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Intersubband infrared absorption in stepped quantum wells under intense irradiation

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Abstract

The effect of an intense THz irradiation on the relative intersubband absorption of electrons in stepped quantum wells of GaAs–GaAlAs is theoretically studied. Analytical expressions for the induced current are obtained by means of the adiabatic and resonant approximations within the matrix density formalism. This method allows one to predict the presence of a marked fine structure on the absorption, together with a shift and broadening of the absorption peaks, when the pump intensity is around the megawatts level.

1. Introduction

It is well known that intense transverse electric fields can modify confined states in quantum wells (QWs). An appropriate method for studying these modifications is the examination of interband optical transitions in a sample subjected to transverse electric fields, either static [1–3] or high frequency fields [4, 5]. In order to give a quantitative description of the interband response, it is necessary to take into account the excitonic effects besides the electron and hole state modifications due to transverse fields.

The analysis of the intersubband response is also interesting when electrons are excited from their fundamental state in the conduction band (c) to an excited state by means of infrared (IR) excitation. Electro-optic modulation of intersubband transitions has already been deeply studied [6]. However, the influence of an intense THz irradiation on those transitions is almost unknown [7]. In this paper we study theoretically the effect of such an irradiation in the intersubband IR absorption of electrons.

Experimentally, the aforementioned irradiation can be achieved by means of free-electron (FEL) or gas lasers with energy densities in the MW cm^{-2} region. Applications of these lasers in the study of the heterostructure response can be found in [8, 9].

A standard way to describe electronic states in QWs of width d and depth w, when they are subjected to a transverse electric field $E_\omega(t) = E_\omega \cos \omega t$, uses the adiabatic approach. This approach can be used if $\hbar\omega \ll \varepsilon_{21}$, where ε_{21} is the energy distance between the two deepest levels (ground and excited levels) of the conduction band c. Because these levels oscillate with

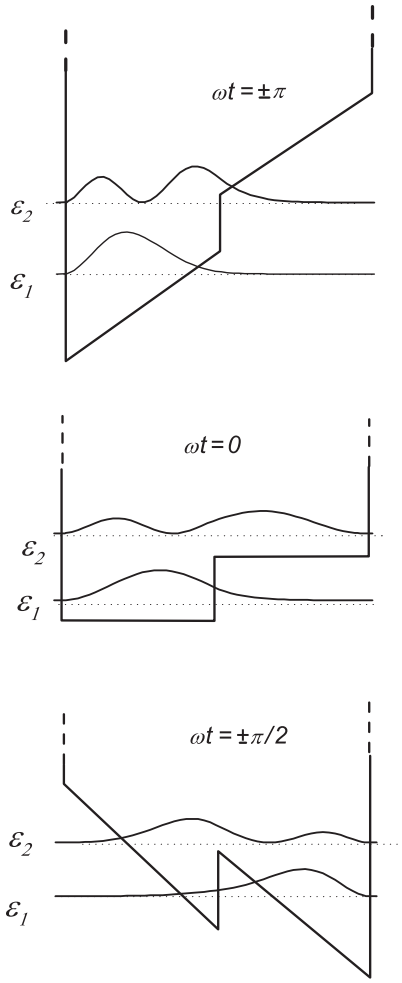

Figure 1. Conduction band diagrams and wavefunctions for stepped QWs under transverse THz irradiation. Dashed lines schematically show the variation of levels under maximal THz field.

frequency ω (see figure 1) intersubband transitions of $(n+1)$ order occur, with the contribution of n THz photons and only one IR photon. These oscillations give rise to a multipeak fine structure of the absorption. Simultaneously, the shape of the absorption peaks is modified by the THz radiation. Such a THz irradiation can be considered as a perturbation whenever the field energy contribution in the well, $|e|E_\omega d/2$, is less than ε_{21}. Otherwise, electronic states have to be numerically calculated.

The procedure we will follow to analyse the absorption is based on the one-particle matrix density equation linearized with respect to the IR field $E_\Omega \exp(-\mathrm{i}\Omega t)$. As commented before, the THz irradiation will be considered within the formalism of the adiabatic approximation. We will describe broadening of the absorption peaks through a phenomenological relaxation frequency, keeping in mind the emission of longitudinal optical phonons, with frequency ω_{LO}, which occurs in the spectral region $\Omega > \omega_{\mathrm{LO}}$. If we take into account both the elastic mechanism of broadening and the LO phonon emission, we can approximate the effective relaxation frequency ν_Ω by a step function: $\nu_\Omega \simeq \nu_{\mathrm{elastic}}$ (due to elastic scattering) if $\Omega < \omega_{\mathrm{LO}}$

and $\nu_\Omega \simeq \nu_{LO}$ if $\Omega > \omega_{LO}$. This approximation is widely accepted because a detailed microscopical analysis of the relaxation does not basically affect the obtained results. An exact calculation including both scattering mechanisms can be found [10]. It would be possible to study electron–LO phonon interaction and elastic scattering as in [10], but under THz pump conditions. However, this microscopical analysis is beyond the scope of the present work. We will also neglect electronic density redistribution between the levels because, in the present case, the level separation $\varepsilon_{21} \sim 100$ meV is greater than the THz energy, $\hbar\omega \sim 1$ meV. Another approximation we will take into account consists in overlooking the THz field influence on the relaxation because the field effect on the wavefunctions is negligible.

We have chosen stepped QWs because, as we will see below, they present suitable characteristics for obtaining a multipeak structure of the absorption.

2. Intersubband absorption

To obtain the relative IR absorption, averaged over the THz pump, we will proceed as follows: first, we deduce the high frequency contribution, $\Delta\rho_{\mathbf{p}}(z, z', t)\exp(-\mathrm{i}\Omega t)$, to the matrix density in the coordinate–momentum representation. This contribution is described by the linearized equation

$$\frac{\partial \Delta\rho_{\mathbf{p}}(z, z', t)}{\partial t} + \frac{\mathrm{i}}{\hbar}\left[\hat{h}(z, t) - \hat{h}(z', t) - \hbar\Omega\right]\Delta\rho_{\mathbf{p}}(z, z', t) + \frac{\mathrm{i}}{\hbar}\left[\widehat{\delta h}(z) - \widehat{\delta h}(z')\right]\rho_{\mathbf{p}}(z, z', t) = 0, \tag{1}$$

where z is the transverse coordinate (growth direction) and $\mathbf{p}$ is the two-dimensional (2D) momentum. The Hamiltonian $\hat{h}(z, t)$ is described by

$$\hat{h}(z, t) = \frac{\hat{p}_z^2}{2m} + w(z) + eE_\omega z\cos\omega t, \tag{2}$$

and the perturbation operator, $\widehat{\delta h}(z)\exp(-\mathrm{i}\Omega t)$, is described by

$$\widehat{\delta h}(z) = \frac{\mathrm{i}e}{\Omega}E_\Omega\hat{v}(z), \tag{3}$$

where $\hat{v}(z) = \hat{p}(z)/2m$ is the velocity operator and $w(z)$ is the potential energy that characterizes the stepped QW.

It is convenient to use parametrized time-dependent wavefunctions, $\varphi^k(z, t)$, which are determined by the eigenvalue problem

$$\hat{h}(z, t)\varphi^k(z, t) = \varepsilon_k(t)\varphi^k(z, t), \tag{4}$$

together with the continuity conditions for the wavefunction and transverse current at each interface, $z = L_i$, between the two materials B (barriers) and W (wells) constituting the structure:

$$\begin{aligned}\varphi^k_{\mathrm{B}}(z, t)\,\big|_{z=L_i} &= \varphi^k_{\mathrm{W}}(z, t)\,\big|_{z=L_i}\,,\\ \frac{1}{m_{\mathrm{B}}}\frac{\partial\varphi^k_{\mathrm{B}}(z, t)}{\partial z}\,\big|_{z=L_i} &= \frac{1}{m_{\mathrm{W}}}\frac{\partial\varphi^k_{\mathrm{W}}(z, t)}{\partial z}\,\big|_{z=L_i},\end{aligned} \tag{5}$$

where the superscript k refers to the level and $L_i = \pm d/2 \pm D$. For numerical purposes we take D as the barrier width separating wells. Because we will study a multiple QW, these barriers must be wide enough to decouple wells (wavefunctions must decay completely between two adjacent wells). Actually, any complete set of functions can be used as a basis in equation (4). The time-dependent Schrödinger equation can also be used but, under adiabatic conditions,

the time-parametrized basis determined by equation (4) appears to be effective. In the basis $\varphi^k(z,t)$ the representation of the density matrix $\Delta\rho_{\mathbf{p}t}(z,z')$ and $\rho_{\mathbf{p}t}(z,z')$ is

$$\begin{aligned}\Delta\rho_{\mathbf{p}}(z,z',t) &= \sum_{kk'} \Delta\rho_{\mathbf{p}}(k,k',t)\varphi^{k*}(z,t)\varphi^{k'}(z',t),\\ \rho_{\mathbf{p}}(z,z',t) &= \sum_{kk'} \rho_{\mathbf{p}}(k,k',t)\varphi^{k*}(z,t)\varphi^{k'}(z',t)\end{aligned} \tag{6}$$

and equation (1) takes the form

$$\begin{aligned}\frac{\partial\Delta\rho_{\mathbf{p}}(k,k',t)}{\partial t} &+ \frac{\mathrm{i}}{\hbar}\left[\varepsilon_k(t) - \varepsilon_{k'}(t) - \hbar\Omega\right]\Delta\rho_{\mathbf{p}}(k,k',t)\\ &+ \sum_{k'}\Big[\Delta\rho_{\mathbf{p}}(k,k',t)\delta^{k'k}(t) - \delta^{kk'}(t)\Delta\rho_{\mathbf{p}}(k',k,t)\Big]\\ &= \frac{\mathrm{i}}{\hbar}\sum_{k'}\Big[\delta h^{kk'}(t)\rho_{\mathbf{p}}(k',k,t) - \rho_{\mathbf{p}}(k,k',t)\delta h^{k'k}(t)\Big],\end{aligned} \tag{7}$$

where the non-adiabatic factor $\delta^{kk'}(t)$ and the perturbation $\delta h^{k'k'}(t)$ are described by

$$\begin{aligned}\delta^{kk'}(t) &= \int_{-d/2}^{d/2}\mathrm{d}z\,\frac{\partial\varphi^{k*}(z,t)}{\partial t}\varphi^{k'}(z,t),\\ \delta h^{kk'}(t) &= \frac{\mathrm{i}e}{\Omega}E_\Omega\int_{-d/2}^{d/2}\mathrm{d}z\,\varphi^{k*}(z,t)\hat{v}(z)\varphi^{k'}(z,t).\end{aligned} \tag{8}$$

We have taken into account that $\delta^{kk'}(t) = -\delta^{k'k*}(t)$ and therefore, the diagonal elements of the non-adiabatic factor are strictly $\delta^{kk}(t) = 0$.

The induced current density can be written by means of $\Delta\rho_{\mathbf{p}}(k',k,t)$ as

$$\begin{aligned}j(t) &= \frac{e}{L^2}\sum_{\mathbf{p}}\int_{-d/2}^{d/2}\mathrm{d}z\lim_{z_1,z_1'\to z}\left[\hat{v}(z_1) + \hat{v}(z_1')\right]\Delta\rho_{\mathbf{p}}(z_1,z_1',t)\\ &= \frac{2e}{L^2}\sum_{\mathbf{p}kk'}v_{k'k}(t)\Delta\rho_{\mathbf{p}}(k,k',t),\end{aligned} \tag{9}$$

where L^2 is the normalization area and $v_{kk'}(t) = \int_{-d/2}^{d/2}\mathrm{d}z\,\varphi^{k*}(z,t)\hat{v}(z)\varphi^{k'}(z,t)$ is the interlevel electronic velocity. For the case of resonant excitation between the first and the second levels, when $\hbar\Omega \sim \varepsilon_{21}$, equation (9) becomes $j(t) \simeq e v_{12}(t)\Delta\rho(2,1,t)$. To obtain this result we have made the summation over 2D momentum according to $\Delta\rho(2,1,t) = (2/L^2)\sum_{\mathbf{p}}\Delta\rho_{\mathbf{p}}(2,1,t)$. Using equation (8) and supposing that only the first level is populated, we can write the differential equation for $\Delta\rho(2,1,t)$ as

$$\frac{\partial\Delta\rho(2,1,t)}{\partial t} + \mathrm{i}\left[\Omega_{21}(t) - \Omega - \mathrm{i}\nu_{21}(t)\right]\Delta\rho(2,1,t) = \frac{eE_\Omega}{\hbar\Omega}v_{21}(t)n_{2\mathrm{D}}, \tag{10}$$

where $\Omega_{21}(t) = [\varepsilon_2(t) - \varepsilon_1(t)]/\hbar$ is the time-dependent interlevel frequency, $\nu_{21}(t) = \nu_\Omega - \delta^{22}(t) + \delta^{11}(t) = \nu_\Omega$ is the effective relaxation frequency, which is described by the phenomenological relaxation frequency ν_Ω, and $n_{2\mathrm{D}}$ is the 2D electronic density. It should be mentioned that ν_Ω has stepped character because it increases in the active region ($\Omega > \omega_{\mathrm{LO}}$) due to LO phonon emission with frequency ω_{LO}. In addition, $\nu_{21}(t) = \nu_\Omega$ because diagonal terms of the adiabatic factor $\delta^{k'k}(t)$ are null. Using the generalized Ohm law, which depends

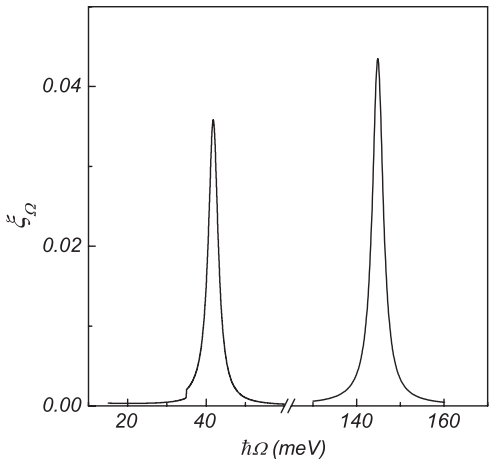

Figure 2. Stepped MQW spectral dependences of ξ_Ω for $E_\omega = 0$ kV cm^{-1} and for the two structures under study. The left peak corresponds to the wide MQW. The right peak corresponds to the narrow MQW.

on time, $\sigma_\Omega(t) = j(t)/E_\Omega$ and solving equation (10) we can write

$$\sigma_\Omega(t) = \frac{e^2 n_{2D}}{\hbar\Omega} v_{21}(t) \int_{-\infty}^{t} \mathrm{d}t'\, v_{21}(t') \exp\left\{-\mathrm{i}\int_{t'}^{t} \mathrm{d}\tau\, [\Omega_{21}(\tau) - \Omega - \mathrm{i}\nu_\Omega]\right\}. \tag{11}$$

The relative absorption, averaged over the THz period [6], can be written as

$$\xi_\Omega = \frac{4\pi}{c\sqrt{\epsilon}} \mathrm{Re} \int_{-\pi/\omega}^{\pi/\omega} \frac{\mathrm{d}t}{2\pi/\omega} \sigma_\Omega(t). \tag{12}$$

Therefore, in order to calculate ξ_Ω we need to know $\Omega_{21}(t)$ and ν_Ω and perform time integrations.

We have studied two different cases: narrow and wide GaAs QWs (100 and 250 Å wide, respectively). In order to discuss a more realistic situation we have considered multiple-quantum-well (MQW) structures consisting of ten GaAs wells separated by AlAs barriers 300 Å wide. The barrier has been chosen wide enough to obtain decoupled wells. We have also used a THz pump with $\hbar\omega = 1.5$ meV. As commented before, we have considered a stepped relaxation energy of $\hbar\nu = 0.66$ meV for $\hbar\Omega < 35$ meV (passive region) and $\hbar\nu = 1.5$ meV for $\hbar\Omega \geqslant 35$ meV (active region). To set a starting point (zero field), in figure 2 we show the energy position and profile of the relative absorption for the narrow and wide QW structures. Figures 3 and 4 show the relative absorption profile for the narrow QW case. One can observe a new fine structure of the absorption which appears around 20 kV cm^{-1}. This structure depends on the electric field in quite a complex way. Together with the decreasing and spreading of the relative absorption, the initial peak for $E_\omega = 0$ kV cm^{-1} gradually splits into several satellite peaks as the field intensity increases, the number of peaks depending on the intensity. The situation is more noticeable for wide QWs (figures 5 and 6). In this case the central peak splits into two relative maxima located at the borders of the relative absorption profile together with a set of minor maxima. In all cases the multipeak fine structure is partially hidden for $\hbar\Omega_{21} > 35$ meV due to the higher effective relaxation energy. Figures 7 and 8 show the evolution of the maxima position as a function of the amplitude of the THz transverse electric field. The origin of the multipeak absorption has a quantum mechanical basis. We will

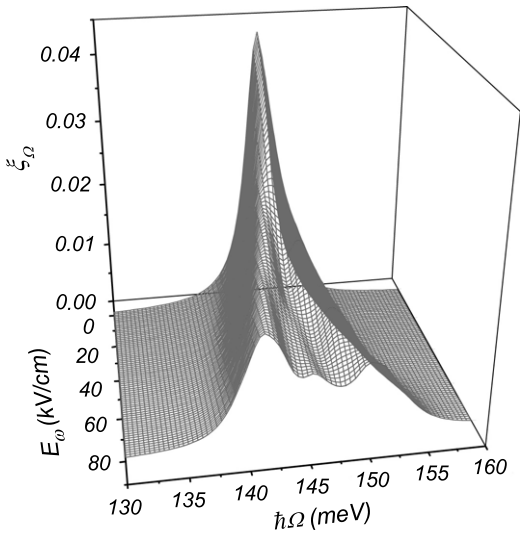

Figure 3. Stepped narrow MQW spectral dependences of ξ_Ω versus E_ω. The irradiation frequency is $\omega = 2.3$ THz.

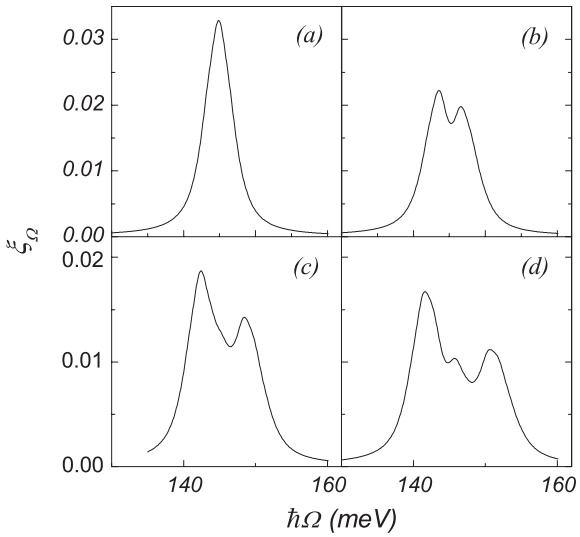

Figure 4. Relative absorption ξ_Ω of the stepped narrow MQW versus $\hbar\,\Omega$ and for $E_\omega = 20\,\mathrm{kV\ cm^{-1}}$ (a), $40\,\mathrm{kV\ cm^{-1}}$ (b), $60\,\mathrm{kV\ cm^{-1}}$ (c), $80\,\mathrm{kV\ cm^{-1}}$ (d). The irradiation frequency is $\omega = 2.3$ THz.

use a simple picture to explain it. Let us consider a rectangular QW under a moderate THz field. The solution of the eigenvalue equation (4), in the parabolic approximation (for low and moderate THz field amplitude), can be written by means of cosine series as follows [7]:

$$\begin{aligned}\varphi_{zt}^{k} &= \varphi_{z}^{k} + \sum_{k'\neq k} \frac{eE_\omega z_{kk'}}{\varepsilon_k - \varepsilon_{k'}}\varphi_{z}^{k'}\cos\omega t + \cdots,\\ \varepsilon_{kt} &\simeq \varepsilon_k + eE_\omega z_{kk}\cos\omega t + \sum_{k'\neq k}\frac{|eE_\omega z_{kk'}|^2}{\varepsilon_k - \varepsilon_{k'}}\cos^2\omega t,\end{aligned} \tag{13}$$

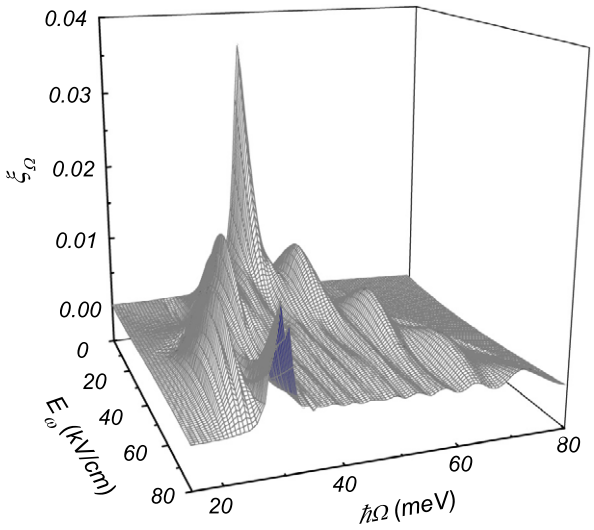

Figure 5. Stepped wide QW spectral dependences of ξ_Ω versus E_ω. The irradiation frequency is $\omega = 2.3$ THz.

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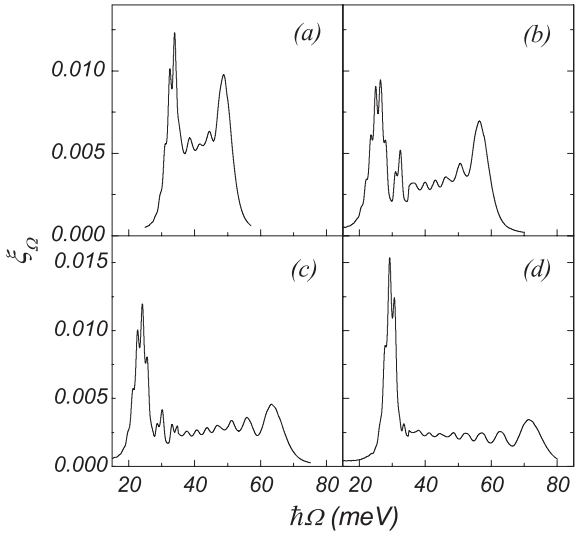

Figure 6. Relative absorption ξ_Ω for the stepped wide MQW versus IR field energy and for $E_\omega = 20$ kV cm^{-1} (a), 40 kV cm^{-1} (b), 60 kV cm^{-1} (c), 80 kV cm^{-1} (d). The irradiation frequency is $\omega = 2.3$ THz.

where the position matrix elements $z_{kk'}$ and the energy level ε_k are determined from the zero-field eigenstate problem: $(\hat{p}_z^2/2m + w_z)\varphi_z^k = \varepsilon_k \varphi_z^k$, with the boundary conditions $\varphi_{|z|=d/2} = 0$. Due to THz field variation, energy levels oscillate and several interlevel transitions are possible for different k, k' values. If one calculates the relative absorption (equations (11) and (12)) using the above wavefunctions, and taking into account the relations $\exp(\mathrm{i}z\cos\psi) = \sum_{l=-\infty}^{\infty} \mathrm{i}^l \mathrm{e}^{\mathrm{i}l\psi} J_l(z)$, $\exp(\mathrm{i}z\sin\psi) = \sum_{l=-\infty}^{\infty} \mathrm{e}^{\mathrm{i}l\psi} J_l(z)$, $\exp(z\cos\psi) = \sum_{l=-\infty}^{\infty} \mathrm{e}^{\mathrm{i}l\psi} I_l(z)$, the

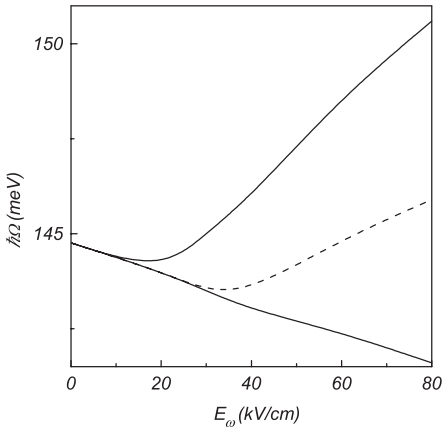

Figure 7. Relative absorption peak evolution for the stepped narrow MQW. The irradiation frequency is $\omega = 2.3$ THz.

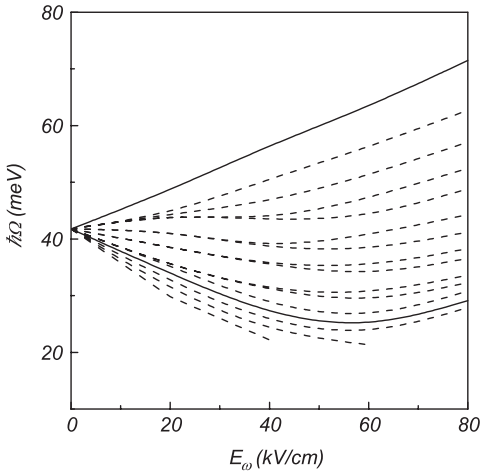

Figure 8. Relative absorption peak position for the stepped wide MQW. The irradiation frequency is $\omega = 2.3$ THz.

solution can be written through the k-order Bessel functions

$$\xi_\Omega \simeq \frac{4\pi}{\sqrt{\epsilon}}\frac{e^2}{\hbar c}\frac{\nu_\Omega |v_{21}|^2}{\Omega} n_{2D} \sum_{k=-\infty}^{\infty} \frac{J_k(\Omega_{21}^{(2)}/4\omega)^2}{(\Omega_{21} - 2k\omega - \Omega)^2 + \nu_\Omega^2}, \tag{14}$$

leading to the multipeak structure. For low THz field intensities (parabolic approximation) $\Omega_{21}^{(2)}$ has small values and only two superimposed peaks appear, which split when the electric field increases. If the THz field is increased beyond the parabolic approach one needs to add more terms to the cosine expansion and the number of peaks increases. For higher THz fields it is not possible to find a simple Bessel expansion for the absorption. It is also very hard to write down a simple cosine expansion for stepped QW and high THz fields, and numerical solutions are needed. However, one can extrapolate that the origin of the absorption fine structure will be the same as in the simple rectangular QW.

3. Conclusions

In summary, we have studied modifications of the intersubband absorption in a stepped MQW structure caused by an intense THz irradiation. Such an irradiation can be experimentally achieved by using FEL or gas lasers, with energy densities around MW cm^{-2}. We have shown that the absorption peak corresponding to an external field $E_\omega = 0$ kV cm^{-1} splits into a set of satellites (fine structure) due to $(n+1)$-order intersubband transitions with the participation of n THz photons and one IR photon. In addition, a strong modification of the absorption, which consists in a noticeable broadening of the zero-field peak and a shift towards higher energy values, is also obtained. We have used in calculations two different MQW structures formed by ten decoupled QWs. The reason is that the relative IR absorption appears to be too weak to be detected in a single QW. On the other hand, hundred-QW structures with identical QWs are used in quantum cascade lasers, so we have discussed a realistic case. Photoconductivity measurements may be more sensitive and a single-QW structure should be suitable for such a case.

We have not taken into account the Coulomb renormalization which diminishes depolarization and exchange effects. This approximation is possible because the interlevel distance is relatively large (at worst, $\varepsilon_{21} \sim 30$ meV for wide QW) and small changes in the level position do not affect results essentially. We have also neglected the THz field effect on the damping because the influence of such a field on the wavefunctions is negligible. Thus, phenomenological broadening should not be essentially dependent on the THz pump.

To conclude, the present work has shown that a large modification of the IR intersubband response in stepped QWs takes place when an intense THz irradiation is applied. We expect the results obtained to stimulate researchers to carry out experiments in this direction.

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