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Polarization anisotropy of sub-band gap oscillatory features in contactless electroreflectance spectrum of $In_xGa_{1-x}P$ layers grown on GaAs (001) substrates

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We report the observation of strongly polarization sensitive sub-band gap oscillatory features in the contactless electroreflectance spectrum of $In_xGa_{1-x}P$ layers grown on GaAs (001) substrates. At a given energy in the sub-band gap region, the peak strength of these oscillatory features decreases from a positive maximum to a negative minimum passing through zero as the polarization of the incident probe beam is rotated by 90° from $[\bar{1}10]$ direction to $[110]$ direction in the (001) plane. The origin of this phenomenon is explained on the basis of optical interference coupled with linear electro-optic effect induced changes in the sub-band gap refractive index of the $In_xGa_{1-x}P$ layers. Numerical simulations based on the above mechanism are shown to reproduce the polarization dependent observations quite well. © *1998 American Institute of Physics.* [S0021-8979(98)04210-8]

I. INTRODUCTION

The study of polarization dependent optical properties of semiconductors give important information about their electronic band structure. In recent times, such measurements have been used to study the process of ordering in epitaxial $In_xGa_{1-x}P$ alloy layers on (001) GaAs substrates.[1] Photoluminescence[2] and electroreflectance[3] measurements on these samples have shown that the spectral features associated with the direct interband transition (DIT) change as the polarization of light is rotated from $[\bar{1}10]$ to $[110]$ direction in the (001) plane, in accordance with theoretical predictions.[4] This article deals with polarization dependence of the oscillatory features (OF) seen in the electroreflectance spectra of $In_xGa_{1-x}P$ samples, at energies lower than the fundamental DIT energy. It is well established that in the case of heterostructure samples, these OF arise in modulation spectroscopy experiments such as photoreflectance (PR) because of a combination of optical interference and sub-band gap refractive index modulation of the film or the substrate or both.[5] In PR the proposed physical mechanisms responsible for the refractive index modulation giving rise to the OF in the sub-band gap region are (i) electric field induced photon assisted sub-band gap tunneling through the Franz–Keldysh (F–K) effect,[5,6] (ii) pump beam induced band filling of the impurity tail states,[5,7] and as was shown recently (iii) pump beam induced periodic modulation of the epilayer temperature.[8] However two of the above mechanisms cannot be invoked in the present case because the absence of a pump beam in contactless electroreflectance spectroscopy (CER) rules out the band filling mechanism or temperature rise due to it. Furthermore, there is no resistive heating in the CER configuration. As such none of the above mechanisms, including the F–K effect, can account for the observed polarization anisotropy in the OF reported here. An understanding of the mechanism of the subgap refractive index modulation leading to the OF can help in separating out its distorting influence on the DIT lineshapes. In this article we provide an explanation for the observed polarization dependence of the OF on the basis of the linear electro-optic effect induced modulation of the sub-band gap refractive index of the epilayer and show that simulations based on this proposed modulation mechanism reproduce the experimental observations quite well.

II. EXPERIMENTAL DETAILS

The samples used in this study were undoped $In_xGa_{1-x}P$ layers grown using metal organic vapor phase epitaxy (MOVPE), on (001) GaAs substrates with a nonintentionally doped GaAs buffer in between. Polarization sensitive OF were observed in many samples out of which we present two cases. In sample A the $In_xGa_{1-x}P$ layer was 2 μm thick while in B it was 1 μm thick. In the contactless electroreflectance (CER)[9] measurements the samples were placed between two electrodes in a capacitorlike arrangement with the top electrode kept $\simeq$0.3 mm from the sample surface. A maximum of 3.5 kV (pp) sinusoidal voltage (330 Hz) was applied on the top transparent electrode (ZnO on glass) for the purpose of modulation.[10] The probe beam was obtained by dispersing light from a 150 W quartz tungsten halogen lamp using a 1/8 m monochromator with $\simeq$4 nm bandpass and detected using a silicon photodetector. Phase sensitive detection of the modulated reflectivity signal was performed using a lock-in amplifier. Polymer film polarizers were used to obtain the polarized probe beam with transmission for the orthogonal polarization being smaller than 1% in the wave-

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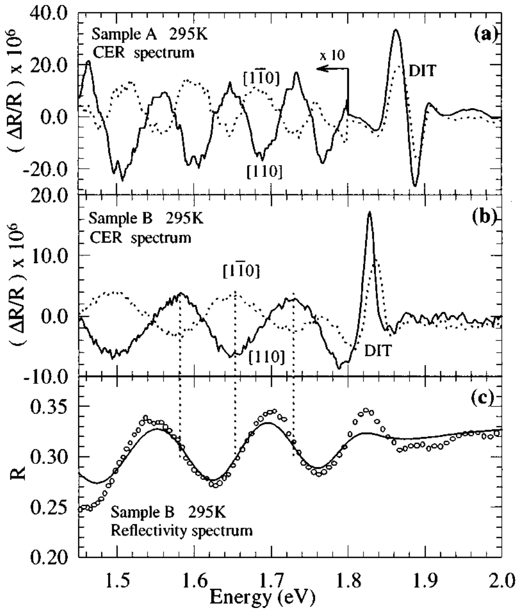

FIG. 1. Experimental CER spectra of samples A (a) and B (b) for two orthogonal polarizations of the probe beam along [110] and $[1\bar{1}0]$ crystallographic directions. (c) The measured (circles) and the fitted (line) reflectivity spectra of sample B. The vertical dotted lines show that the peaks of the oscillatory features in the CER spectrum coincide with the halfway points of the interference oscillations in the R spectrum.

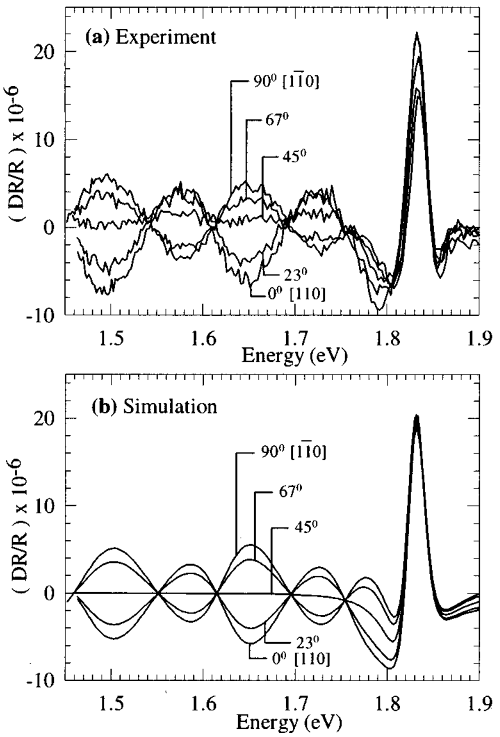

FIG. 2. (a) The experimental and (b) the simulated CER spectra of the sample B for different polarizations of the incident probe beam.

length range of interest. The reflectivty (R) spectrum was normalized using a standard aluminium coated mirror.

III. RESULTS AND DISCUSSION

Figures 1(a) and 1(b) show the CER spectra of the two samples A and B respectively with the incident probe light polarized along the [110] and $[1\bar{1}0]$ crystallographic directions. The structure in these spectra beyond 1.8 eV is due to the DIT at the fundamental gap of the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{P}$ alloy. The strength of the DIT differs for the [110] and the $[1\bar{1}0]$ polarizations due to ordering which is commonly observed in these materials.[3] However the most prominent difference between the two light polarizations along [110] and $[1\bar{1}0]$ is the ''phase rotation'' of the OF seen at energies below the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{P}$ band gap DIT. A more detailed study shows that the peak strength of these OF actually decreases gradually from a positive maximum to a negative minimum passing through zero as the polarization of the incident probe beam is rotated by 90° from $[1\bar{1}0]$ direction to [110] direction in the (001) plane. This is depicted in the CER spectra of the sample B in Fig. 2(a) for various polarizations of the incident beam with respect to the [110] crystallographic axis. Although such a phenomenon was apparent in the polarization dependent electroreflectance spectra of ordered $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{P}$ alloys reported by Kanata *et al.*,[3] these authors did not discuss it in their article probably because of its weak strength as compared to the main transitions at the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{P}$ band edge. Ordering does break the fourfold symmetry of the (001) plane of a crystal with ZnS structure and creates a difference in the refractive indices for light polarized along [110] and $[1\bar{1}0]$, however it cannot explain the above phenomenon especially since it occurs at energies far below the fundamental gap.

We note here that there is a considerable body of literature devoted to the study of ''spectral rotation'' of sub-band gap features in electroreflectance which has been reviewed by Pollak and Shen.[11] The spectral rotation in these cases refers to the energy shift, observed as a phase shift, of the sub-band gap features with change of the dc bias applied in addition to the modulating voltage. The strength of these features is strongly damped at energies lower than the fundamental gap. In these cases, interference effects due to refractive index modulation caused by (i) exciton quenching and (ii) F–K effect under inhomogenous fields have been put forth as the causes giving rise to this phenomenon. In our case, the spectral rotation with variation in the polarization of light is evidently quite different from the above mentioned phenomenon. The distinguishing features of the phenomenon observed by us are (i) for a given polarization the amplitude of the OF remains practically unchanged even upto 380 meV below the band gap, (ii) strong anisotropy between [110] and $[1\bar{1}0]$ directions resulting in a 180° phase reversal of the OF for light polarized along these two directions, (iii) a direct dependence of the amplitude of the OF on the angle between the principal orientations [110] or $[1\bar{1}0]$ and the polarization vector, and (iv) a sudden phase reversal by 180° as the polarization angle goes through 45°. We shall show here that all these aspects are related to the linear electro-optic effect in these structures.

The fact that the OF in the present case are related to optical interference is evident from the following two observations. First, the periods of the OF in CER match those in the plain *R* spectra which can be seen by comparing the CER spectrum of sample B in Fig. 1(b) and its *R* spectrum (circles) in Fig. 1(c). The OF in the *R* spectra arise because of interference between the light reflected from the $In_xGa_{1-x}P$ surface and the $In_xGa_{1-x}P$/GaAs interface. Second, comparing Figs. 1(a) and 1(b) we find that the period of the OF in the CER spectra scale with the thickness of the $In_xGa_{1-x}P$ layer in accordance with what is expected on the basis of optical interference. The OF in CER arise because the external modulating ac electric field modulates the refractive index of the $In_xGa_{1-x}P$ layer in phase with the modulating field. This modifies the optical path length of the beam reflected from the $In_xGa_{1-x}P$/GaAs interface which in turn affects the interference oscillations. The difference between the interference oscillations in the reflected signal with and without the external modulating field results in the OF in the CER spectra.

We suggest that the mechanism of sub-band gap refractive index modulation in the present case is the linear electro-optic effect. Due to the linear electro-optic effect, in a semiconductor alloy crystal having ZnS symmetry with an external electric field (F) applied along the (001) direction, the refractive index as seen by light incident on the (001) face polarized along $[\underline{1}10]$ direction is increased to $n=n_0+\delta n$ while for the $[1\bar{1}0]$ polarization it is decreased to $n=n_0-\delta n$ where n_0 is the refractive index in the absence of the field and

$$\delta n=\tfrac{1}{2}\, n_0^3 r_{41} F. \tag{1}$$

r_{41} being the linear electro-optic coefficient of the $In_xGa_{1-x}P$ epilayer.[12] This can explain the observed polarization anisotropy of the OF as follows. When the incident light is polarized along [110], the increased refractive index, due to the applied electric field, shifts the interference oscillations in the reflected light to one direction (towards lower energies) while for the $[1\bar{1}0]$ polarization, the decreased refractive index shifts them in the opposite direction (towards higher energies). As a result the difference signal between the reflected light under field off and field on conditions would have opposite sign for the two polarizations of the light. This is the origin of the observed polarization anisotropy in the OF. Also the fact that the OF in CER arise mainly due to shifts in the interference oscillations in the reflected signal rather than a change in their amplitude can be infered as follows. Comparing the CER and *R* spectra of sample B in Figs. 1(b) and 1(c), it is evident that the peak of the OF in CER coincide with the halfway points of the interference oscillations in the *R* spectrum (see the vertical dotted lines), a phenomenon which is also seen in our simulations to be discussed later. This can happen only if peak shifts were the main cause for the appearance of the OF in CER because OF dominated by change in amplitude of the interference oscillations would have resulted in the peaks in the CER and the *R* spectra occuring at the same energy. Finally the above mechanism also suggests that if both the polarizations [110] and $[1\bar{1}0]$ have equal intensity (i.e., when light is polarized at 45° to the [110] direction) then it is likely that the shifts in the interference oscillations for the two polarizations would tend to cancel out resulting in no OF in the CER spectrum. That this is indeed so is shown in Fig. 2(a) for light polarized at 45° to the [110] direction.

We note here that in the F–K theory, the external modulating electric field affects the dielectric function at the band gap and higher energies by modifying the transition probabilities of electrons from the valence band to the conduction band. In this theory, in the expression for change in the dielectric function, there does arise a term which is linear in the electric field in the case of noncentrosymmetric crystals but its effect is only to cause a shift (usually not measureable) in the critical point energy.[13] However there are reports in the literature of structures in the electroreflectance spectra (at energies higher than the band gap and strongly energy dependent), whose strength varies linearly with the applied electric field. Their origin has been explained on the basis of piezoelectric effect induced changes in the dielectric function.[14,15] The linear electro-optic effect invoked here has similar tensorial characteristics. The change in the dielectric function at sub-band gap energies due to the linear electro-optic effect arises from modification of the polarizability of the valence electrons (at optical frequencies) due to the externally applied modulating field.[16] In such a case the quantity $n_0^3 r_{41}$ has a very weak energy dependence in the sub-band gap region[12,16] therefore δn for all practical purposes is a constant in the 400 meV range below the $In_xGa_{1-x}P$ band gap considered here. Simulations performed by us show that the OF amplitude is proportional to the magnitude of δn, which explains the observed nearly energy independent amplitude of the OF in the CER spectra presented here. In the following sections we discuss the details of the simulations performed to reproduce the observed polarization dependence of the OF in the CER spectrum, based on the linear electro-optic effect.

For the purpose of simulations, in the energy range 1.45–1.9 eV, the reflectivity (R) spectrum of our structure was modeled by considering reflection in air (≡medium 1) from a dielectric film of thickness d ($In_xGa_{1-x}P$≡medium 2) on a semi-infinite dielectric substrate (GaAs≡medium 3). In the above energy range, the 300-μm-thick GaAs substrate has a finite absorption coefficient ($\alpha_{\mathrm{GaAs}}\simeq 2\times 10^4\ \mathrm{cm}^{-1}$), so it completely absorbs all the light penetrating it. This prevents any light from reaching the $In_xGa_{1-x}P$/GaAs interface after reflection from the back of the GaAs substrate and provides for the assumption of a semi-infinite substrate. However the imaginary part of the refractive index κ_{GaAs} ($\simeq 0.2$) is too small in comparison with its real part n_{GaAs} ($\simeq 3.7$). As a result κ_{GaAs} has negligible contribution to the reflectivity at the $In_xGa_{1-x}P$/GaAs interface so it is taken to be zero in our simulations. On the other hand, to account for the vanishing of the OF at energies higher than the band gap of the $In_xGa_{1-x}P$ film ($\simeq$1.83 eV), along with real part of the refractive index of the $In_xGa_{1-x}P$ film $n_{In_xGa_{1-x}P}$, it is necessary to consider a finite imaginary part of the refractive index $\kappa_{In_xGa_{1-x}P}$ of the $In_xGa_{1-x}P$ film at those energies. For such a model, the reflectivity is given by

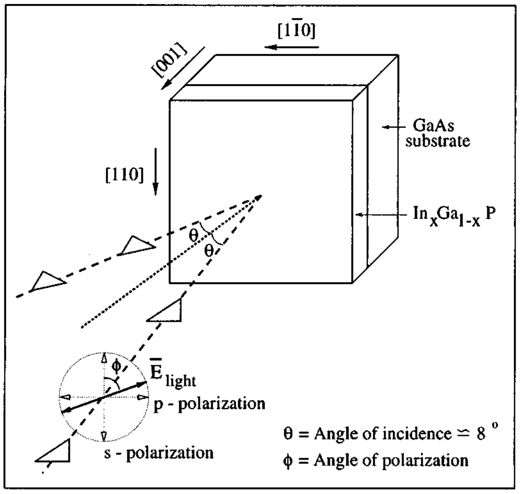

FIG. 3. Schematic of the relative orientation of the sample B with respect to the incident probe beam and its polarization.

$$R=r^s r^{s^*}\cos^2\phi+r^p r^{p^*}\sin^2\phi$$

with

$$r^{s,p}=\frac{r_{12}^{s,p}+r_{23}^{s,p}e^{i4\pi d(u_2+iv_2)/\lambda}}{1+r_{12}^{s,p}r_{23}^{s,p}e^{i4\pi d(u_2+iv_2)/\lambda}}, \tag{2}$$

where ϕ defines the polarization angle of the incident beam as shown in Fig. 3, $r^s(r^{s^*})$ and $r^p(r^{p^*})$ are the complex (complex conjugate) amplitude reflection coefficients for s and p polarized components of the incident probe beam and λ is wavelength of the incident light in air. The quantities $r_{12}^{s,p}$ and $r_{23}^{s,p}$ are complex amplitude reflection coefficients at the air/In$_x$Ga$_{1-x}$P and In$_x$Ga$_{1-x}$P/GaAs interfaces, respectively. These along with the quantities u_2 and v_2 depend on $n_{\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{P}}$, $\kappa_{\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{P}}$, n_{GaAs}, and the angle of incidence (θ) of the light in air. The detailed formulas relating these quantities are taken from Ref. 17. The λ dependent values of n_{GaAs} were taken from Ref. 18 while those of $n_{\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{P}}$ and $\kappa_{\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{P}}$ along with θ and d were estimated by fitting the experimental R spectra. For $\kappa_{\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{P}}$, an Urbach tail in the absorption coefficient of the In$_x$Ga$_{1-x}$P film had to be considered to account for the damping of the OF in the R and the CER spectra at energies slightly below the band gap of the In$_x$Ga$_{1-x}$P film. The fitted R spectrum for sample B is shown by the continuous line in Fig. 1(c). The OF in the CER spectra can be simulated by calculating the difference between the reflectivity of the structure with and without the applied modulating field[5] as follows

$$\left(\frac{\Delta R}{R}\right)_{\mathrm{intr}f}=\frac{R_{\mathrm{field\ on}}-R}{R}. \tag{3}$$

In order to determine $R_{\mathrm{field\ on}}$ we replace $n_{\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{P}}$ by $n_{\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{P}}+\delta n$ when calculating $r^s(r^{s^*})$ and by $n_{\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{P}}-\delta n$ when calculating $r^p(r^{p^*})$ as explained earlier. Initially we assume a constant δn change throughout the In$_x$Ga$_{1-x}$P film thickness. To account for the DIT transition at the In$_x$Ga$_{1-x}$P band gap, the conventional third derivative lineshape function (TDFF)[19] was fitted to the experimental CER spectra for $\phi=45°$ polarization. The parameters thus obtained were used to get the simulated DIT line shape which was then added to the simulated OF to get the total spectrum. For the sake of simplicity, the same parameters were used to get the DIT line shape for all values of ϕ. This procedure therefore will not reproduce the experimentally observed variation of the DIT amplitude (due to ordering) with change in the angle of polarization, but we overlook this point as it is not the main aim of the present study. The resultant combined simulated spectra as a function of the polarization angle ϕ is shown in Fig. 3(b). To match the experimental data a $\delta n=2.8\times10^{-6}$ was required in the simulations. It is evident that the polarization dependence of the OF is well reproduced in the simulations including their absence for $\phi=45°$.

Next, we explore the consequences of the spatial dependence of δn inside the In$_x$Ga$_{1-x}$P layer. In the depletion approximation, the field $F(x)$ as a function of distance x inside the space charge region of a uniformly doped semiconductor falls off linearly from its value $F_{\max}$ at the surface ($x=0$) to zero at a distance W as

$$F(x)=F_{\max}-\frac{eN_d}{\epsilon}x;\quad W=\sqrt{\frac{2\epsilon}{eN_d}(V_{\mathrm{bi}}+V_{\mathrm{ext}})},$$
$$F_{\max}=\sqrt{\frac{2N_d e}{\epsilon}(V_{\mathrm{bi}}+V_{\mathrm{ext}})}, \tag{4}$$

where e is the electronic charge, N_d is the doner concentration ($\simeq1\times10^{16}\ \mathrm{cm}^{-3}$ in our samples), $\epsilon(=11.8\times8.85\times10^{-14}\ \mathrm{F\ cm}^{-1})$ is the dielectric constant, V_{bi} the built in potential at the surface (typically ≃0.5 V), and V_{ext} the magnitude of the externally applied modulation voltage at the surface.[20] Thus even in the absence of the externally applied voltage ($V_{\mathrm{ext}}=0$) there exists a nonzero electric field at the surface and a corresponding depletion width which with the above parameters comes to $W\simeq0.26\ \mu$m. This field would already create a difference in the refractive indices for the two orthogonal polarizations of the light but their effects would be too small to detect in a simple reflectance experiment. In CER spectroscopy, the finite V_{ext} brings about additional change in the existing field, resulting in additional difference in the refractive indices for the two orthogonal polarizations of the light. Although this additional change is ususally smaller still, nevertheless in CER we are able to detect it since we are tuned to measure effects due to just the change in the refractive indices. We also note here that in the low field regime (which is valid here as $\Delta R/R\ll10^{-4}$),[19] the DIT amplitude is proportional to $\Delta F_{\max}^2$ which means that it should vary linearly with V_{ext}. Figure 4(a) shows the variation of the DIT amplitude as a function of the high voltage (V_{app}) applied to the front electrode. The DIT amplitude was estimated by fitting the TDFF line shape function[19] to the experimental data. The variation is evidently linear which suggests that V_{ext} is directly proportional to V_{app}. This is not surprising since by modeling the CER experimental configuration as two capacitors in series, one due to the depletion capacitance of the sample (thickness W and relative permitivity ≃11.8) and the other due to the air gap (thickness ≃0.3 mm), we get the same result that V_{ext} is directly proportional to V_{app} though many orders of magnitude smaller.

As indicated previously, the OF in CER arise due to shifts in the positions of the interference fringes occuring as

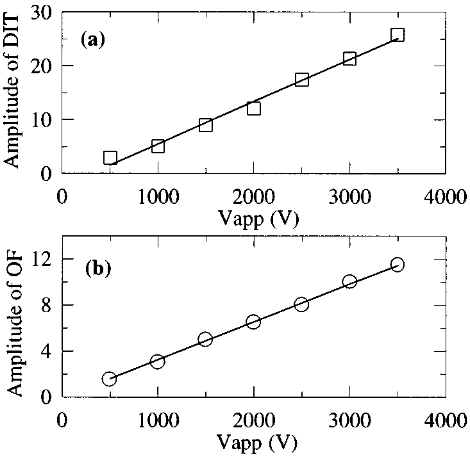

FIG. 4. The dependence of the amplitudes of the DIT transition (a) and the OF (b) on the high voltage applied to the front electrode.

a result of the additional phase difference of the beam reflected from the $In_xGa_{1-x}P/GaAs$ interface because of the modulation of the refractive index of the $In_xGa_{1-x}P$ layer. Our simulations show that for a small total additional optical path difference $\Delta\Psi$, the OF amplitude is directly proportional to $\Delta\Psi$. In the present situation, taking into account the spatial dependence of the field and the linear electro-optic effect, the total additional optical path difference is given by

$$\Delta\Psi=\frac{1}{2}\,n_0^3r_{41}\left[\int_0^{W'}F'(x)dx-\int_0^{W^0}F^0(x)dx\right], \tag{5}$$

where W' and $F'(x)$ are the depletion width and the electric field with the external modulation voltage V_{ext} on and W^0 and $F^0(x)$ are their values when the external modulation voltage V_{ext} is off. Upon evaluation, using the expressions in Eq. (4), the above expression simplifies to

$$\Delta\Psi=\tfrac{1}{2}\,n_0^3r_{41}V_{\mathrm{ext}}. \tag{6}$$

We therefore expect the OF amplitude to vary linearly with V_{app}. That this is indeed true is evident from the plot in Fig. 4(b), justifying the above analysis based on the linear electro-optic effect. Our earlier assumption of a constant δn throughout the thickness d of the $In_xGa_{1-x}P$ film for the simulations is equivalent to considering $\Delta\Psi=\delta nd$. This assumption does not affect our simulations because the OF are sensitive to the total magnitude of $\Delta\Psi$ rather than its detailed spatial dependence. This assumption only underestimates the value of δn, since the δn required to get the same $\Delta\Psi$ over a distance W would be larger than that required if the change occured over a distance d because $d>W$. However the value of δn obtained from the simulations (along with $d=1.05\times10^{-6}$ m for sample B) is useful in that it gives us a measure of $\Delta\Psi=2.94\times10^{-12}$ m. Although the value of r_{41} for $Ir_xGa_{1-x}P$ is expected to be affected by ordering, for an order of magnitude estimation we take $r_{41}\simeq1.26\times10^{-12}$ m/V which is obtained by linear interpolation between the value of this coefficient for InP and GaP.[21] Using this along with $n_0\simeq3.5$ in Eq. (6) we get $V_{\mathrm{ext}}=0.17$ V. This small value of V_{ext} is in accordance with the ''low field regime'' criteria invoked earlier. Also this value of V_{ext} tallies very well with its estimated value based on the two capacitor in series model for CER discussed previously.

IV. CONCLUSION

In conclusion we have explained the origin and the polarization dependence of the sub-band gap oscillatory features in the CER spectrum of $In_xGa_{1-x}P$ films on GaAs (001) substrates and have been able to simulate the experimental observations. We have, to the best of our knowledge, for the first time provided direct evidence for the linear electro-optic effect being the modulation mechanism responsible for giving rise to the OF in the CER spectra of heterostructures. In addition our work shows that the OF can be supressed and its distorting influence on the line shapes can be eliminated if the incident probe light is polarized at 45° to the [110] direction. A situation where this may prove extremely useful is the following. In the CER spectrum of a GaAs quantum well with thick $In_xGa_{1-x}P$ barriers on (001) GaAs substrates, the presence of the OF in the energy range 1.45–1.82 eV can distort or render indistinguishable the transitions associated with the quantum well which are also expected to occur in this energy range. With the incident probe light polarized at 45° to the [110] direction the OF can be supressed while bringing out the undistorted spectral features associated with the quantum well.

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