# Hot luminescence and Landau-level fine structure in bulk GaAs

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Additional peaks in the luminescence spectra of GaAs in a magnetic field in the region around the one-LO-phonon Raman line are reported. Experiments were performed under conditions close to resonance with interband magneto-optical transitions between Landau levels of low electronic quantum number $n$ ($n \leq 3$) for magnetic fields ranging up to 11 T. From the magnetic-field dependence of these features, and the fact that for different excitation energies they occur at the same spectral positions on an absolute scale as long as the magnetic field is kept constant, we argue that they are due to hot luminescence from the recombination of the free excitons associated with Landau levels. Magneto-Raman profiles (Raman intensity versus magnetic field) at the one-LO-phonon position were also measured. Fan plots obtained with the two methods coincide and show a characteristic fine structure of fan lines. The components of this multitude of features have separations that are about five times smaller than those of the dominant transitions between heavy-mass levels and electron states, which were considered in previous works on one-phonon magneto-Raman scattering. Since the same transitions between Landau levels are observed in both experiments, the additional features reflect the electronic structure of the magnetically quantized system. A theoretical interpretation of the fine structure is obtained when admixed valence-band states are taken into account.

## I. INTRODUCTION

Resonant Raman scattering has been used recently as a rather sensitive technique to study Landau levels in semiconductors.[1–3] In Raman scattering, oscillations of the LO-phonon intensity with magnetic field have been observed. Magneto-Raman profiles, i.e., the Raman intensity at the LO-phonon frequency as a function of the magnetic field, exhibit peaks when either the energy of incident laser photons or that of the scattered lights is in resonance with transitions between Landau levels. In such experiments resonant Raman scattering is used as a kind of modulation spectroscopy,[4] where an internal modulation is applied to the system by the phonon mode. Using this technique various aspects of electronic structure and electron-phonon interactions in a magnetic field have been studied, such as the nonparabolicity of the conduction band,[1,5] admixed valence levels,[2,3,6,7] resonant magnetopolarons,[2] or double resonances.[3,6,8,9]

When Raman spectra of GaAs are excited in a magnetic field with laser energies in the region just above the $E_0$ fundamental gap, additional peaks around the one-LO-phonon line are observed.[10] The spectral positions of these peaks, which occur with strengths up to that of the LO phonon, depend on the magnetic field. In this work we present a careful analysis of these features under different experimental conditions. Section II introduces the experimental procedure. Results from the systematic investigation of the additional lines in various scattering configurations with circularly polarized light and for different excitations energies are presented and discussed in Sec. III. Fan plots from magneto-Raman profiles, recorded at the one-LO-phonon line, are also given since they contain important subsidiary information for the interpretation of the magnetic-field dependence of the additional peaks in their relation to the electronic structure of the material. The investigation is summarized in Sec. IV.

## II. EXPERIMENT

The material studied in this work is $n$-type GaAs grown by liquid-phase epitaxy on a [001] GaAs substrate, the same sample as that used in our previous experiments. The mobility of the epitaxial layer is $\mu \simeq 10^5$ $cm^2/V^{-1}s^{-1}$ and the compensated carrier concentration $n = 7\times 10^{12}$ $cm^{-3}$ at 77 K. Raman experiments were performed with the sample at a temperature of about 10 K in backscattering geometry with the magnetic field applied perpendicular to the surface (Faraday configuration). Stokes Raman spectra were recorded in four scattering configurations with circularly polarized light: $\bar{z}(\sigma^{\pm},\sigma^{\mp})z$ and $\bar{z}(\sigma^{\pm},\sigma^{\pm})z$, where $\bar{z}$ and $z$ indicate the directions of incident and scattered photons, respectively. Incident light came from a Ti:sapphire laser pumped by an $Ar^+$-ion laser. The scattered light was dispersed with a double monochromator and detected with a cooled RCA C31034A GaAs photomultiplier tube using conventional photon-counting electronics. The Raman spectra were measured for two exciting laser energies of 1.580 and 1.605 eV with magnetic fields ranging from 5 to 11 T. Magneto-Raman profiles were recorded with the double monochromator set at the Stokes one-LO-phonon line. While scanning the magnetic field from 5 to 11 T the energy $\hbar\omega_L$ of the incident photons was kept fixed. The spectrometer slits were opened to give a spectral bandpass of about 10 $cm^{-1}$. Thus the signal retrieved can be regarded as a measure for the Raman scattering efficiency.

## III. RESULTS AND DISCUSSION

Typical Raman and luminescence spectra for various magnetic fields and for two different laser energies, $\hbar\omega_L = 1.580$ and 1.605 eV, are shown in Fig. 1. They were taken in a configuration with complementary circular polarizations, $\bar{z}(\sigma^+,\sigma^-)z$. Both sets of spectra show the one-LO-phonon line and also a series of additional features. The one-LO-phonon peak stays constant relative to the laser energy and its intensity exhibits the characteristic oscillations with the magnetic field which were extensively studied before.[2,3] These intensity oscillations can be assigned to resonances of incoming or scattered (outgoing) photons with transitions between conduction and valence Landau levels. The additional peaks have a characteristic dependence on magnetic field and laser energy. They move towards lower frequency shifts (higher absolute energies) when the magnetic field is increased. On an absolute photon-energy scale the positions of the additional peaks remain constant when different exciting laser energies are used at the same magnetic field. To facilitate the comparison, such scales are given at the top of Figs. 1(a) and 1(b). The relative intensities of the additional features depend on the magnetic field and also on laser energy. In the spectra obtained for other circular polarizations similar peaks appear which show the same behavior.[10]

When it comes to the observation of additional features in the Raman spectrum of GaAs one might first expect them to reflect van Hove singularities of the two-phonon density of states. Such effects have been demonstrated recently in GaAs under uniaxial stress, where, in a situation close to double resonance, additional peaks appear which could be attributed to scattering from combinations of acoustic phonons.[11] Raman scattering in GaAs around the one-LO-phonon line for zero magnetic field has been studied by Trommer and Cardona.[12] A series of peaks involving scattering by two transverse acoustic

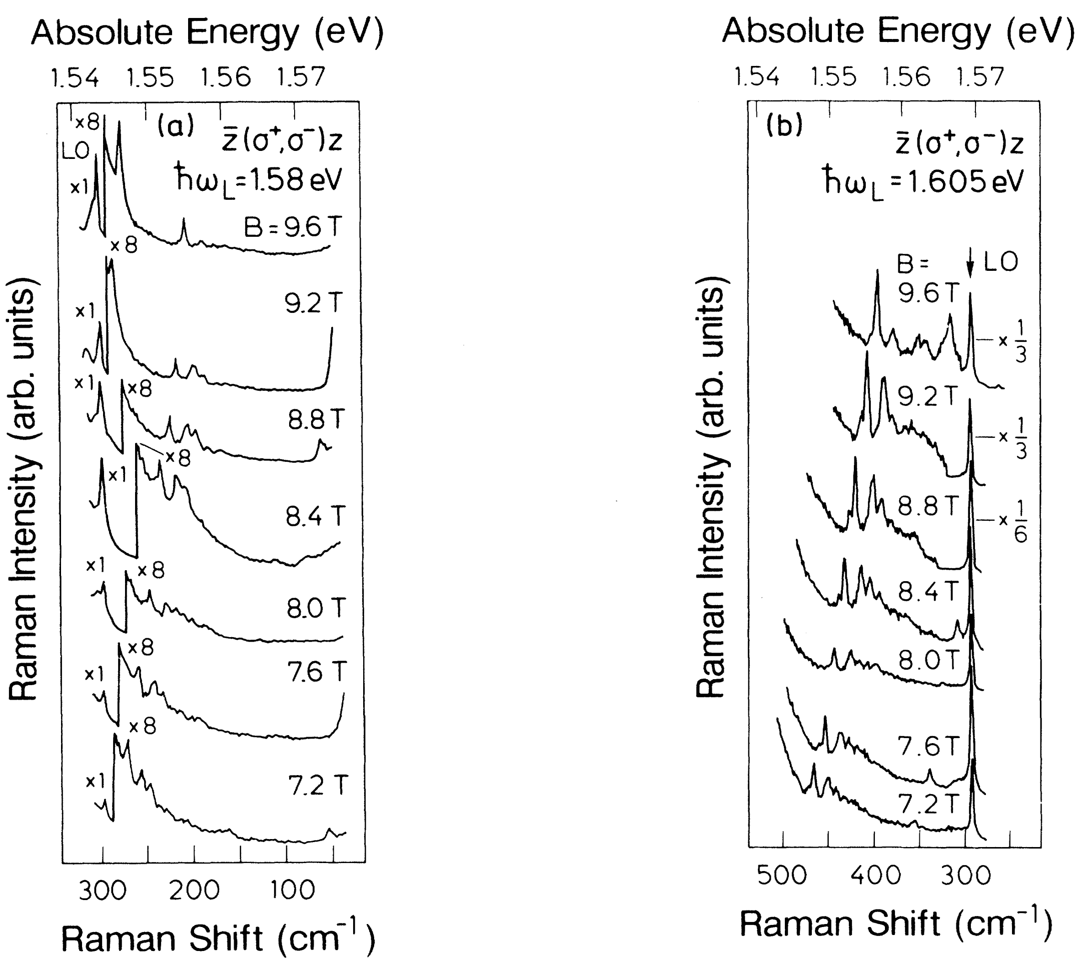


FIG. 1. Typical luminescence-Raman spectra of GaAs(001) at 10 K for different magnetic fields in the $\bar{z}(\sigma^+,\sigma^-)z$ configuration. The spectra were recorded with laser energies of (a) $\hbar\omega_L = 1.580$ eV and (b) $\hbar\omega_L = 1.605$ eV. The multiplication factors indicate that the particular parts of the spectra were multiplied by them prior to plotting.

phonons at different critical points of the Brillouin zone ($L$, $X$, and $K$) have been observed about 150 $cm^{-1}$ off the laser line. Multiphonon peaks due to combinations of optical and acoustic phonons with energies above the one-LO-phonon frequency were also found. When a magnetic field is applied, the electronic structure of the system is changed in accordance with Landau quantization[13] but vibrational structure must remain unaffected. Therefore the multiphonon peaks mentioned above should stay fixed at their positions with respect to the laser line when the magnetic field is changed. The magnetic field may cause intensity oscillations of these peaks due to resonances with Landau-level transitions but it should not change their spectral positions. The additional peaks in our Raman spectra have a completely different behavior than that expected for structure due to multiphonon Raman scattering. They appear to move in accordance with Landau levels, i.e., they depend on the magnetic field. In view of these characteristics it is suggested that they originate from hot luminescence due to magneto-optical transitions between Landau levels.

In Fig. 2 fan plots of energy positions of the hot-

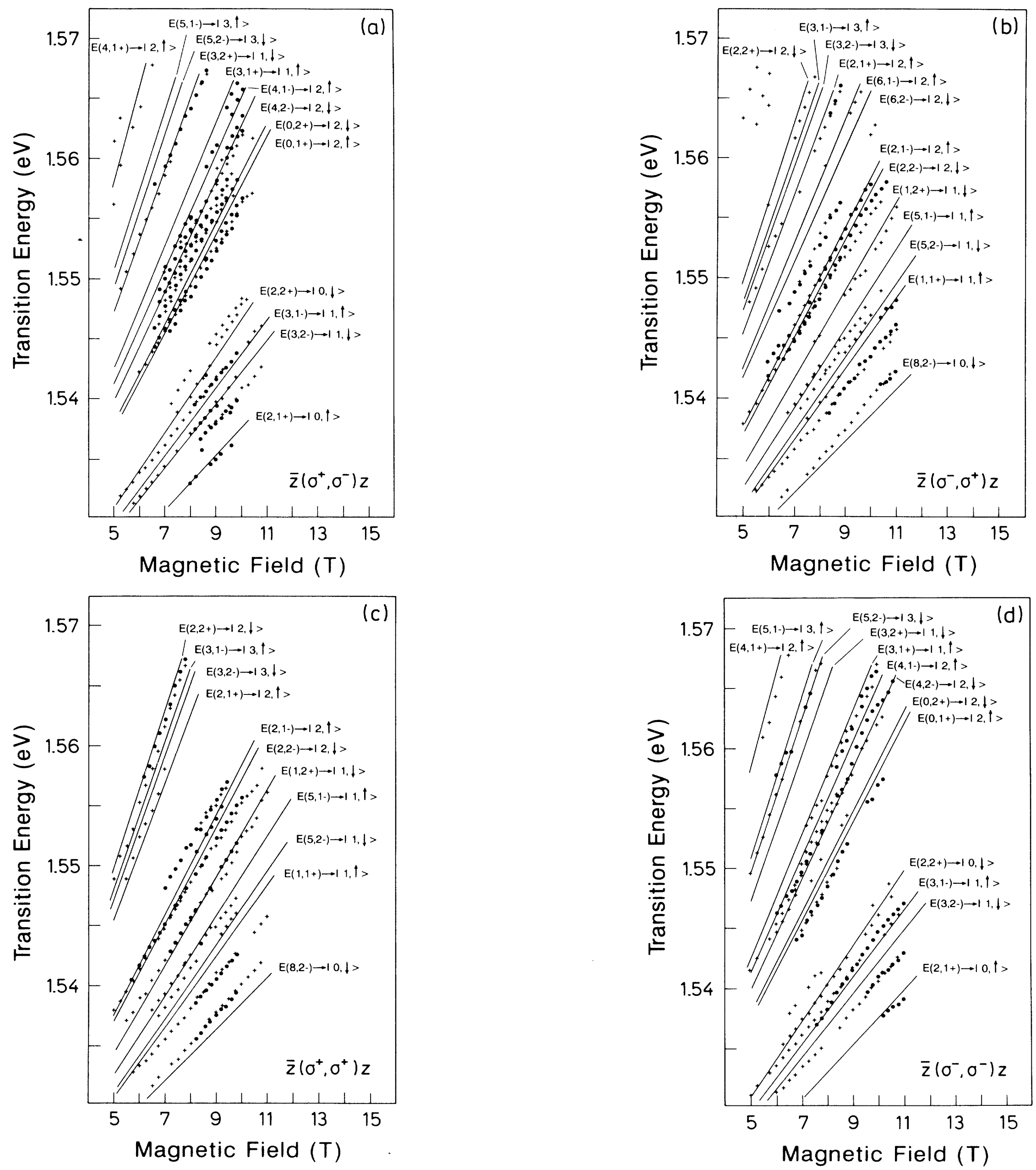


FIG. 2. Fan plots of peak positions of the hot luminescence for two different laser energies $\hbar\omega_L = 1.580$ eV (dots) and $\hbar\omega_L = 1.605$ eV (crosses). Solid lines are calculated Landau-level transition energies. The horizontal arrows point from the final state in the luminescence process to the corresponding initial state. The fan plots were measured in four scattering configurations: (a) $\bar{z}(\sigma^+,\sigma^-)z$, (b) $\bar{z}(\sigma^-,\sigma^+)z$, (c) $\bar{z}(\sigma^+,\sigma^+)z$, and (d) $\bar{z}(\sigma^-,\sigma^-)z$.

luminescence peaks as a function of the magnetic field are shown for two laser energies in four different scattering configurations. Data from excitation with $\hbar\omega_L = 1.580$ eV photons are given by dots, those taken with a laser energy of $\hbar\omega_L = 1.605$ eV are marked by crosses. The coincidence of fan lines measured with two different excitation energies can be readily observed. The solid lines represent theoretical transitions between Landau levels and will be discussed below. We note that all the lines converge towards $1.520 \pm 0.002$ eV for zero magnetic field, which is the energy of the $E_0$ fundamental gap of GaAs at low temperature [1.519 eV at 2 K (Ref. 14)]. We find that the intensities of the luminescence features increase linearly with the laser power. This is a strong indication that this "geminate" recombination involves correlated electron-hole pairs (excitons) which are associated with Landau levels rather than free electrons and holes.[15] The importance of Coulomb effects on the energies of interband transitions has been realized early[16] and a variational approach to evaluate the exciton ground-state energy, depending on the magnetic field and the Landau quantum number $n$, has recently been introduced and applied to interpret magneto-Raman data in the limit of high magnetic fields.[2] As will be illustrated in detail below, special attention has to be paid to these effects in our experimental range, since excitonic corrections are largest for transitions with small Landau quantum numbers $n$. Hot luminescence in GaAs without a magnetic field has been studied by many authors. Most efforts have been directed towards the understanding of hot electron relaxation in $p$-type bulk GaAs, where effects like conduction-band nonparabolicity and intervalley scattering can be studied.[15,17–19] In the presence of a magnetic field, investigations of hot magnetophotoluminescence were performed in GaAs/$Al_xGa_{1-x}$As multiple-quantum-well heterostructures.[20,21] There the hot luminescence due to exciton recombination associated with band-to-band transitions between conduction and valence Landau levels and also due to transitions between Landau levels and acceptor states has been observed. The filling of subbands with increasing excitation intensity and the saturation of the luminescence due to the finite population of the levels have been demonstrated.[21]

Our experimental finding of these additional features in the Raman spectra of bulk GaAs in a magnetic field, which for the above reasons may be attributed to hot luminescence between Landau levels, raises the question of identifying the observed fan lines and establishing their connection with the electronic structure of the system. An immediate though empirical way to do this is by comparison with results from magneto-Raman experiments such as those given in Ref. 3. This is a valid approach since the magneto-Raman resonances have been shown to be connected with Landau-level structure.[6] Therefore we have also measured magneto-Raman profiles at the one-LO-phonon line. Fan plots of peak positions for various laser energies (dots) are shown in Fig. 3 for the (a) $\bar{z}(\sigma^+,\sigma^-)z$ and (b) $\bar{z}(\sigma^+,\sigma^+)z$ configurations, respectively. The solid lines represent theoretical transitions which will be discussed below. All the fan lines converge towards the energy of $1.560 \pm 0.002$ eV for zero magnetic field. With $E_0 + \hbar\omega_{LO} = 1.556$ eV and an LO-phonon energy of $\hbar\omega_{LO} = 37$ meV, this implies that the fan lines are due to outgoing resonance, i.e., the energies of scattered Stokes photons match with interband magneto-optical transitions.[3] A comparison of the hot-luminescence fan lines of Fig. 2 with those obtained by magneto-Raman scattering (Fig. 3) shows that both experiments yield identical features in configurations with the same circular polarizations of the scattered photons, when the outgoing character of the magneto-Raman resonances is taken into account. The experimental fan lines of Fig. 3(a) compare

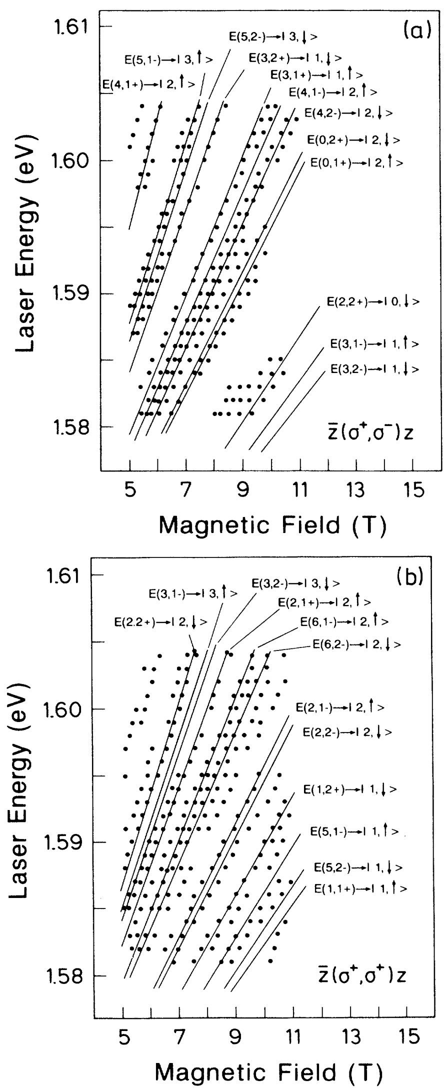


FIG. 3. Fan plots of laser energy vs magnetic field obtained from magneto-Raman profiles at the one-LO-phonon position in (a) $\bar{z}(\sigma^+,\sigma^-)z$ and (b) $\bar{z}(\sigma^+,\sigma^-)z$ configurations. The fan lines converge towards $E_0 + \hbar\omega_{LO}$ with the field approaches zero. Thus the resonances have outgoing character. Solid lines represent theoretical transitions between Landau levels. The horizontal arrows point from the final to the initial states in the luminescence process. The energy of one LO phonon was added to the calculated transition energies to account for the outgoing character of the resonances.

well with those of Figs. 2(a) and 2(d); Fig. 3(b) shows features which are also found in Figs. 2(b) and 2(c). From this we conclude that the hot-luminescence fan plots which are summarized in Fig. 2 do indeed reflect electronic structure from transitions between Landau levels. It is interesting to note, however, that the magneto-Raman fan plots of Fig. 3 show more lines and more detailed structure than the hot-luminescence fans in Fig. 2. This highlights the sensitivity of the magneto-Raman technique as a modulation spectroscopy type of experiment when compared to the more direct luminescence method.

The common origin of hot-luminescence features and magneto-Raman resonances suggests that an assignment of the fan lines in Figs. 2 and 3 to theoretical magneto-optical interband transitions should be possible following the scheme which has been successfully applied to interpret one-LO-phonon magneto-Raman data in InP and GaAs in previous investigations.[2,3] There Landau levels have been calculated using the theory of Trebin *et al.*,[22] who constructed an $8\times8$ $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian for zinc-blende-structure semiconductors by invariant expansion up to second order in $k$ in the space of the $J=\frac{1}{2}$, $\frac{3}{2}$, and $\frac{1}{2}$ manifolds, i.e., the $\Gamma_6^c$, $\Gamma_8^v$, and $\Gamma_7^s$ conduction, valence, and spin-orbit split-off bands. In such a scheme, which is based on the treatment of these bands as quasidegenerate, the energies of magneto-optical intraband and interband transitions can be predicted with rather high accuracy. It has consequently been applied to the interpretation of cyclotron resonance[23] as well as transmission,[24] reflectivity,[2,25] and magneto-Raman[2,3,5] experiments. Even though conduction Landau levels in GaAs can be calculated to a good approximation with the analytical expression based on a two-band model, which was used to determine the band nonparabolicity in Ref. 5, a treatment within the full theory is mandatory to determine energies and wave functions of the valence levels of heavy- and light-mass ladders. These levels show considerable mixing of the basis states and in some cases there are four or more admixtures with comparable weights. An approximate solution has been given earlier by Luttinger, who treated the $\Gamma_8^v$ manifold as decoupled into two $2\times2$ blocks.[26] That way he arrived at analytical expressions for the eigenvalues of the valence ladders. The wave functions consist of two admixtures which differ by 2 in the Landau quantum numbers of their oscillator parts. We write them as

$$E(n,1\pm)=a_1^{\pm}|n-2, J_z=+\tfrac{3}{2}\rangle+a_2^{\pm}|n, J_z=-\tfrac{1}{2}\rangle$$

and

$$E(n,2\pm)=b_1^{\pm}|n-2, J_z=+\tfrac{1}{2}\rangle+b_2^{\pm}|n,J_z=-\tfrac{3}{2}\rangle\ .$$

$E(n,1+)$ and $E(n,2+)$ represents light-mass ladders, $E(n,1-)$ and $E(n,2-)$ belong to valence levels of heavy mass. Outgoing resonances of one-LO-phonon magneto-Raman profiles in Ref. 3 were assigned to interband magneto-optical transitions between heavy-mass valence levels and conduction states and the wave functions $E(n,1-)$ and $E(n,2-)$ were used to systematically identify dominant admixtures within the heavily-mixed-valence levels obtained from the $8\times8$ $\mathbf{k}\cdot\mathbf{p}$ calculation. Theoretical considerations on resonant Raman scattering in a magnetic field which included all processes possible within the above wave functions and conduction states showed that indeed those transitions where $E(n,1-)$ and $E(n,2-)$ occur as dominant admixtures in the heavy-mass valence levels experience the strongest resonant enhancement and are thus observed in experimental magneto-Raman profiles.[6]

For these reasons the theoretical fan plots given in Figs. 2 and 3 of Ref. 3 should be sufficient to identify the fan lines of hot luminescence and magneto-Raman fan plots of this investigation. One realizes, however, that the fan plots given in Figs. 2 and 3 of this work exhibit far more structure beyond that which was attributed to the dominant transition between heavy-mass levels and electron states in Ref. 3. As far as the magneto-Raman fan plots of Fig. 3 are concerned, evidence for additional lines has been already found in Ref. 3 but it was not pursued further. The situation is particularly pronounced for the two scattering configurations given in Fig. 3, since under such conditions double resonances, which dominate the magneto-Raman profiles in the other two configurations, occur only at higher excitation energies and do not prohibit the observation of weak features by their strong scattering intensities [compare Figs. 3(a) and 3(b) with Figs. 2(a) and 3(a) of Ref. 3). The theoretical transitions in Figs. 2 and 3 which correspond to those of Ref. 3 are labeled by $E(n+2,2-)\rightarrow|n,\downarrow\rangle$ for the scattering configurations where $\sigma^-$ polarized photons are analyzed [compare Figs. 2(a), 2(d), and 3(a) with Fig. 2(a) of Ref. 3], with $n$ corresponding to the Landau index given in Fig. 2(a) of Ref. 3. The corresponding transitions for $\sigma^+$-polarized photons are labeled by $E(n,1-)\rightarrow|n,\uparrow\rangle$ in Figs. 2(b), 2(c), and 3(b). They can be found for the same Landau $n$ in Fig. 3(a) of Ref. 3. All these dominant transitions involve coupling between the $|n, J_z=\pm\frac{1}{2}\rangle$ admixtures of the heavy-mass valence levels and the $|n,\downarrow(\uparrow)\rangle$-conduction states. To understand the fine structure of fan lines which is found in Figs. 2 and 3 a more detailed analysis of interband magneto-optical transitions is necessary which also takes weaker transitions into account The first step towards this is to consider that the fan lines given in Figs. 2 and 3 of Ref. 3 are actually doublets of transitions with a separation which can be regarded as a $g$-factor splitting. This splitting, which amounts to the sum of electron and heavy-hole $g$ factors and is dominated for GaAs by the heavy-hole contribution,[6] was neglected in the fan lines of these figures because it could be considered as small compared to the experimental accuracy. With the experimental precision of the present investigations, however, such splittings can be distinguished and therefore the partner transitions are given: $E(n+2,2-)\rightarrow|n,\downarrow\rangle$ is close to $E(n+2,1-)\rightarrow|n,\uparrow\rangle$ in Figs. 2(a), 2(d), and 3(a). $E(n,1-)\rightarrow|n,\uparrow\rangle$ is next to $E(n,2-)\rightarrow|n,\downarrow\rangle$ in Figs. 2(b), 2(c), and 3(b). In Fig. 4 we show the diagram of band structure of GaAs at 10 T in order to understand the transitions observed in the experimental data presented in Figs. 2 and 3. The analysis of the oscillator strengths of interband transitions calculated with the

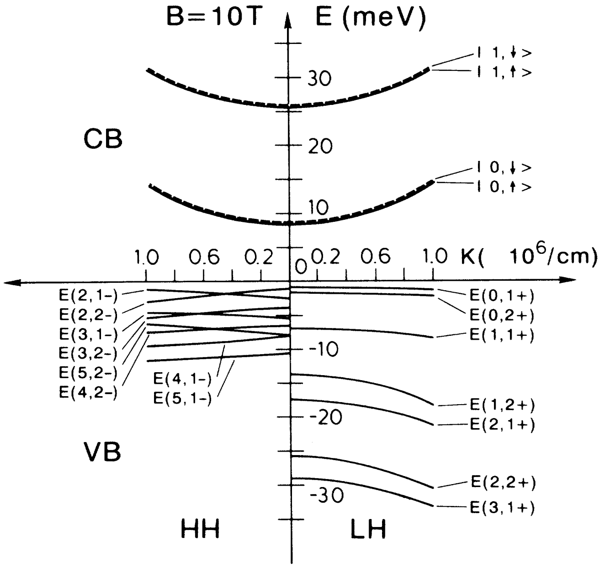


FIG. 4. Diagram of the Landau-level ladder energies vs wave vector $K$, along the direction of the magnetic field, at 10 T. We present only Landau levels relevant to this work. The positive energy axis corresponds to the conduction-band (CB) Landau levels while the negative one corresponds to valence-band (VB) levels of the heavy holes (HH) (left) and the light holes (LH) (right). The notation is explained in the text.

theory of Ref. 22 yields weaker transitions between levels of the light-mass ladders and electron states. Such transitions can also be assigned to experimental fan lines in Figs. 2 and 3. They are labeled by $E(n,1+)\rightarrow|n,\uparrow\rangle$ and $E(n,2+)\rightarrow|n,\downarrow\rangle$ for $\sigma^+$-polarized photons in Figs. 2(b), 2(c), and 3(b). For $\sigma^-$-polarized photons [Figs. 2(a), 2(d), and 3(a)] the $E(n+2,1+)\rightarrow|n,\uparrow\rangle$ and $E(n+2,2+)\rightarrow|n,\downarrow\rangle$ transitions are given. The irregular spacings of these transitions for small Landau quantum numbers illustrate the impact of quantum effects on the eigenenergies of light-mass valence levels[26,22] (see light-hole ladders in Fig. 4). In addition to the above transitions which can be found in a systematic way when the valence eigenvalues are investigated for dominant contributions in accordance with Luttinger's approximation, a few other fan lines are also given in Figs. 2 and 3. They are found to have comparable oscillator strengths but the weight coefficients of the coupling admixtures are only the third largest of the wave functions or smaller. Their valence parts were labeled according to the most dominant admixtures. With the consideration of all these transitions most of the fine structure of fan lines found in both hot-luminescence and magneto-Raman experiments can be described.

Since all the calculations of Landau levels were performed in a single-particle picture, special care has to be taken to include Coulomb effects. A variational approach has been introduced in Ref. 2 which allows the calculation of the ground-state binding energy of excitons associated with Landau levels in the limit of high magnetic fields, i.e., when the cyclotron energy is large compared with Coulomb corrections. This calculation yields the binding energy of the excitons as a function of magnetic field and Landau quantum number $n$ for a system of reduced effective mass $\mu$. When admixed valence levels are considered, the assignment of a single Landau $n$ and a mass $\mu$ to a particular valence level or exciton is no longer unambiguous. We therefore determined the effective mass directly from the magnetic-field dependence of the respective transition and chose as Landau $n$ that of the conduction level involved. This is justified by the $(\Delta n=0)$ selection rule for the dominant interband transitions[6] and the rather pure character of the conduction-Landau-level wave functions. The exciton binding energies obtained that way were subtracted from the theoretical transition energies mentioned above. All theoretical fan lines in Figs. 2 and 3 represent such corrected values. From Fig. 2 it can be realized that the hot-luminescence features are found in the region of energies close to the $E_0$ fundamental gap. Therefore interband transitions of small Landau quantum numbers $n$ are involved. Calculations show that for $n\leq 2$ the exciton binding energies exceed 4 meV in the range of magnetic fields investigated here. The contribution due to Coulomb effects is thus an important correction to the energies of these interband magneto-optical transitions, especially when the fine structure is considered. In fact, due to quantum effects for low-index valence levels, not including the influence of excitons will change not only the spectral positions but also the order of transitions and an assignment of theoretical fan lines to experimental features becomes impossible.

The mechanism which is responsible for the occurrence of band-to-band hot luminescence between Landau levels can be viewed as an intraband relaxation of excited carriers with the emission of phonons. The electrons and holes thus reach a quasiequilibrium state at the extrema of the Landau bands from which the recombination occurs. A microscopic picture for the case of multiple-LO-phonon cascades within Landau levels has been developed by Belitskii *et al.*[27] and expressions for the case of two-LO-phonon resonant Raman scattering have been given.[28,29] From this a theoretical model which treats optical and acoustic multiphonon emission in a magnetic field could be developed. Due to nonvanishing matrix elements of the Fröhlich electron-phonon interaction for couplings between Landau levels of different oscillator quantum number $n$ when the crystal momentum has finite values, interlevel scattering should also be considered. Time-resolved luminescence experiments will yield information on the carrier relaxation and multiphonon emission in a magnetic field and might thus be helpful to obtain a more detailed view of these processes.

## IV. CONCLUSIONS

Hot luminescence in a magnetic field occurs in bulk GaAs due to the recombination of free excitons associated with Landau levels. Fan plots of these transitions show the same features which are obtained from magneto-Raman profiles recorded at the one-LO-phonon position. A characteristic fine structure of transitions between Landau levels is observed by both methods. Theoretical interband magneto-optical transitions can be assigned to the experimental fan lines from hot luminescence and magneto-Raman scattering when the mixing of valence levels with heavy and light masses is taken into account.

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