_{1-x}Zn_xTe$ substrates with $x\approx 0.04$ (for which the well width was $L=660$ Å with $S\approx 10$ meV) or with $x\approx 0.12$ (for which $L=370$ Å and $S\approx 36$ meV). The barrier height for heavy-hole excitons is approximately 43 meV. (iii) A further specimen (with $L=660$ Å) was grown with $Cd_{0.94}Zn_{0.06}Te$ barriers on a (110) $Cd_{1-x}Zn_xTe$ substrate with x nominally equal to 0.03. The barrier heights for exciton states are of order 30 meV (see also Sec. IV G for the nature of these states).

(b) *Specimens grown at Warsaw.* Here CdTe/(001)GaAs hybrid substrates were used, with the CdTe thickness being 4.5 μm. Prior to the growth of the actual QW structure, a 0.2 μm thick $Cd_{1-y}Mg_yTe$ graded buffer (with y increasing smoothly from 0 to 0.3), followed by a 1 μm thick buffer layer of $Cd_{0.7}Mg_{0.3}Te$, was deposited; the barrier layers for the CdTe quantum well were also of $Cd_{0.7}Mg_{0.3}Te$ and for these structures the strain splitting S was approximately 15 meV. The CdTe well widths ranged between 400 and 1500 Å. The barrier height for heavy-hole excitons is approximately 460 meV.

The relevant sample properties are summarized in Table I.

B. Magneto-optical experiments

The specimens were studied with photoluminescence (PL), transmission, reflectivity, and photoluminescence excitation (PLE) spectroscopy in magnetic fields up to 8 T. Most experiments were carried out with the specimens at 2 K, with some additional measurements at 26 K. For the PL studies, a helium-neon laser was used, while for PLE the specimens were excited by a tunable Ti-sapphire laser. The specimens could be turned about an axis perpendicular to the magnetic field such that experiments could be carried out in either the Faraday or the Voigt configuration (magnetic field, respectively, parallel to or perpendicular to the quantum well growth direction and to the direction of light propagation) and also at intermediate orientations of the field.

III. EXPERIMENTS ON (001) QUANTUM WELLS

A. PL in zero field

If the quantum wells were infinitely deep, the energies of photons emitted during the recombination of heavy-hole excitons in states of a particular value of N would be given by

$$E_N = E_0 + N^2h^2/8M_{HH}L^2, \quad (1)$$

where $M_{HH}=m_e^*+m_{HH}^*$ is the HH exciton translational mass for motion in the growth direction (m_e^* is the electron effective mass and m_{HH}^* is the HH effective mass for this direction). E_0 is the exciton recombination energy in a well of

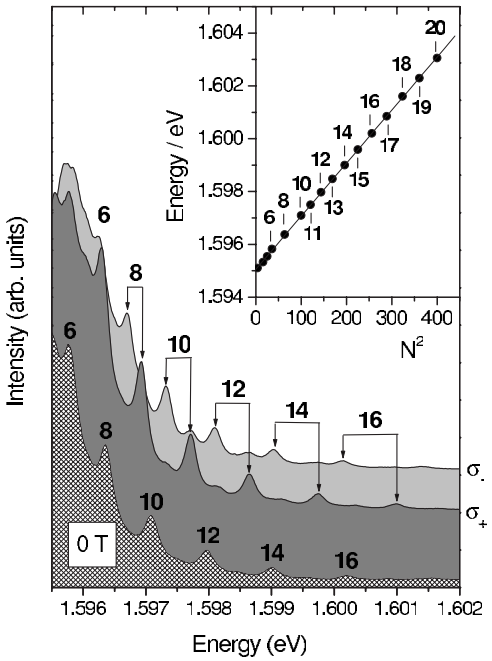

FIG. 1. PL spectra at 2 K from a 1671 Å wide CdTe quantum well on a $Cd_{0.96}Zn_{0.04}Te$ substrate. The lowest trace is at zero-magnetic field; the central and uppermost traces are at a magnetic field of 4 T in σ_+ and σ_- polarizations, respectively. The inset shows how the zero-field transition energies depend on N^2. The values of the translational quantization index N are shown (in the inset, solid circles represent even values of N and open circles represent odd values). The slope of the line in the inset corresponds to a translational mass of $0.67m_0$, in good agreement with the value of $0.64m_0$ expected from the data of Ref. 11.

infinite width. Figure 1 shows the PL spectrum from a Grenoble CdTe well, in which a series of clearly resolved signals corresponding to different values of N is observed.

The relative intensities of the signals with different values of N are predicted to be proportional to the squares of the Fourier components $F_N^2(k_{\text{photon}})$ of the exciton envelope function at the wave vector $k_{\text{photon}}=2\pi/\lambda_{\text{photon}}$ of the photon in the material (it is the existence of such components which makes it possible for the photon to interact with the exciton).[6,7] Here λ_{photon} is the wavelength in the material (~2500 Å) (Ref. 7) and

$$F_N^2(k_{\text{photon}}) = A_N^2 \cos^2(k_{\text{photon}}L/2)$$

for odd values of N and

$$F_N^2(k_{\text{photon}}) = A_N^2 \sin^2(k_{\text{photon}}L/2)$$

for even values, where

$$A_N^2 = \{(N\pi/L)/[(N\pi/L)^2 - k_{\text{photon}}^2]\}^2(2/L).$$

In the particular example of Fig. 1, $(L/\lambda_{\text{photon}})=0.67$, so that the signals for even values of N are expected to be much stronger than those for odd values, as is observed. For well widths of about 625 Å, the even and odd transitions should have the same value of F_N while for wells narrower than this, it is the odd transitions which are expected to become stronger.

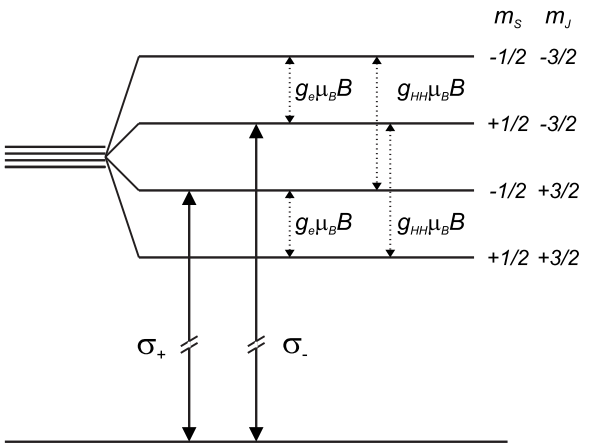

FIG. 2. Energy levels in a magnetic field for a heavy-hole exciton at rest for the magnetic field along the z axis (see text). The electron-hole exchange interaction is assumed negligible. The allowed optical transitions for creation/recombination of the exciton are shown by the solid vertical lines. The diagram is drawn for negative values of g_{HH} and g_e, with $g_{\text{HH}} > g_e$.

In the inset of Fig. 1 the photon energies at zero-magnetic field are plotted against N^2 and show the expected linear dependence, confirming that the infinitely deep well provides a good description of the behavior. Similar results were obtained for all the specimens described in the present study and the infinite well provides a good approximation to the data. In the case of specimens with $Cd_{0.90}Mn_{0.10}Te$ barriers, the depth of the quantum wells can be changed significantly in the presence of magnetic fields at low temperatures and corrections for this have to be made as outlined in Sec. III H.

Strictly, the spectra should be considered within the coupled exciton-photon (polariton) system (as in Refs. 7 and 12); however, the smallest value of K_z for which the splittings caused by magnetic fields could be resolved (see Sec. III B) was about 1×10^6 cm^{-1}, which is well above the region of the polariton dispersion curve where the exciton-photon interaction is strong (see, e.g., Ref. 7), so that the simple uncoupled excitonic description is valid.

Finally, although Fig. 1 shows a PL spectrum, it is possible to observe the excitonic transitions in reflectivity, PLE, and transmission spectra, in some cases with sharper signals and with values of N as high as 30. The energies of the transitions obtained by the different techniques are in good agreement with each other.

B. Effects of a magnetic field along [001]

In our (001) CdTe quantum wells, it is the heavy-hole exciton states that lie lowest. For an exciton at rest (i.e., with zero translational wave vector K_z), the energy levels when a magnetic field B is applied in the [001] direction are as shown in Fig. 2. Here, m_s $(=\pm 1/2)$ and m_J $(=\pm 3/2)$ refer to the magnetic quantum numbers of the electron and heavy-hole, respectively, g_e and g_{HH} are the electron and heavy-hole g values, and μ_B is the Bohr magneton (g_e is negative[13–16] and we shall see later that g_{HH} is also negative).

The selection rules for the optical transitions that create or annihilate the excitons result, for this field direction, in two

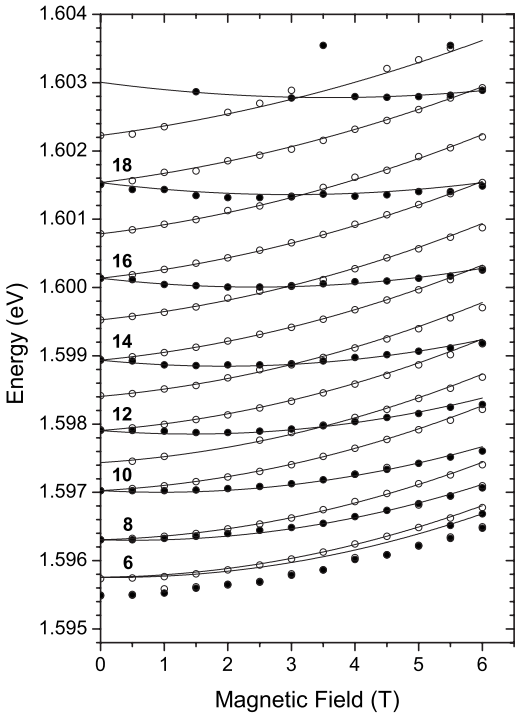

FIG. 3. PL transition energies at 2 K for excitons in a 1671 Å wide CdTe (001) quantum well when a magnetic field is applied along the [001] direction (the Faraday configuration). Open circles denote σ_+ polarization and closed circles σ_-. The translational quantum numbers N are shown. For these excitons, g_{exc} is positive. The continuous curves are of the form of Eq. (4).

dark states and two bright states. Transitions from the bright states are of σ_+ and σ_- polarizations, as shown, and can therefore be distinguished easily. In Fig. 2 we have not included any effects of the exchange interaction between the electron and hole, since the exchange splitting in these wide wells is believed to be small [of order 0.07 meV[17] (see also Sec. III D)] and in any case does not directly affect the Zeeman splitting of the heavy-hole exciton transitions. The energy difference between the σ_+ and σ_- transitions should therefore be linear in magnetic field and we can define an exciton g value g_{exc} such that

$$E_{\sigma+} - E_{\sigma-} = g_{\mathrm{exc}}\mu_B B. \tag{2}$$

For the exciton at rest it follows that[18]

$$g_{\mathrm{exc}} = g_{\mathrm{HH}} - g_e. \tag{3}$$

If the exciton is unaffected when it acquires kinetic energy, each bright transition would be expected to split linearly with magnetic field into two components, the splitting being independent of the translational quantization index N. In addition, the transition energy should show a diamagnetic shift, again independent of N. In practice, this is clearly *not the case*, as can be seen in Fig. 1, where spectra taken in the two polarizations at a finite magnetic field are shown.

This surprising behavior is shown more clearly in the fan diagram of Fig. 3, where the splitting (and hence g_{exc}) is seen to increase considerably as N increases. For each value of N, the two Zeeman components also move upward in energy

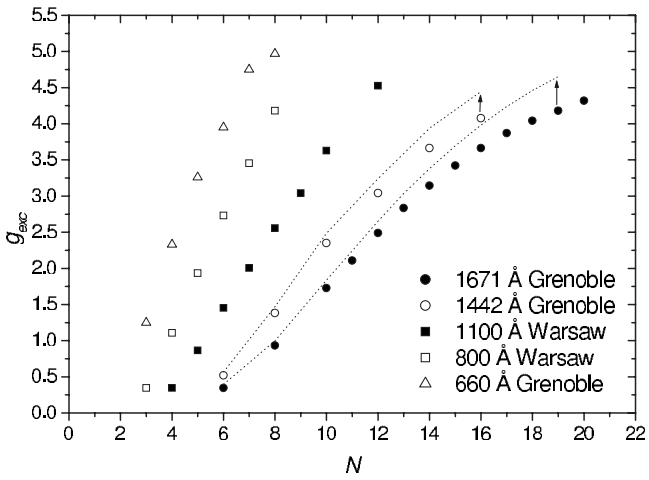

FIG. 4. Values of g_{exc} as functions of the translational quantum number N for a series of (001) CdTe quantum wells of different widths for magnetic fields applied along the growth direction at 2 K. For all these samples, the strain is similar, the heavy-hole to light-hole splittings being in the region of 12–15 meV, except for the 660 Å well, for which it is 10 meV. The data for the 1442 and 1671 Å wells have to be corrected for the effects of magnetism in the barriers, as discussed in Sec. III H; this correction, shown by arrows, leads to the broken curves.

owing to diamagnetic shifts, such that the displacement of the average energy of each pair is proportional to B^2. Close inspection of Fig. 3 shows that these diamagnetic shifts *decrease* as N gets larger; this is also surprising.

The fits to the experimental data in Fig. 3 are of the form

$$E = E_N + DB^2 \pm g_{\mathrm{exc}}\mu_B B. \tag{4}$$

The parameters g_{exc} and D which, respectively, characterize the Zeeman splittings and the diamagnetic shifts depend on N (as shown in Sec. III E, they are in fact functions of $K_z = N\pi/L$).

C. Exciton g values for fields along [001]

For all the (001) quantum wells which we have studied, the values of g_{exc} for magnetic fields along the growth axis (the Faraday configuration) are found to depend on N. Some examples of the behavior are shown in Fig. 4, where it is seen that for narrower wells the change in g_{exc} occurs more rapidly at smaller values of N than in the wider wells. In fact, it is the ratio of N to the well width L, i.e., the value of K_z, and hence the kinetic energy, which controls the value of g_{exc}, as can be seen in Fig. 5, where the values of g_{exc} are plotted against $K_z = N\pi/L$ for the same series of wells. The upper sets of data points in Fig. 5, which are for quantum wells which all have similar LH-HH strain splittings (12–15 meV) but different widths, are shown to fall onto a common curve, indicating a universal dependence of g_{exc} on kinetic energy (as characterized by K_z). We therefore write

$$g_{\mathrm{exc}} = g_{\mathrm{HH}} - g_e + g(K_z). \tag{5}$$

The source of this additional term is discussed in Sec. IV.

When the strain is increased, the values of g_{exc} are strongly affected, as shown in Fig. 5, where the crosses refer

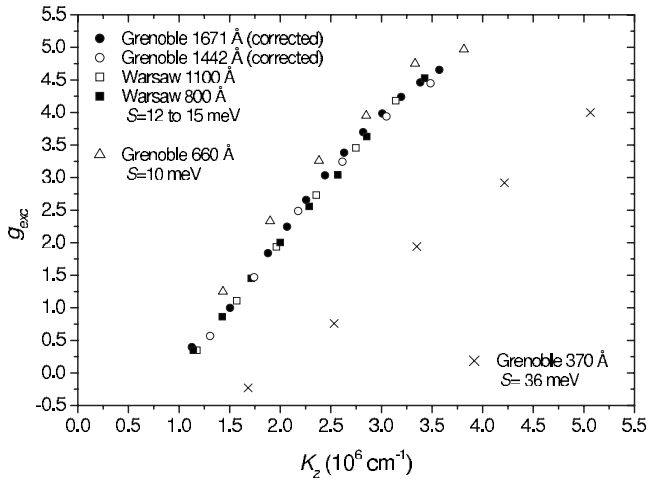

FIG. 5. The values of g_{exc} from Fig. 4 replotted as functions of $K_z=N\pi/L$. The symbols have the same meaning as in Fig. 4. Additional data represented by the crosses are for a (001) CdTe well for which the strain is much larger, the heavy-hole light-hole splitting S being about 36 meV. The data for the 1442 and 1671 Å wells, which have dilute magnetic barriers, have been corrected for field-induced changes in barrier heights (see Sec. III H).

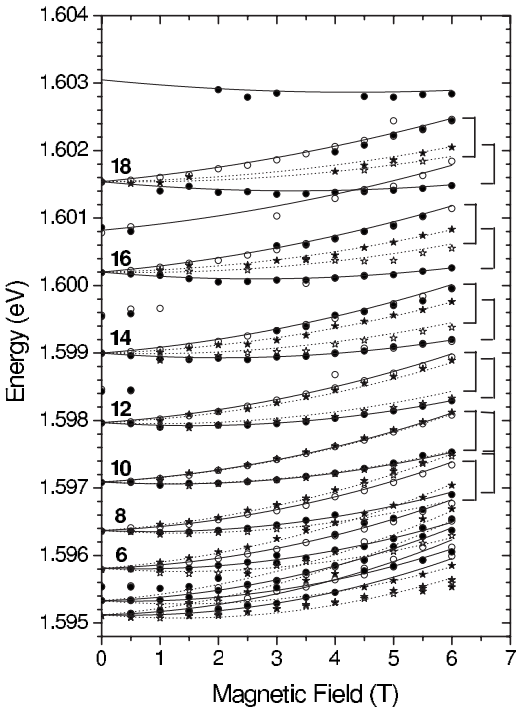

FIG. 6. PL transition energies at 2 K for excitons in a 1671 Å CdTe (001) quantum well when a magnetic field is applied at 60° to the [001] direction. Open circles denote σ_+ polarization and closed circles σ_-. Transitions which are dark when $\theta=0$ become allowed at other angles and are represented by the star symbols and broken curves. The intervals marked on the right-hand side correspond to the electron Zeeman splittings at 6 T; states which were originally bright and dark with the same values of N and of m_J but with opposite values of m_s, differ in energy by this amount.

to additional data, obtained for a CdTe well grown on a $Cd_{0.88}Zn_{0.12}Te$ substrate, for which the LH-HH splitting is about 36 meV. This strain dependence is also demonstrated by the small shift [in the opposite direction, (see Fig. 5)] between the data set for the 660 Å well and the sets for the 800, 1100, 1442, and 1671 Å wells; the strain is lower in the 660 Å well than in the latter wells. The effects of strain are discussed further in Sec. III F. The dependence of g_{exc} on wave vector described above has also been observed in a specimen of GaAs and in a specimen of ZnSe, as reported in Ref. 8.

D. Effects of inclined fields

We now turn our attention to the behavior of the g values for (001) quantum wells when the magnetic field is inclined at an angle θ to the growth axis. As an example, in Fig. 6, we show a fan diagram for the transition energies when $\theta=60°$. There are three aspects of the behavior: first, the Zeeman splittings between the bright states have decreased markedly relative to their values for $\theta=0°$; second, the diamagnetism changes; and third, the previously dark states start to become visible (this is because the large LH-HH strain splitting ensures that m_J remains a good quantum number while, since the electron g value is essentially isotropic, the $m_s=\pm 1/2$ states of the electron become mixed by the inclined magnetic field). In this section we concentrate on the Zeeman splittings; the diamagnetic properties will be discussed in Sec. III E.

On the right-hand side of Fig. 6 we show intervals which represent the *electron* Zeeman splitting (which is essentially isotropic) at a field of 6 T. The energies of the originally dark states differ from those of the bright states by exactly this amount to within the experimental accuracy, so that an upper limit can be set to the magnitude of the electron-hole exchange interaction, assumed to be of the form $a\vec{\sigma}\cdot\vec{J}$, where $\vec{\sigma}$ and $\vec{J}$ refer, respectively, to the electron spin and hole angular momentum. Such an interaction would cause a shift in the average energy of the dark states relative to that of the bright states of order $(9/2)a\cos\theta$ (see, e.g., Ref. 19), so that the data in Fig. 6 show that a is indeed less than 0.1 meV.

Figure 7 shows how the values of g_{exc} extracted from the fan diagrams change as a function of θ. The values of g_{exc} for each value of N are shown and converge at $\theta=90°$ to a common value of $g=1.62$ which is independent of K_z and which corresponds in magnitude to that for an electron.[13–16]

For heavy-hole excitons at rest, one would expect the transitional energies of the bright states to be such that

$$g_{\mathrm{exc}}=g_{\mathrm{HH}}\cos\theta-g_e.$$

Since the electron g value is expected to be essentially isotropic, the data for excitons with finite values of K_z (Fig. 7) show that $g(K)$ in Eq. (5) must also have a $\cos\theta$ dependence, so that

$$g_{\mathrm{exc}}=-g_e+g_{\mathrm{HH}}\cos\theta+g(K_z)\cos\theta. \tag{6}$$

Note here that although the magnetic field is applied at an angle θ to the growth axis, the exciton's translational motion remains quantized along that axis.

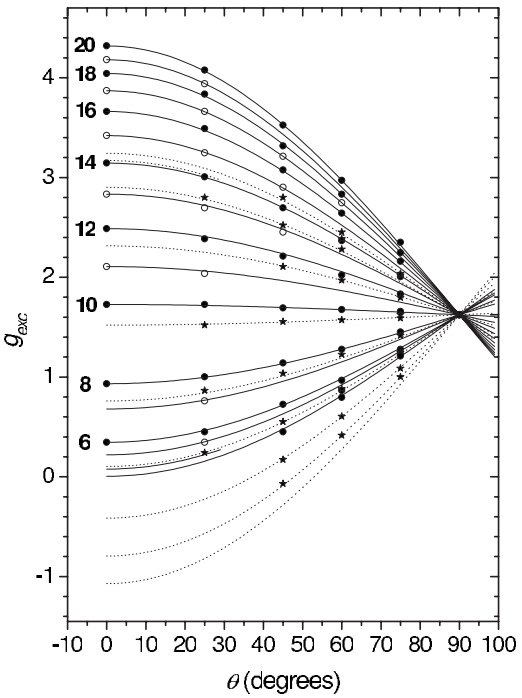

FIG. 7. Values of g_{exc} as a function of the direction of the magnetic field with respect to the growth axis. $\theta=0$ corresponds to the Faraday and $\theta=90°$ to the Voigt configuration. The continuous curves [which are of the form of Eq. (6)] show the behavior for the states of different translational quantization index N which are bright states when $\theta=0$ (for clarity, only the even values of N, represented by filled circles, are specifically indicated; open circles represent odd values of N). At large values of θ previously dark states start to become visible and are represented by the broken curves. Broken curves correspond to states which were dark at $\theta=0$.

An important conclusion from Fig. 7 is that, since g_e is negative, the g values presented in the figure must be positive. This is in agreement with the calculation in Sec. IV.

E. Diamagnetism

The upper sets of data in Fig. 8 show how the diamagnetic parameter D depends on K_z for (001) wells of two different widths and with $Cd_{0.7}Mg_{0.3}Te$ barriers, the field being in the (001) direction (Faraday configuration). The two sets of data superimpose, showing that D, like g_{exc}, is a function of K_z. A clear decrease in D with K_z is observed in these and in all other specimens. To describe the behavior we write

$$D = D_0 + D_0(K_z), \tag{7}$$

where D_0 is the value of the diamagnetic parameter for excitons at rest and for fields along [001].

The lower set of data points is for the field at 90° to the growth direction (Voigt configuration). For these data we write

$$D = D_{90} + D_{90}(K_z), \tag{8}$$

where D_{90} is the value of the diamagnetic parameter for excitons at rest with this direction of the field. In contrast to the behavior of g_{exc}, it is clear that a K_z-dependent variation in the diamagnetic parameter occurs for both the Faraday and Voigt configurations, the variation being greater in the latter. The origin of this behavior is discussed in Sec. IV F.

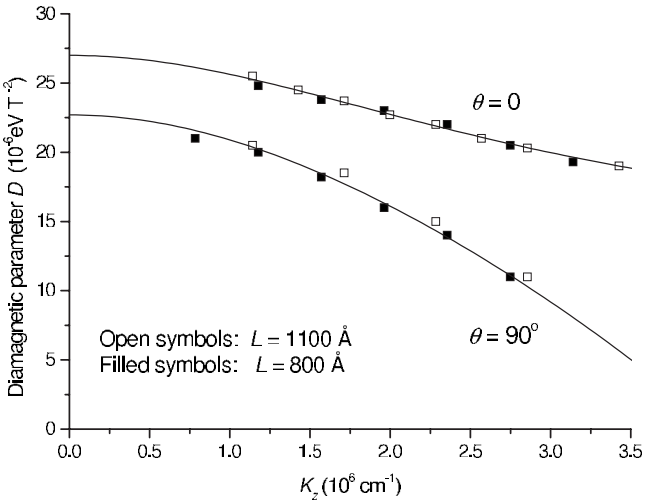

FIG. 8. Dependence of the diamagnetic parameter D on K_z for two CdTe (001) quantum wells with the magnetic field along the growth direction (Faraday, upper data set) and at 90° to it (Voigt, lower set). The continuous curves are calculated using the model in the text (Sec. IV F).

F. Change in strain

The effect of changing strain in the quantum well through changing the substrate and comparing different specimens was shown in Fig. 5. We have carried out an additional experiment in which a specimen with an 800 Å well was glued face down onto a glass slide and the substrate removed by etching. On cooling to 2 K, differential thermal contraction caused the strain-induced LH-HH splitting S in the quantum well to increase by $\delta S=2.2$ meV compared to the original specimen (the value of δS was calculated from the observed much smaller change in the HH transition energy). At the same time, the data points in Fig. 5 shifted to slightly larger values of K_z. For example, the point for which $g_{\mathrm{exc}}=2.5$ was displaced by an amount $\delta K_z=0.14\times10^6$ cm^{-1}. The ratio $\delta K_z/\delta S$ for this strain-induced change is thus about 6×10^4 cm^{-1} meV^{-1}. This is in agreement with the prediction from the analytic formula that will be discussed later in Sec. IV E. These observations for the glass-mounted sample in which nothing but the strain was changed support the supposition that the strain splitting is a key factor in explaining the differences between the sets of data in Fig. 5 rather than some other variable (such as the well depth or well width).

G. Experiments on (110) quantum wells

A PLE spectrum for the CdTe quantum well grown on a (110) substrate was shown in Fig. 3(b) of Ref. 8 for a magnetic field along the growth axis (and hence in the direction of center-of-mass quantization). In contrast to that of (001) quantum wells, the Zeeman splitting of the exciton lines (corresponding to a g value of magnitude 1.3) was found to

be *independent* of wave vector. The diamagnetic constant does, however, decrease with the translational wave vector, the dependence being similar to that in the upper curve of Fig. 8 for the (001) quantum wells.

H. Effects of magnetic barriers

The CdTe quantum wells with widths 1671 and 1442 Å have barriers which are of $Cd_{0.9}Mn_{0.1}Te$ and which are therefore magnetic. In zero applied magnetic field, the barrier height for heavy-hole excitons is about 150 meV. However, when a magnetic field in the [001] direction is present, the barrier heights for excitons with $m_J=-3/2$, $m_s=+1/2$ and with $m_J=+3/2$, $m_s=-1/2$, respectively, increase and decrease because of the strong exchange coupling between the charge carriers and the magnetization of the manganese spin system.[20] The changes in barrier heights are given by

$$\Delta E_B(m_J = \pm 3/2, m_s = \mp 1/2) = \pm \bar{x}(N_0\alpha_e - N_0\beta_e)\langle S_z^{\mathrm{Mn}}\rangle,$$

where $\bar{x}$ is the effective manganese fraction (less than the true concentration x because of antiferromagnetic coupling between any adjacent pairs of manganese ions) and $N_0\alpha_e$ (=−0.22 eV) and $N_0\beta_e$ (=0.88 eV) (Ref. 21) represent the exchange interactions between manganese spin system and the electrons and holes, respectively.

The magnetization of the manganese spin system is given by[20]

$$\langle S_z^{\mathrm{Mn}}\rangle = -\frac{5}{2}B_{5/2}\left(\frac{g_{\mathrm{Mn}}\mu_B B}{k_B(T+T_0)}\right),$$

where $B_{5/2}$ is a Brillouin function for spin 5/2, g_{Mn} (=2.0) is the g value for Mn^{2+} ions, and T_0 is a Weiss constant. For bulk material, with $x=0.1$, $\bar{x}\approx 0.04$ and $T_0\approx 1$ K.

These changes in barrier height can amount to several tens of meV and thus lead to changes in the confinement energies of the excitons. Because the changes in barrier height are different for the two exciton spin states, they result in a contribution to (which turns out to be a decrease in) the spin splitting. The effect becomes more significant for states which are higher in the well, i.e., those with larger values of K_z.

Since $\bar{x}$ and T_0 are strongly affected when $Cd_{0.9}Mn_{0.1}Te$ lies at an interface with the nonmagnetic CdTe,[21] rather than using the bulk values for $\bar{x}$ and T_0, we have chosen values which reproduce the changes in experimental data for the Zeeman splittings when the temperature is raised from 2 to 26 K (the result of the temperature increase is to enhance the exciton Zeeman splittings at the highest values of K_z by about 10%, the direction of the change being consistent with the g values plotted in Figs. 4 and 5 being positive, as shown in Sec. III D). We have then corrected the 2 K data to remove the (negative) contributions to the values of g_{exc} caused by magnetically induced changes in the barrier height. The corrections, which apply to the 1442 and 1671 Å wells, are shown in Fig. 4.

IV. MODEL

A. Background

It has been known for some time that the combination of the relative motion of the electron and hole with the translational motion of the exciton as a whole can lead to remarkable effects. For example, in classic reports by Thomas and Hopfield[22] and by Gross *et al.*,[23] exciton movement perpendicular to a magnetic field was shown to lead to the appearance of a quasielectric field. In contrast, until our own previous report,[8] there appear to have been no accounts of how the magnetic properties of excitons are influenced when their translational wave vectors are *along* the direction of an applied magnetic field, as is the case in the present paper when the field is applied in the growth direction of the quantum well.

As noted in Sec. I, we shall consider the exciton within the adiabatic approximation[3–5] as a hydrogenlike composite particle formed by the electron and hole mutually orbiting each other (the internal motion) plus a translational motion of their center of mass. In the hydrogenic ground state, the wave function for an exciton traveling with translational wave vector K_z can therefore be written as

$$\Psi(m_J, m_s, m_l, K_z) = s\Phi_{1S}(m_J, m_s, m_l)\exp(\pm iK_z Z). \quad (9)$$

Here Φ_{1S} is a hydrogenic function which describes the internal motion (for a $1S$ state, the orbital magnetic quantum number m_l is zero), s is a normalizing factor, and Z refers to the coordinate of the center of mass. In the quantum well, counterpropagating traveling waves form standing waves[6] such that $K_z=N\pi/L$.

In Ref. 8, we suggested that the dependence of g_{exc} on K_z for magnetic fields in the growth direction of (001) wells was a result of mixing between states of the form given above with higher lying nP hydrogenic states which we take to be of the form

$$\Psi(m_J, m_s, m_l, K_z') = s\Phi_{nP}(m_J, m_s, m_l)\exp(\pm iK_z Z). \quad (10)$$

The mixing process is ascribed to a coupling between the internal motion of the electron and hole within the exciton and the translational motion of the exciton itself. The additional experimental data for CdTe wells described in Sec. III provide further evidence for this concept but with considerable modifications to the details of the model discussed in Ref. 8. In particular, the data indicate that the coupling is due to terms in the Luttinger Hamiltonian[10] (which describes the band structure) when these terms are modified to include the translational motion, as will be described below in Sec. IV B.

The approach which we adopt is similar to that used by Altarelli and Lipari[2] in their treatment of the ground-state energies of excitons in low magnetic fields. Altarelli and Lipari[2] calculated the contributions to the Zeeman splitting and diamagnetic shift of the different spin components of the $1S$ exciton ground state as a result of mixing with excited hydrogenic states caused by terms in the modified Luttinger Hamiltonian. The calculation was carried out for excitons which were at rest ($K_z=0$) and in an unstrained environment. In contrast, the calculation that will be described below is for excitons with finite translational wave vector and in a

strained material (so that the HH states are well removed from the LH ones). We shall concentrate only on those contributions to the Zeeman splittings and diamagnetic shifts which depend on K_z, since it is these that we have measured experimentally. In contrast to the work in Ref. 2, we shall not attempt to calculate the exciton g values and diamagnetic shifts for excitons *at rest*, instead, we shall calculate that part of $g_{\rm exc}$ which depends on the translational wave vector.

We note here that, although we have used quantum well structures to study the ways in which the magnetic properties depend on K_z, the mechanism that we propose for the enhancement suggests that the dependence is not simply the result of quantum confinement[24] on the individual g values of the electrons and holes: spin-flip Raman scattering[25] and quantum beat[26,27] studies of electrons in relatively narrow quantum wells show that the electron g factor remains essentially unchanged by confinement effects if the well width remains greater than 300 Å (which is smaller than the narrowest used in the present investigation). We assume that the hole g values are also not strongly affected by confinement in our very wide wells. In Refs. 25–27, the exciton g values were also determined and were used to obtain hole g values through use of Eq. (3) but without allowance for any of the mixing processes that we will discuss below and which lead to replacing Eq. (3) by Eq. (6).

B. Luttinger Hamiltonian and its extension to excitons

Following Eq. 3.8 of Ref. 3 (see also Refs. 1 and 2), the Hamiltonian which describes the exciton with translational wave vector $\vec{\mathbf{K}}$ in a magnetic field can be written in terms of the relative and translational motion of the electron and the hole as

$$H = H^{(c)}\left(\frac{\vec{\mathbf{p}}}{\hbar} + \frac{e\vec{\mathbf{A}}}{\hbar} + \alpha\vec{\mathbf{K}}\right) - H^{(v)}\left(-\frac{\vec{\mathbf{p}}}{\hbar} + \frac{e\vec{\mathbf{A}}}{\hbar} + \beta\vec{\mathbf{K}}\right) - \frac{e^2}{4\pi\varepsilon_r\varepsilon_0 r} + H_{\rm exchange}, \tag{11}$$

where ε_r is the relative permittivity, $\vec{\mathbf{r}}$ is the electron-hole separation, and $\vec{\mathbf{p}}$ is the momentum operator for the electron within the exciton (of equal magnitude but of direction opposite to that of the hole). Here $\alpha = m_e/(m_e+m_{\rm HH})$, $\beta = m_{\rm HH}/(m_e+m_{\rm HH})$, and the vector potential $\vec{\mathbf{A}}$ is given[1–3] by $\frac{1}{2}\vec{\mathbf{B}}\times\vec{\mathbf{r}}$.

The first two terms are obtained from the Luttinger Hamiltonian[10] which describes the energies of conduction-band electrons and valence-band holes in zinc-blende semiconductors (as modified by Cho *et al.*, see Eqs. 3.2 and 3.3 of Ref. 3). We have

$$H^{(c)}(\vec{\mathbf{k}}) = \frac{\hbar^2 k^2}{2m_e^*} + g_e\mu_B\vec{\sigma}\cdot\vec{B} \tag{12}$$

and

$$-H^{(v)}(\vec{\mathbf{k}}) = \left(\gamma_1 + \frac{5}{2}\gamma_2\right)\frac{\hbar^2k^2}{2m_0} - \gamma_2\frac{\hbar^2}{m_0}(k_x^2J_x^2 + k_y^2J_y^2 + k_z^2J_z^2) - 2\gamma_3\frac{\hbar^2}{m_0}(\{k_xk_y\}\{J_xJ_y\} + \text{cycl.perm.}) - 2\mu_B\kappa\vec{J}\cdot\vec{B} - 2\mu_Bq(B_xJ_x^3 + B_yJ_y^3 + B_zJ_z^3), \tag{13}$$

where $\{J_xJ_y\} = \frac{1}{2}(J_xJ_y + J_yJ_x)$, etc. Here γ_1, γ_2, γ_3, κ, and q are the (dimensionless) Luttinger parameters and m_0 and m_e are, respectively, the electron rest mass and the conduction-band effective mass. The directions x, y, and z refer to the crystal [100], [010], and [001] axes. We have omitted k-linear terms in the Hamiltonian since the effects of these are expected to be small. We shall also neglect the final term in Eq. (11), which represents the electron-hole exchange interaction and whose effects, as already noted, are expected to be small.

In Secs. IV C–IV I, we shall consider the mixing between the different hydrogenic states that arises because of the terms in the modified Luttinger Hamiltonian which involve the internal momentum $\vec{\mathbf{p}}$ and the internal coordinate $\vec{\mathbf{r}}$. The mixing causes the energies of the $m_J = 3/2$ and $m_J = -3/2$ exciton ground states to be lowered. The decreases in energy contain terms which are dependent on K_z and which are proportional to B^2, leading to changes in the diamagnetic shifts; they also contain K_z-dependent terms which are linear in B and of opposite sign for $m_J = 3/2$ and $m_J = -3/2$, thus leading to contributions to the Zeeman splitting.

C. Energies of the unperturbed ground spin states in the (001) quantum wells

In the absence of any mixing between hydrogenic states, the energies of the bright spin states of the heavy-hole excitons in a low magnetic field along the [001] direction are given, for $m_J = \pm 3/2$, $m_e = \mp 1/2$, by

$$E_{\pm 3/2,\mp 1/2} = -R + \frac{\hbar^2K_z^2}{2M_{\rm HH}} \pm \frac{1}{2}g_{\rm HH}\mu_BB \mp \frac{1}{2}g_e\mu_BB + D_0B^2$$

relative to the heavy-hole band gap. Here R is the exciton Rydberg constant and D_0 is the diamagnetic parameter for this field direction at $K_z = 0$. The values of $M_{\rm HH}$, $g_{\rm HH}$, and D_0 are derived from the eigenvalues of Eq. (13). It follows that, for magnetic fields in the [001] direction in the (001) quantum well, $g_{\rm HH} = -6\kappa - \frac{27}{4}q \approx -6\kappa$, since the magnitude of q is much less than that of κ (e.g., Ref. 28).

D. Contributions to the Zeeman splitting in (001) wells due to mixing

If the magnetic field is in the zx plane and is directed at an angle θ to the growth axis z, we have $2A_x = -By\cos\theta$, $2A_y = B(x\cos\theta - z\sin\theta)$, and $2A_z = By\sin\theta$, where x, y, and z are the internal coordinates of the electron relative to the hole and $\theta = 0$ and $\theta = 90°$ correspond, respectively, to the Faraday and Voigt arrangements. When these values are substituted into the exciton Hamiltonian of Eq. (11) there arise several terms which can mix the $1S$ ground states with excited states, in particular, those for which the hydrogenic wave function is of nP form. Irrespective of the direction of the magnetic

field, the exciton translational motion remains quantized along the growth axis.

We consider first the terms which arise from the γ_3 part of the Hamiltonian. If only those γ_3 terms which contain K_z are retained (since transverse center-of-mass motion is negligible, particularly since the incident light is at near-normal incidence), we obtain

$$H_{\gamma_3} = H_+ + H_- + H_z,$$

in which

$$H_+ = \frac{\gamma_3 \hbar \beta}{2m_0}\left(p_+ - ir_+\frac{eB\cos\theta}{2}\right)(J_-J_z + J_zJ_-)K_z, \quad (14)$$

$$H_- = -\frac{\gamma_3 \hbar \beta}{2m_0}\left(p_- + ir_-\frac{eB\cos\theta}{2}\right)(J_+J_z + J_zJ_+)K_z, \quad (15)$$

and

$$H_z = \frac{\gamma_3 \hbar \beta}{2m_0} zeB\sin\theta(J_yJ_z + J_yJ_x)K_z. \quad (16)$$

Here $p_\pm = p_x \pm ip_y$, $r_\pm = x \pm iy$, $p_x = -i\hbar\partial/\partial x$, etc. The coordinates x, y, and z are those of the electron relative to the hole.

We consider first the effects of the terms H_+ and H_- (we shall see below that it is these two terms which lead to the wave-vector dependence of the exciton g values, which is not affected by H_z). Since, for hydrogenic states, one can write

$$\langle nP|p_x|1S\rangle = \frac{i\hbar}{2a_B^2}\left(1 - \frac{1}{n^2}\right)\langle nP|x|1S\rangle,$$

etc., and, since we shall be considering mixing between $1S$ and nP states, we can simplify the expressions for H_+ and H_- to give

$$H_\pm = \pm\frac{i\gamma_3\beta}{2}\left[\frac{\hbar^2}{2ma_{\text{exc}}^2}\left(1 - \frac{1}{n^2}\right) \mp \mu_B B\cos\theta\right] \times r_\pm(J_\mp J_z + J_zJ_\mp)K_z \quad (17)$$

when considering mixing with a given nP level. Here a_{exc} is the exciton Bohr radius.

The angular-momentum operators in one part of $H_{\gamma 3}$ will mix $1S$ states for which $m_J = +3/2$ with excited states for which $m_J = +1/2$ and the other part will mix $1S$ ($m_J = -3/2$) with excited states with $m_J = -1/2$, i.e., the $1S$ heavy-hole states will be mixed with excited states which are *light hole* in character.

The operators $r_\pm$ will mix hydrogenic $1S$ orbital functions with those of nP form, for example, $2P$ states, for which the orbital magnetic quantum number is $m_l = \pm 1$. Thus, to summarize, $H_{\gamma 3}$ will mix the $1S$ heavy-hole states which have $m_J = \pm 3/2$, $m_l = 0$ with light-hole nP states with $m_J = \pm 1/2$, $m_l = \pm 1$ and the same translational wave vector $K_z = N\pi/L$.

From perturbation theory, the energies of $1S$ ground states with $K_z = N\pi/L$ are changed by amounts,

$$\delta E_{\pm 3/2} = -\frac{|M_\pm|^2}{\Delta E_n},$$

where

$$|M_+|^2 = \left|\left\langle nP, m_J = +\frac{1}{2}, m_l = +1\right| H_+ \left|1S, m_J = +\frac{3}{2}\right\rangle\right|^2 K_z^2,$$

and

$$|M_-|^2 = \left|\left\langle nP, m_J = -\frac{1}{2}, m_l = -1\right| H_- \left|1S, m_J = -\frac{3}{2}\right\rangle\right|^2 K_z^2, \quad (18)$$

owing to mixing with the nP states. The energy denominator ΔE_n that appears above is given by

$$\Delta E_n = E_{nP,\text{LH},K_z} - E_{1S,\text{HH},K_z} = R\left(1 - \frac{1}{n^2}\right) + S + \hbar^2\left(\frac{1}{2M_{\text{LH}}} - \frac{1}{2M_{\text{HH}}}\right)K_z^2. \quad (19)$$

Here M_{LH} and M_{HH} are the translational masses of the light-hole and heavy-hole excitons and S is the strain-induced splitting between the light-hole and heavy-hole states. At small values of K_z, the denominator is dominated by the terms in R and S and is almost independent of wave vector, so that the changes in energies of the two states are then proportional to K_z^2. In this regime, the changes in energy (which are field dependent) can be expressed as field-dependent changes in the effective translational masses. At larger values of K_z, where the denominator in Eqs. (18) increases rapidly with wave vector, the energy changes are no longer proportional to K_z^2.

In the expressions for $\delta E_{\pm 3/2}$, there are two parts which involve the magnetic field. There are those proportional to B^2, which provide an increasingly negative contribution to the ground-state energy and therefore a negative contribution to the diamagnetism, which we discuss further in Sec. III E. There are then those which arise from the cross terms which are produced when the square bracket in Eq. (17) is squared. These give contributions to $\delta E_{\pm 3/2}$ which are linear in the magnetic field and which are of opposite signs for the $m_J = +3/2$ and $m_J = -3/2$ excitons (since the cross terms that arise from squaring the square bracket in the expressions for H_+ and H_- are, respectively, negative and positive). The dispersion curves for the $m_J = +3/2$ and $m_J = -3/2$ excitons will thus be perturbed differently, the difference being proportional to $B\cos\theta$: in other words, there will be a *contribution* to the g value of the exciton, with the observed angular dependence.

For a $1S$ exciton with translational quantum wave vector K_z, the contribution to the g value due to mixing with an nP state with the same translational wave vector is then given by

$$\delta g_1 = \frac{(\delta E_{3/2} - \delta E_{-3/2})}{\mu_B B} = \left(\frac{24\gamma_3^2\hbar^2\beta^2 \cos\theta}{m_0}\right)\left(\frac{v_n w_n}{\Delta E_n}\right)K_z^2, \tag{20}$$

where $v_n = -\langle nP, p_x|\partial/\partial x|1S\rangle a_{\rm exc}$ and $w_n = \langle nP, p_x|x|1S\rangle a_{\rm exc}^{-1}$. For $n=2$, $v_2 = 0.279$, and $w_2 = 0.745$.

Because of the mixing there will be, in addition to δg_1, a change in the g value of the exciton of the form

$$\delta g_2 = (g_{nP} - g_{1S})\sin^2\phi, \tag{21}$$

where to a good approximation

$$\tan 2\phi = 2M_+/\Delta E$$

evaluated at zero field and where g_{nP} $[= g_{\rm orb}\cos\theta - 2\kappa(\cos^2\theta + 2\sin^2\theta)^{1/2} - g_e]$ and g_{1S} $(= -6\kappa\cos\theta - g_e)$ are the g values of the $1S$ and nP states. Here the terms involving κ arise, respectively, from the LH and HH characters of the nP and $1S$ states and $g_{\rm orb}$ is an orbital contribution for the nP LH state of order $(2\gamma_1 + 5\gamma_2 - 2/m_e^*)$.

The total contribution to the exciton g value from mixing *with this particular* nP state is then given by

$$\delta g = \delta g_1 + \delta g_2. \tag{22}$$

We considered above the operators in the modified Luttinger Hamiltonian which involve γ_3. The terms which involve γ_1 and γ_2 also cause K_z dependent mixing between the $1S$ exciton state and higher lying nP states and this leads to K_z-dependent contributions to the *diamagnetic* shifts (see below). However, there are no cross terms of the form which arise in the mixing by the γ_3 operator (because there are no excited states which are connected to the ground state by *both* the internal momentum operator $\vec{p}$ and the distance operator $\vec{r}$), and hence *there are no* K_z-dependent contributions to Zeeman splitting from the γ_1 and γ_2 parts of the Hamiltonian.

For CdTe, the mixing produced by the γ_3 term with any particular nP state is small, so that contributions to the g value of the form given by Eq. (22) can be summed over all such states. For different values of K_z we have carried out the summation, using values of n from 2 to 10, by which time the summation has converged to within 1% (as found by selective calculations to greater accuracy). Mixing by $r_\pm$ cannot occur between $1S$ and nD, nF, etc., states because of the electric-dipole selection rules.

There is, however, mixing between the $1S$ state and p-symmetry continuum states which has to be taken into account. We have calculated the resulting contribution to the g value by integrating over these states. The contribution turns out to be about 40% of the total, with continuum states with energies up to 10 R required to give convergence to 1%. The result of the summation over discrete states and integration over continuum states gives the quantity $g(K_z)$.

To fit the experimental data for the specimens with strain of about 14 meV in the Faraday configuration ($\theta=0$), we have used the value of $g_{\rm HH} = -0.75$ as the point to which the data converge at $K_z = 0$. We have then used a value of $0.09m_0$ for the electron effective mass and the values of the Luttinger parameters given in Ref. 11 ($\gamma_1 = 4.7 \pm 0.3$, $\gamma_2 = 1.45 \pm 0.15$, and $\gamma_3 = 1.9 \pm 0.2$). As seen in the upper curve of Fig. 9, agreement with experiment is excellent. We shall see below (Sec. IV F) that similar values of the Luttinger parameters also account very well for the wave-vector dependence of the diamagnetic properties. The calculated values of $g_{\rm exc}$ do not depend on the choice of $a_{\rm exc}$ [which cancels out in Eq. (20)] and are only slightly dependent on the choice of γ_1 and γ_2. Since the contributions from δg_1 [Eq. (20)] are found to be much greater than those from δg_2 [Eq. (21)], the result is relatively insensitive to the value of $g_{\rm nP}$. The fraction f_0 of the ground state that remains of $1S$ character is approximately 0.96, which justifies the use of perturbation theory.

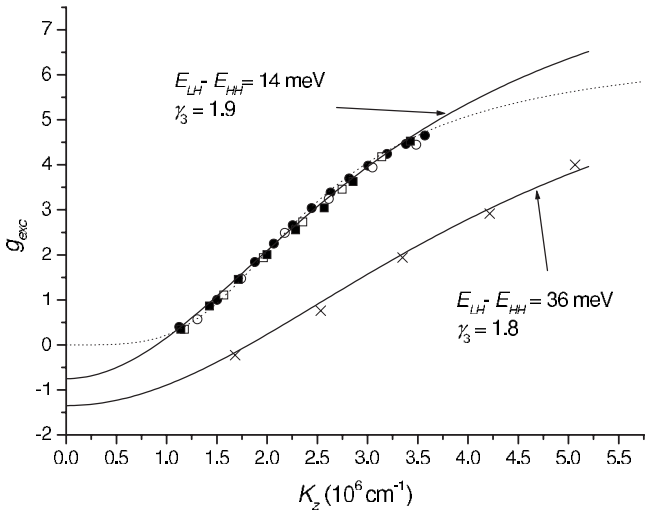

FIG. 9. The values of $g_{\rm exc}$ obtained from the model in the text (continuous curves) compared to two sets of the experimental data for (001) wells. The symbols have the same meaning as in Fig. 5. The upper sets of data points are for four wells with different widths, ranging from 800 to 1671 Å, but with approximately the same strain (an average of $S=14$ meV). The lower set of points is for the more highly strained specimen with $S \approx 36$ meV. The intercepts at $K_z=0$ are taken to be -0.75 and -1.25 (see text) and the curves are calculated with $\gamma_3=1.9$, $S=14$ meV (upper curve), and with $\gamma_3=1.8$ (lower curve). The broken curve is calculated from Eq. (27) based on the earlier model of Ref. 8.

In the case of the well with strain splitting of 36 meV, our best fit is obtained with $\gamma_3=1.8$ and $g_{\rm HH}=-1.35$. It is possible that these parameters are strain dependent, though more data are needed if this is to be confirmed.

For the Voigt configuration (where $\cos\theta=0$), the expected g value is $g_{\rm exc}^{\rm Voigt} = -g_e - 2\kappa\sin^2\phi$. Since κ is of order 0.17 and since, in this configuration, $\sin^2\phi$ is about 0.02, we expect an exciton g value which is essentially equal to the negative of the electron g value and which is independent of K_z. This is in agreement with the experimental observations (Fig. 7).

Finally, we note that the operator H_z [Eq. (16)] does not contribute to the exciton g values since it does not contain the internal momentum operator and therefore does not lead to the cross terms which are linear in magnetic field. However, it does lead to energy shifts proportional to B^2 and thus contributes to K_z-dependent changes in the diamagnetism (see Sec. IV F).

E. Analytic formula for the exciton g values

Since the range of variation of the denominator in Eq. (19) is small for those terms which contribute significantly to the mixing, we can approximate ΔE by

$$\Delta E = E_R + S + fK_z^2,$$

where

$$f = \hbar^2\left(\frac{1}{2M_{\mathrm{LH}}} - \frac{1}{2M_{\mathrm{HH}}}\right)$$

and where E_R is an averaged value (of order R) of the denominator. Summation now leads to an expression of the form

$$\sum \delta g_1 = C\frac{fK_z^2}{E_R + S + fK_z^2}\gamma_3^2, \tag{23}$$

where C is a fitting parameter. Curves of this form can, in fact, be drawn which are indistinguishable over the experimental range from the continuous lines plotted in Fig. 9 and provide a universal description of the K_z-dependent properties of the values of the exciton g values.

If γ_3 is constant, an increase in strain by a small amount δS shall cause a point with a given value of g_{exc} in Fig. 5 to be displaced by an amount δK_z given by

$$\frac{\delta K_z}{K_z} = \frac{\delta S}{2(E_R + S)}.$$

For specimens with $S=14$ meV and for $g_{\mathrm{exc}}=2.5$ (for which $K_z=2.3\times10^6$ cm^{-1}), this leads to $\delta K_z/\delta S \approx 5\times10^4$ cm^{-1}, in good agreement with the experimental result of Sec. III F.

F. Diamagnetic shifts in the (001) wells

We noted above that mixing by the γ_3 operator causes a K_z-dependent (negative) contribution to the energy of both $J_z=\pm3/2$ ground states which is proportional to the square of the magnetic field. The contribution to the diamagnetic constant when the magnetic field is along [001] (Faraday arrangement) is of the form

$$\delta D_0(K_z) = -\frac{3}{2}\left(\frac{\gamma_3\beta\hbar e a_{\mathrm{exc}}}{m_0}\right)^2\left(\frac{w_n^2}{\Delta E_n}\right)K_z^2 \tag{24}$$

for mixing with a particular excited nP state with the same translational wave vector. ΔE is given by Eq. (19). In a similar way to adding the contributions to the g value, the contributions can be summed and integrated over values of n and over the continuum states as described in Sec. IV D. The negative total contribution thus obtained is then combined with the (positive) diamagnetic parameter D_0 for zero K_z (we consider D_0 as an experimental parameter which we do not attempt to calculate) to give the upper continuous curve in Fig. 8. The Faraday data are accounted excellently with a value of the product $\gamma_3 a_{\mathrm{exc}}$ of 152 Å. The calculated curve can also be approximated very well with a negative K_z-dependent contribution to the diamagnetic parameter which has the same analytical form as Eq. (23).

In the Voigt configuration (magnetic field at 90° to the [001] direction) the situation becomes more complicated, since K_z-dependent mixing processes by operators which involve γ_1, γ_2, and γ_3 *all* become possible, with contributions to the diamagnetism which involve both light-hole and heavy-hole excited states. By similar arguments to those above, we obtain a K_z-dependent contribution of the form

$$\delta D_{90}(K_z) = -\frac{3}{4}\left(\frac{\gamma_3\beta\hbar e a_{\mathrm{exc}}}{m_0}\right)^2\left(\frac{w_n^2}{\Delta E_n}\right)K_z^2 - \left(\frac{(\gamma_1 - 2\gamma_2)\beta\hbar e a_{\mathrm{exc}}}{m_0}\right)^2\left(\frac{w_n^2}{\Delta E'}\right)K_z^2, \tag{25}$$

where ΔE_n is given by Eq. (19) and where

$$\Delta E_n' = E_{nP,\mathrm{HH}} - E_{1S,\mathrm{HH}} = R\left(1 - \frac{1}{n^2}\right). \tag{26}$$

There is no K_z dependence in $\Delta E'$ since the term in $(\gamma_1 - 2\gamma_2)$ mixes $1S$ heavy-hole exciton states with *heavy-hole* nP states. The contributions from Eq. (25) have, as before, to be summed and integrated over all nP and continuum states.

In fitting the data for the field at 90° to the growth axis, we have again taken the diamagnetic parameter at $K_z \to 0$ to be an experimental parameter (it needs not to be the same as for field directions along the growth axis, since the hole masses are anisotropic). The lower curve in Fig. 8 is calculated with $\gamma_3 a_{\mathrm{exc}}=152$ Å and with $(\gamma_1 - 2\gamma_2)a_{\mathrm{exc}}=100$ Å and reproduces the K_z dependence of the Voigt data very well. A reasonable choice of parameters which fit the data in Fig. 8 and which lie within the experimental accuracy of the values found by Dang *et al.*[11] is $\gamma_3=2.1$, $(\gamma_1 - 2\gamma_2)=1.4$, and $a_{\mathrm{exc}}=72$ Å. The value of γ_3 is close to that required to fit the exciton g values (Fig. 9) and a_{exc} is close the value of 64 Å expected from the binding energy of 11 meV if a dielectric constant of 10.3 is assumed.[29]

G. Magnetic properties of (110) quantum wells

In the case of (110) quantum wells we can define axes X, Y, and Z such that X is along $[1\bar{1}0]$, Y along [001], and Z is along [110] (the growth direction). For a well of width L, the center-of-mass exciton translational wave vector becomes quantized according to $K_Z = N^{110}\pi/L$. If the symmetry was purely axial about the Z axis and if the magnetic field was along this axis, then one would expect K_Z-dependent contributions to the exciton g value given by equations of the form of Eq. (20) but with γ_3^2 replaced by $\gamma_2\gamma_3$.[30]

The fact that no wave-vector dependence of the exciton g values is, in fact, observed for this quantum well is therefore surprising. One possibility is that the symmetry is indeed to a good approximation axial but that the splitting in energy between the $J_Z=\pm3/2$ and the $J_Z=\pm1/2$ states is very large [so that the denominator in the equivalent of Eq. (20) is large]. This explanation is unlikely, however, since there is a strong wave-vector dependence of the diamagnetic shift, similar to that for (001) quantum wells. A more likely possibility is therefore that the axial approximation is *not* appropriate and that the X and Y axes cannot be taken as being equivalent in our (strained) (110) quantum well. At present, we have insufficient information to solve this problem and

work on a further range of specimens with well-characterized strain distributions is necessary in order to understand the properties of (110) wells.

H. Mixing by higher order terms in the Hamiltonian

In our preliminary investigations[8] of the behavior of the g values we considered a model which involved only mixing between $1S$ heavy-hole exciton states and nP heavy-hole excited states. Mixing between such states is not caused by terms derived from the Hamiltonian of Eq. (11) but arises because of higher order terms whose form can be obtained from the theory of invariants.[31] For (001) quantum wells with the magnetic field along the growth axis, an operator was sought which mixes heavy-hole states in such a way as to produce K_z-dependent contributions to g_{exc}; the operator also needed to be such that it had no wave-vector-dependent effects on the g values when the magnetic field is applied along the growth direction of (110) quantum wells. The only term which satisfied these requirements was $\delta H = c\Sigma_i J_i^3 \kappa_i$, where c is a constant (often written as $a_2\gamma_v$), $\kappa_i = k_i(k_{i+1}^2 - k_{i-1}^2)$, and i refers in turn to each of the x, y, and z directions.[4,5] This term contains components of the form $-cK_z^2[J_-^3 p_+ + J_+^3 p_-]/8\hbar$, where $p_\pm = p_x \pm ip_y$, which, for (001) quantum wells, can mix the $1S$ ($m_J = \pm 3/2$) and nP ($m_l = \mp 1$) HH states. Only the mixing between $1S$ and $2P$ states was considered and the values of g_{exc} for the field along the growth direction of (001) quantum wells could be described by a universal curve of the form

$$g_{\mathrm{exc}} \approx \frac{1}{2} G\left(1 - \frac{1}{\sqrt{1 + \alpha^2 (a_{\mathrm{exc}} K_z)^4}}\right), \tag{27}$$

where G and α are adjustable parameters and g_0 is the value at $K_z = 0$. An example of such a curve is shown by the broken curve in Fig. 9, where g_0 is taken to be 0, rather than the values used in Sec. IV D.

In the model of Ref. 8, to account for the behavior of g_{exc}, the values required for c are 2.0 and 0.7 eV nm^3 for CdTe on $Cd_{0.96}Zn_{0.04}Te$ and for CdTe on $Cd_{0.89}Zn_{0.11}Te$, respectively, being concluded that in that model the c must be strongly strain dependent. Unfortunately, there are no published data which these values can be compared, so that an independent test of the model could not be provided.

The data presented in the present paper for both the Zeeman splittings and the diamagnetic shifts (together with their anisotropy) for a greater range of CdTe quantum wells have, however, enabled an alternative source of the mixing to be identified. Of particular importance has been the clear identification of the role of strain in changing the light-hole to heavy-hole splitting and thus altering the degree of mixing. This clearly points to the importance of excited states which are of light hole rather than heavy-hole character. The calculations in Sec. IV D show that the effect of altering the strain gives good agreement with experiment without the need to make drastic assumptions about the strain dependence of the parameter that controls the mixing (in that case, γ_3). Further, the mixing process caused by the γ_3 term leads to predictions for both the Zeeman splittings and the diamagnetic shifts that are in excellent quantitative agreement with experiments for values of γ_3 which are similar to those which have been published previously.

While contributions to the K_z-dependent magnetic properties which result from mixing due to high order terms such as δH cannot be excluded, we conclude that mixing which involves the Luttinger γ parameters provides a much more satisfactory and self-consistent description of the experimental data.

I. Other semiconductors

In Ref. 8 we showed (limited) data for the values of g_{exc} as functions of K_z for (001) quantum wells of ZnSe (500 Å) and of GaAs (2400 Å). The present calculation provides a good fit to the data for ZnSe if the published[32] value of γ_3 of 1.67 is chosen. For GaAs, the mixing with excited states appears to be too large for the simple perturbation approach used in Sec. IV to be adopted and fitting of these data will require a fuller calculation.

V. SUMMARY AND CONCLUSIONS

We have observed very large changes in the Zeeman splittings and in the diamagnetism of excitons as they acquire kinetic energy in wide quantum wells of CdTe (widths ranging from 660 to 1671 Å). The changes are found to be functions of the translational wave vector K_z, irrespective of the width of the well, and a model has been described which accounts both qualitatively and quantitatively for the properties observed in both Faraday and Voigt configurations and at intermediate directions of the magnetic field relative to the growth axis of the wells.

The model involves mixing between the hydrogenic $1S$ (heavy-hole) ground state of the exciton and the higher lying (light-hole) nP states and is ascribed to terms in the Luttinger Hamiltonian as extended to excitons in the center-of-mass or adiabatic approximation. Excellent quantitative agreement with all the experimental observations is provided with choices of the Luttinger parameters γ_1, γ_2, and γ_3 which are similar to those obtained from the literature.[32] All three parameters contribute to the K_z-dependent part of the diamagnetic shifts, since the corresponding terms in the Hamiltonian all lead to depression of the ground state by amounts which depend on the square of the magnetic field. In contrast, only the term involving γ_3 leads to K_z-dependent contributions to the exciton g values for (001) wells: the reason for this is that only this part of the Hamiltonian results in K_z-dependent matrix elements of the internal momentum operator $\vec{\mathbf{p}}$ and K_z-dependent matrix elements of the vector potential which both connect the $1S$ ground states with the *same* excited state. The changes in g_{exc} can be described by an analytic curve [Eq. (23)] which is indistinguishable from the full calculation.

For the heavy-hole exciton states which lie lowest in CdTe, the excited states which are mixed in by the γ_3 part of the extended Luttinger Hamiltonian are of light-hole nature, so that the magnetic properties become sensitive to the strain-induced splitting between the light and heavy holes. A further success of the model is that it leads to a satisfactory

quantitative explanation of the observed strain dependence.

One consequence of the analysis is that the changes in the g values of the exciton are due to the combined effect of the internal motion and the translational motion: the changes cannot therefore be ascribed to the electron or to the hole individually but to the exciton as an entity. Caution should therefore be exercised in simply using Eq. (3) to deduce hole g values from experimental determinations of the g values of electrons and excitons.

The model is applicable to wide quantum wells made from all zinc-blende semiconductors and confirms that the huge motion induced changes in magnetic properties, observed here for CdTe and previously[8] also for ZnSe and GaAs, should be universal for such materials.

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[1] J. O. Dimmock, in *Semiconductors and Semimetals*, edited by R. K. Willardson and A. C. Beer (Academic, New York, 1967), Vol. 3, p. 259.
[2] M. Altarelli and N. O. Lipari, Phys. Rev. B **7**, 3798 (1973).
[3] K. Cho, S. Suga, W. Deybrodt, and F. Willman, Phys. Rev. B **11**, 1512 (1975).
[4] E. L. Ivchenko and G. Pikus, *Superlattices and Other Microstructures* (Springer-Verlag, Berlin, 1995).
[5] E. L. Ivchenko, *Semiconductor Nanostructures* (Alpha Science International, Harrow, UK, 2005).
[6] Y. Merle d'Aubigné, Le Si Dang, A. Wasiela, F. d'Albo, and A. Million, J. Phys. (Paris), Colloq. **48**, 363 (1987).
[7] H. Tuffigo, R. T. Cox, N. Magnea, Y. Merle d'Aubigné, and A. Million, Phys. Rev. B **37**, 4310 (1988).
[8] J. J. Davies, D. Wolverson, V. P. Kochereshko, A. V. Platonov, R. T. Cox, J. Cibert, H. Mariette, C. Bodin, C. Gourgon, I. V. Ignatiev, E. V. Ubylvovk, Y. P. Efimov, and S. A. Eliseev, Phys. Rev. Lett. **97**, 187403 (2006).
[9] H. Mariette, F. Dal'bo, N. Magnea, G. Lentz, and H. Tuffigo, Phys. Rev. B **38**, 12443 (1988).
[10] J. M. Luttinger, Phys. Rev. **102**, 1030 (1956).
[11] Le Si Dang, G. Neu, and R. Romestain, Solid State Commun. **44**, 1187 (1982).
[12] V. A. Kiselev, B. S. Razbirin, and I. N. Uraltsev, Phys. Status Solidi B **72**, 161 (1975).
[13] M. V. Alekseenko and A. I. Veinger, Sov. Phys. Semicond. **8**, 143 (1974).
[14] M. Willatzen, M. Cardona, and N. E. Christensen, Phys. Rev. B **51** 17992 (1995).
[15] A. A. Sirenko, T. Ruf, M. Cardona, D. R. Yakovlev, W. Ossau, A. Waag, and G. Landwehr, Phys. Rev. B **56**, 2114 (1997).
[16] S. Tsoi, I. Miotkowski, S. Rodriguez, A. K. Ramdas, H. Alawadhi, and T. M. Pekarek, Phys. Rev. B **69**, 035209 (2004).
[17] W. Wardzyński and M. Suffczyński, Solid State Commun. **10**, 417 (1972).
[18] In Ref. 8 the sign of g_{exc} was not determined and g_{exc} was written as $g_e - g_{\mathrm{HH}}$. In the present paper we have determined the sign and use the definition in Eq. (3) so as to be consistent with other workers.
[19] R. T. Cox and J. J. Davies, Phys. Rev. B **34**, 8591 (1986).
[20] For a review of dilute magnetic semiconductors, see, e.g., J. Furdyna, J. Appl. Phys. **64**, R29 (1988).
[21] J. A. Gaj, R. R. Galazka, and G. Fishman, Solid State Commun. **29**, 435 (1979).
[22] D. G. Thomas and J. J. Hopfield, Phys. Rev. **124**, 657 (1961).
[23] E. F. Gross, B. P. Zakharchenya, and O. V. Konstantinov, Sov. Phys. Solid State **3**, 305 (1961).
[24] See, e.g., J. J. Davies, D. Wolverson, I. J. Griffin, O. Z. Karimov, C. L. Orange, D. Hommel, and M. Behringer, Phys. Rev. B **62**, 10329 (2000), and references therein.
[25] A. A. Kiselev, E. L. Ivchenko, A. A. Sirenko, T. Ruf, M. Cardona, D. R. Yakovlev, W. Ossau, A. Waag, and G. Landwehr, J. Cryst. Growth **184/185**, 831 (1998).
[26] Q. X. Zhao, M. Oestrich, and N. Magnea, Appl. Phys. Lett. **69**, 3704 (1996).
[27] Q. X. Zhao, N. Magnea, and M. Willander, J. Cryst. Growth **184/185**, 835 (1998).
[28] E. Molva and Le Si Dang, Phys. Rev. B **32**, 1156 (1985).
[29] J. Strzalkowski, S. Joshi, and C. B. Crevell, Appl. Phys. Lett. **28**, 350 (1976).
[30] This can be seen by transformation of the coordinates x, y, z to X, Y, Z or by using the transformation given by G. Fishman, Phys. Rev. B **52**, 11132 (1995).
[31] G. L. Bir and G. E. Pikus, *Symmetry and Strain-Induced Effects in Semiconductors* (Wiley, New York, 1974), Chap. IV.
[32] *Numerical Data and Functional Relationships in Science and Techology*, edited by O. Madelung, U. Rössler, and M. Schulz, Landolt-Börnstein, New Series, Group III, Vol. 41B (Springer-Verlag, Berlin, 1999).