Confinement effects in bulk samples derived from the Franz-Keldysh effect

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Differential electroabsorption spectra of thick $Ga_{0.47}In_{0.53}As$ epilayers demonstrate a significant distortion of the Franz-Keldysh effect if the sample is thinner than the mean free path. Such confinement effects are clearly visible in 100-nm-thick samples, where they reduce the range and amplitudes of Franz-Keldysh oscillations. Further oscillations appear well above the gap which are related to quantum-confined states. Additional sharp spectral features are observed near the gap which are related to field-induced interface states. The spectra are compared with calculations based on the envelope approximation. [S0163-1829(99)03820-5]

The coherence length of excited states in bulk semiconductors can be derived from the range of Franz-Keldysh oscillations observed in differential electroabsorption spectroscopy.[1] This technique measures the change of transmittance induced by a small modulation of the field in the sample. The spectral range δE of Franz-Keldysh oscillations increases in proportion to the field F: $\delta E = eFL$, where L is the coherence length of excited states equivalent to the mean free path for coherent motion. The coherence length of $Ga_xIn_{1-x}As_yP_{1-y}$ epilayers varies at 20 K from 160 nm in the ternary alloy down to 60 nm for some quaternary compositions.[2] Similar spectra with numerous oscillations are also observed in the photoreflectance of GaAs heterostructures, where partial screening of the built-in field by photo-excited carriers provides comparable modulation conditions.[3,4] An increased range of oscillations has been used as an argument for improved sample quality after surface treatment,[5] assuming that this range depends only on the coherent lifetime of an excited state. This reasoning, however, is not justified if the sample thickness d is less than the mean free path L, because the barriers limit the energy an accelerated electron hole pair can gain in the field to the smaller value eFd. In the basis of eigenstates, the boundaries modify the free carrier states to quantized states of a broad quantum well.

We investigated electroabsorption spectra of 1-μm-thick $InP/In_{0.53}Ga_{0.47}As/InP$ heterostructures, with the thicknesses of the well material chosen as 600, 200, 100, and 60 nm, to explore the confinement-induced effects on the spectral lineshape of the Franz-Keldysh effect. The samples were grown by metal-organic vapor phase epitaxy (MOVPE) using the less hazardous liquid compounds tertiary butyl As (TBAS) and tertiary butyl P (TBP) as substitutes for the highly toxic hydrate gases AsH_3 and PH_3.[6] The samples were mounted on the tip of a cryostat and studied at 20 K. Electric fields were applied by dc bias between the n-doped substrate and a 6-nm-thin Pt contact on top, and altered by a square-wave modulation voltage $\Delta U = \pm 100$ mV of 1-kHz frequency. The relative change of the transmitted intensity I yields the change of the absorption constant: $d\Delta\alpha = -\Delta I/I$.

Figure 1 compares spectra of three samples at the same field of 32 kV/cm. The field is determined from the scaling of peak positions E_n with peak number n,[7] using the reduced mass $m^* = 0.037 m_o$ of heavy holes which, due to their larger oscillator strength, dominate the spectra:

$$(E_n - E_g)^{3/2} = \frac{3eh}{8}\sqrt{\frac{F^2}{m^*}}n \tag{1}$$

A different bias is needed to create the same field in each sample due to partial screening of the applied voltage by space charge in the contact region. The modulation field is estimated to about ± 0.5 kV/cm, but, due to screening effects, it can slightly vary with bias voltage and for different samples. The field is sufficiently large to ionize excitons completely. Weak-modulation spectra are then fully described by the envelope functions of electrons and holes because the Coulomb enhancement is not sensitive to small variation of the field.[8] There is no difference in the spectra of 600- and 200-nm-thick samples or to spectra measured on 600-nm-thick samples prepared by conventional sources.[1] Franz-Keldysh oscillations extend beyond the onset of transitions from the split-off band at 1.17 eV, and overlap with the Franz-Keldysh effect at that gap. The beat in the oscillations results from the interference of the dominant heavy-hole and weaker light-hole transitions which have different periods due to their different reduced mass.[1,9] The arrow marks the last oscillation which strictly obeys the scaling law of Eq. (1). This yields a mean free path of 150 nm which is

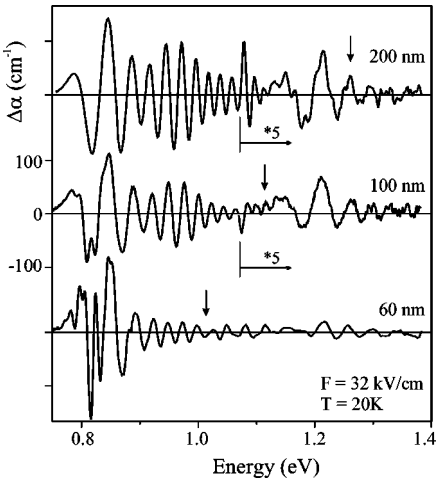

FIG. 1. Differential electroabsorption spectra for samples with different thickness at the same electric field of 32 kV/cm. The modulation voltage is ± 0.1 V. Arrows mark the range of Franz-Keldysh oscillations. Parts of the spectra are enlarged as indicated.

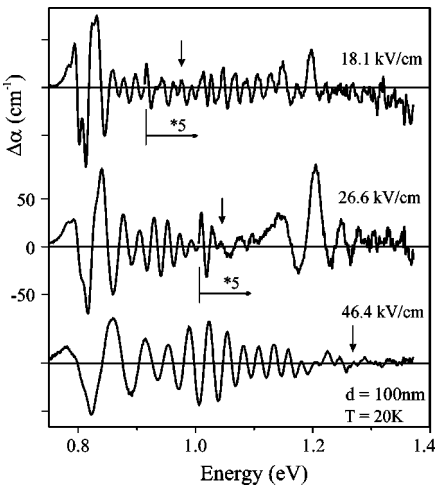

FIG. 2. EA spectra of the 100-nm-thick sample for different electric fields. The modulation voltage is ± 0.1 V.

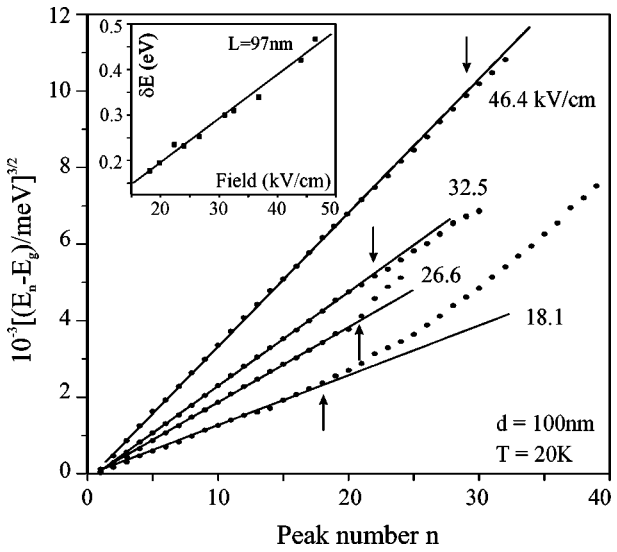

FIG. 3. Peak positions E_n of Franz-Keldysh oscillations vs peak number n for the 100-nm-thick sample. The inset shows the linear increase with field of the range of Franz-Keldysh oscillations indicated by the arrows.

confirmed by measurements at a smaller field, where the oscillations do not overlap with the response of the split-off band.

Despite the same field, spectra of thinner samples show significant differences, obvious by a direct comparison for a sample thickness of 100 nm. Beats of heavy- and light-hole spectra are still present, but the oscillations are more rapidly damped and disappear before they reach the gap of the split-off band. The arrow at 1.11 eV again denotes the energy where peak positions begin to deviate from the prediction by Eq. (1). Even more obvious changes occur near the gap at 0.81 eV, where the first peaks develop a fine structure. Such a fine structure at the gap is even more pronounced in the spectrum of the 60-nm-thick sample. Averaging over this fine structure leads to peaks which, below 1 eV, line up according to Eq. (1). However, the oscillations are rapidly damped and the beats of heavy and light holes are hardly visible. The oscillations observed above 1 eV do not obey Eq. (1), and therefore are not part of the Franz-Keldysh effect.

The spectrum of the thickest sample is the only one which shows an undistorted Franz-Keldysh effect corresponding to coherent acceleration limited by scattering. The modifications in the spectra of the thinner samples are attributed to the barriers which reflect carriers before collisions destroy the coherent motion. The boundaries alter the free-particle states into confined states of quantum wells. The energy range above the gap where oscillations line up as predicted for the Franz-Keldysh effect is always close to $\delta E = eFd$, the field-induced potential across a sample of thickness d.

The effect of boundaries is most evident at low field, as demonstrated in Fig. 2 by spectra of the 100-nm-thick sample. Except for a minor distortion near the gap, the spectrum at 46 kV/cm below 1.25 eV shows no deviation from the Franz-Keldysh effect. At lower fields, however, features attributed to carrier confinement become evident: the fine structure of the leading peaks, and deviation of the proper alignment of the oscillations at higher energy. At 18 kV/cm only a few of the numerous oscillations can be attributed to the Franz-Keldysh effect.

The range of the Franz-Keldysh oscillations is obtained from plots of peak position versus peak number, shown for the 100-nm-thick sample in Fig. 3. At 46 kV/cm, almost all peaks line up according to Eq. (1) and are therefore attributed to the Franz-Keldysh effect. Deviations become noticeable near peak number 30 located about 450 meV above the gap. Although an impressive range for coherent acceleration, this value remains far below the 700 meV anticipated for a mean free path of 150 nm. The energy where deviation from perfect alignment is evident decreases with decreasing field accompanied by an increasingly pronounced fine structure of the peaks near the gap. Replacing this fine structure by envelope peaks, we find that for 18 kV/cm still more than 15 peaks, reaching to about 1 eV, obey Eq. (1), and thus are attributed to Franz-Keldysh oscillation. The inset in Fig. 3 shows that the range δE of peaks which belong to the Franz-Keldysh effect increases linearly with field. Since δE is the energy carriers acquire in the electric field, the linear relation reveals that the mean free path, not the time between collisions, is independent of field and carrier energy.[1] The slope of the straight line, the ratio $\delta E/F$, leads to $L \sim d$, as expected for coherent motion restricted to the sample thickness d.

A deeper insight into the role of boundaries is obtained from tunnel resonance calculations,[10] which yield the envelope functions of eigenstates in these thick but finite samples. Figure 4 shows valence- and conduction-band edges of the heterostructure for the 100-nm-thick sample at 18 kV/cm. Wave functions above the barriers do not contribute to the electroabsorption spectra, but within the well two different types of wave functions exist whose subtle differences are responsible for the different types of oscillations observed in the experimental spectrum below E_b, the gap of barriers material. Since these states are confined, the energy spectrum consists of discrete but closely spaced levels. These states behave like a continuum as long as the level spacing is smaller than their lifetime broadening and the field is sufficiently strong. By tilting the potential, the field produces two spectral regions involving distinctly different eigenstates. In the first region extending from the gap E_g to $E_1 = E_g + eFd$,

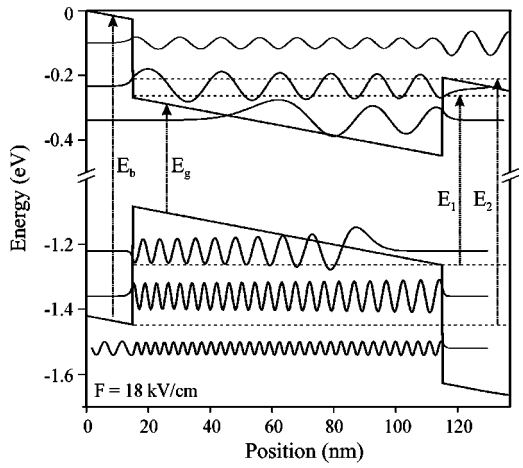

FIG. 4. Electron and hole envelope functions at 18 kV/cm of in a 100-nm-wide $Ga_xIn_{1-x}As$ well with InP barriers. Field and barrier height determine spectral ranges involving different types of wave functions.

all excited states involve electron and hole envelopes which reach to the bottom of the respective bands and extend over most of the sample. Except for their cutoff by the step potential of the barrier, they are all identical and correspond to eigenstates of a particle in a constant field, the Ai functions, which describe the Franz-Keldysh effect in bulk material. The different periods of electrons and hole states are due only to their different mass. The cutoff by the barriers has little influence as long as the wave functions extend over most of the structure, because normalization keeps their amplitude at the barrier small. Wave functions and electroabsorption spectra therefore become indistinguishable from those of bulk samples up to E_1.

The situation is different for the lowest electron and hole states. Confined by the field to the corner of their potential wells, they split away from the extended states. These strongly confined states which for electrons and holes occur at opposite interfaces have substantial transition probabilities only to Ai functions which just reach into their corners, separated in energy by the gap. Transitions involving these states bound to the interface cause the sharp features in the spectra near E_g. Since the transition probability to the rapidly oscillating wave function at higher energy is much smaller, the fine structure in the spectra induced by these interface states is found only near the gap.

Above E_1 and below $E_2=E_b-eFd$, optical transitions involve at least one state which does not reach to the bottom of the potential well. The periods of these wave functions are determined by the width of the potential well rather than by the field. These states correspond to quantum-confined states of large quantum numbers which are modified by the Stark effect. Consequently, the sequence of peaks in the spectra gradually deviates from that of Franz-Keldysh oscillations, as such states begin to contribute to the absorption spectrum. The range of quantum-confined states shrinks with increasing field, and vanishes for large fields when E_2 becomes smaller than E_1. At such a high field the Franz-Keldysh effect determines the spectrum completely, and the oscillations reach up to E_1 as long as the states remain confined to the well.

In conclusion, we have shown that boundary conditions modify the Franz-Keldysh oscillations. Their spectral range corresponds to the mean free path for coherent motion only in samples of sufficient thickness. This critical thickness is 150 nm at 20 K for the ternary alloy $Ga_xIn_{1-x}As$, and even larger values are anticipated for binary compounds like GaAs. In the case of thinner samples, fields and boundaries create different types of wave functions which can be distinguished by their contribution to the electroabsorption spectrum. States which reach to the bottom of the tilted valence and conduction bands produce Franz-Keldysh oscillations as in bulk material over an energy range which, however, is limited by the thickness of the sample. A second type of oscillation at higher energy involves rapidly oscillating quantum-confined states which respond to the field by the quantum-confined Stark effect. Although this may result in many oscillations in an electroabsorption spectrum, the distribution of peaks differs from the lineup of Franz-Keldysh oscillations, which allows one to separate these contributions. The most obvious effect of the barrier is the appearance of very narrow spectral features which seem to split the first peaks of the Franz-Keldysh effect. This fine structure results from the excitation of electrons or holes which are confined by the field to opposite interfaces of the sample. These states can be considered as field-induced interface states, which are best seen at lower field. It is interesting to note that these highly structured spectra are obtained on samples grown with alternative metal-organic sources, which obviously have reached the same quality as samples obtained with conventional but highly poisonous hydrate gases.

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