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ABSTRACT

The generation of a nonequilibrium population of optical phonons by photoexcited hot electrons in semiconductor quantum wells is investigated theoretically. The microscopic model of electron-phonon interaction proposed by Huang and Zhu has been used to compute the distributions of confined longitudinal optical phonons and interface modes in GaAs/AlAs quantum wells as a function of well width. Experimental tests of the calculated distributions by Raman scattering are discussed.

1. INTRODUCTION

There has been much interest in the properties of quasi-two-dimensional microstructures fabricated from two semiconductors with different band gaps such as GaAs and AlAs.[1] These structures are referred to as quantum wells (QW) because electrons and holes are confined by the potential wells created by the different layers. Optical phonons may also be confined in such QW.[2] If the optical phonon (OP) frequencies in the two layers do not overlap (as in case of GaAs/AlAs QW) and the layer thicknesses are large enough, OP cannot propagate from one layer to the next. The confinement of electrons and OP in QW has significant influence on the electron-phonon interaction and hence on hot electron relaxation in QW.[3] In addition to these effects, the presence of interfaces between layers in QW introduces new vibrational modes known as interface phonons.[4,5] These interface modes are expected to play an important role in hot electron relaxation in QW with well widths of the order of nm.[6,7]

Relaxation of hot electrons via electron-phonon interactions in bulk semiconductor samples has been studied extensively.[8] It is now recognized that in semiconductors, such as GaAs, interaction of electrons with the longitudinal optical (LO) phonons and zone-edge phonons is responsible for the relaxation of hot electrons in subpicosecond time scales. Several experimental techniques have been developed to study hot electron relaxation in semiconductors. One of the optical techniques involves using Raman scattering to study non-equilibrium phonons generated by hot electron relaxation. Both intravalley and intervalley electron-phonon interactions can be studied by this technique.[8] Hot electrons are excited with a picosecond or subpicosecond pump laser. The time evolution of the non-equilibrium optical phonon (NEOP) population generated by the hot electrons is monitored via Raman scattering with a time-delayed probe pulse.[9] Since crystal momentum is conserved in Raman scattering, this technique can probe only part of the NEOP produced by the hot electrons. In bulk GaAs this technique has been shown to be quite sensitive for studying the electron-LO phonon interaction. This is because the momentum of the LO phonon probed by Raman backscattering with a visible laser (wavelength between 600-500 nm) is very near the peak of the NEOP distribution.[10]

So far understanding of the electron-phonon interaction and hence of hot electron relaxation in QW has lagged behind that for bulk samples. There have been controversies as to the correct form of the electron-LO phonon interaction (Fröhlich interaction) for confined phonons in QW.[7] Recently, Kim and Yu[11] have tried to extend the NEOP technique to study electron-phonon interaction in GaAs/AlAs QW. They found that the NEOP population excited by subpicosecond laser pulses decreased monotonically as the QW width L was decreased. So far there is no quantitative explanation of their results. In this paper we will examine the distribution of NEOP population excited by hot electrons in GaAs/AlAs QW as a function of L. Our results show that the distribution of NEOP generated by intersubband scattering is quite different from that produced by intrasubband scattering. In addition, the distributions for the confined phonon modes and for the interface modes vary differently with L. Thus by measuring the NEOP populations excited by picosecond laser pulses, it is possible to study the Fröhlich interaction in QW just as in bulk GaAs.

2. MODEL CALCULATIONS & RESULTS

Figure 1 shows schematically a GaAs QW of thickness L surrounded on both sides by barriers of AlAs. The z-axis is taken to be perpendicular to the layers. To calculate the NEOP distribution generated by a hot electron gas, it is necessary to know the electronic energy bands, the confined phonon modes and the electron-phonon interactions.

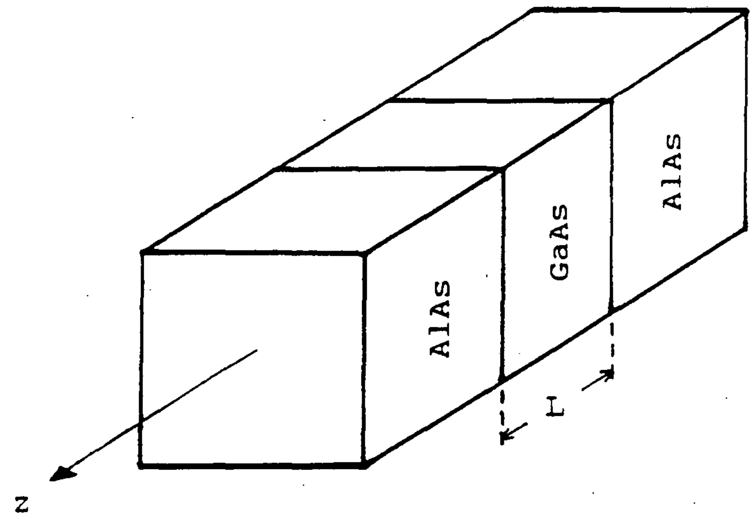

FIG. 1 Schematic diagram of a symmetric GaAs/AlAs QW.

To simplify the calculation we will assume that L is small enough so that at most two (lowest) subbands (to be labeled as n=1 and 2) are significantly populated by hot electrons. The energies and wave functions of the electrons in these subbands are calculated with Bastard's four-band model.[12] We will assume that the hot electron distributions in these two subbands are Maxwell-Boltzmann. This assumption is appropriate for a low density electron gas excited by picosecond laser pulses. The GaAs phonon wave vector **q** in the QW will be decomposed into (Q,q_z), where Q is the component parallel to the well. Due to confinement q_z is quantized into: $q_z=m\pi/(L+a)$ where m is an integer larger than 2 and a is the thickness of a monolayer of GaAs.[13] The confined LO phonons will be denoted as LOm. In addition to the confined modes, there are four interface modes usually labeled as IF^+_s, IF^+_{as}, IF^-_s and IF^-_{as}.[4,5] In case of GaAs/AlAs QW, the modes with the superscript + have frequencies between the AlAs TO and LO phonons. This mode is also referred to as the AlAs interface mode. The modes indexed by the superscript - are the corresponding GaAs interface modes. The subscripts s and as refer to the parity of these modes under reflection about the center of the well. Since the signs + and - are sometimes used to denote parity, we will avoid using + and - to label the interface modes. Instead we will label the symmetric GaAs interface mode as IF1 and the symmetric AlAs interface mode as IF2. In this paper we will neglect the antisymmetric interface modes since only the symmetric interface modes are observable in Raman scattering. To calculate the electron-phonon interaction we use the microscopic model of Huang and Zhu (HZ)[3] in which the Fröhlich Hamiltonian for the confined modes is given by:

$$H_F = -|e|\phi_m(z)F(Q) \tag{1}$$

where e is the electronic charge, ϕ_m is the electric potential associated with the confined phonon LOm, and F(Q) is a function of the phonon coordinate Q defined by HZ in Ref.3 as Eq. (37). HZ proposed the following analytic expressions for ϕ_m:

$$\phi_m = \begin{cases} \cos[m\pi z/(L+a)] - (-1)^{m/2} & m=2, 4,.. \\ \\ \sin[\mu_m \pi z/(L+a)] + C_m z/(L+a) & m=3, 5,.. \end{cases} \tag{2}$$

The constants μ_m and C_m are defined in Ref. 3. For the IF modes HZ found that the microscopic model and the dielectric continuum model gave identical results for the electron-interface phonon interaction.[3] As a result we will use the Hamiltonian given by Mori and Ando[6] for (the symmetric modes of) the dielectric continuum model:

$$\mathcal{H}_{MA} = \left[\frac{\pi\hbar\omega_{i\mathbf{Q}}e^2}{A}\right]^{1/2}\left[\beta_1^{-1}(\omega_{i\mathbf{Q}})\tanh(QL/2) + \beta_2^{-1}(\omega_{i\mathbf{Q}})\right]^{-1/2}\frac{e^{i\,\mathbf{Q}\cdot\mathbf{R}}}{\sqrt{Q}}f_S(Q,z)$$

$$\times\left[a_{i\mathbf{Q}} + a^{\dagger}_{i,-\mathbf{Q}}\right] \tag{3}$$

where the functions $f_S(Q,z)$ and $\beta_i(\omega)$ are given in Ref. 6. The matrix elements of the above electron-phonon Hamiltonians are calculated with electron wave functions for an infinitely deep potential well.

In bulk GaAs, phonons emitted during hot electron relaxation have a well-defined range of values of Q determined by the electron dispersion as a result of momentum conservation.[10] In case of QW, only the component of the wave vector parallel to the layers (**Q**) is conserved in scattering. For typical Raman backscattering geometry in GaAs QW with visible lasers, Q has values less than 10^5 cm^{-1}. In bulk GaAs the Fröhlich matrix element $|H_F|^2$ is proportional to $1/q^2$. Thus one effect of confinement of LO phonon in QW is to make the Fröhlich interaction dependent on L:

$$|H_F|^2 \propto [Q^2+(m\pi/L)^2]^{-1} \tag{4}$$

For L smaller 10 nm, q_z is much larger than Q so $|H_F|^2$ decreases with L as $(m\pi/L)^{-2}$. Kim and Yu[11] have pointed out that this is one reason for the decrease in the NEOP generation efficiency with L they observed. Another factor contributing to the decrease is that there is a minimum value of Q (Q_{min}) below which the NEOP population drops to zero. Q_{min} depends on the well width, and for small L, Q_{min} can become too large to be observed in Raman backscattering. To test these ideas quantitatively we have calculated the NEOP distribution in GaAs/AlAs QW as a function of L.

The NEOP distributions are obtained by solving a set of rate equations for the population N_{iQ} of the phonon mode i and wave vector **Q**, and the electron population N_e:

$$\frac{\partial N_{iQ}}{\partial t} = \frac{2\pi}{\hbar}\sum_{\mathbf{K}}\Big[\, |\langle N_{iQ}+1,\mathbf{K}-\mathbf{Q}|\,\mathscr{H}_{eph}\,|N_{iQ},\mathbf{K}\rangle|^2 f_{\mathbf{K}}(1-f_{\mathbf{K}-\mathbf{Q}})\,\delta(E_{\mathbf{K}-\mathbf{Q}}-E_{\mathbf{K}}+\hbar\omega_{iQ}) - |\langle N_{iQ}-1,\mathbf{K}+\mathbf{Q}|\,\mathscr{H}_{eph}\,|N_{iQ},\mathbf{K}\rangle|^2 f_{\mathbf{K}}(1-f_{\mathbf{K}+\mathbf{Q}})\,\delta(E_{\mathbf{K}+\mathbf{Q}}-E_{\mathbf{K}}-\hbar\omega_{iQ})\Big] \qquad (5)$$

$$\frac{\partial}{\partial t}(N_e\langle E\rangle) = -\sum_{i,\mathbf{Q}}\hbar\omega_{iQ}\frac{\partial N_{iQ}}{\partial t} + \frac{E_0\,\partial N_e}{\partial t}\Big]_{laser} \qquad (6)$$

In Eq. (5) $\hbar$ is Planck's constant; H_{eph} is the appropriate electron-phonon interaction. K, E_K and f_K are, respectively, the electron wave vector, energy and distribution function. f_K is assumed to be always Maxwell-Boltzmann, otherwise the calculation would be much more complex. We have neglected the decay of phonons due to their finite lifetime, so our results are valid only for comparison with picosecond and subpicosecond experiments. In Eq. (6) <E> is the average electron energy defined by:

$$<E> = \sum f_K E_K / \sum f_K \qquad (7)$$

and E_0 is the initial kinetic energy of the electrons excited by a monochromatic laser pulse. We will examine separately the NEOP distributions generated by <u>intra</u>subband and <u>inter</u>subband scattering in GaAs/AlAs QW.

2.1 Intrasubband Scattering

For intrasubband scattering we will consider only wells with L small enough for the interface modes to contribute significantly to hot electron relaxation. Equations (5) and (6) are integrated subjected to the following initial conditions: a two-dimensional electron gas of areal density 2×10^{10} cm^{-2} is excited at time=0 in the n=1 subband with an average energy of 200 meV. The lattice temperature is assumed to be 10 K. From parity consideration only phonon modes whose H_{eph} is even are allowed in intrasubband scattering of hot electrons. Of such phonons the more important ones are the interface modes: IF1 and IF2 and the lowest order confined LO phonon: LO2. Their relative contributions depend on the well thickness. As pointed out by several authors,[6,7] intrasubband scattering of electrons by interface modes becomes more important than confined phonon modes when L is less than 10 nm. Figure 2 shows the non-equilibrium phonon distributions generated by hot electrons 1 ps after excitation. Results for L=2 nm and 6 nm are shown for comparison.

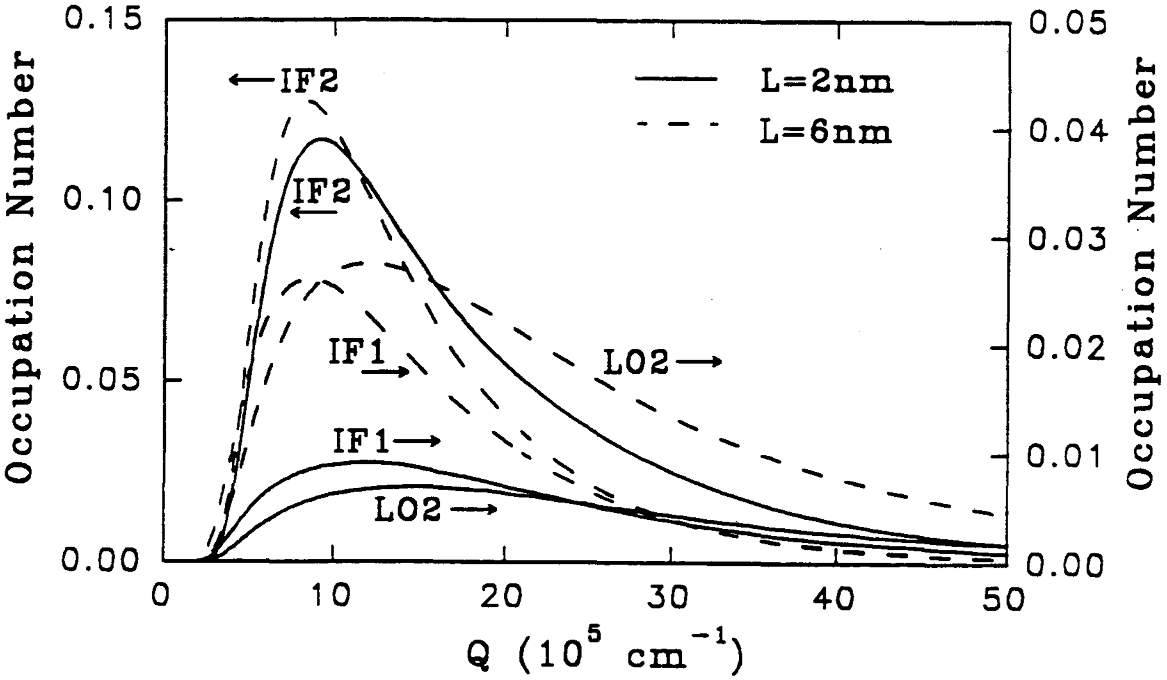

FIG. 2 A plot of the populations of IF1, IF2, and confined LO2 phonons versus Q for L=2 and 6 nm.

Qualitatively these curves are similar to the NEOP distribution generated by hot electrons in bulk GaAs calculated by Collins and Yu[10] (reproduced in Fig. 3).

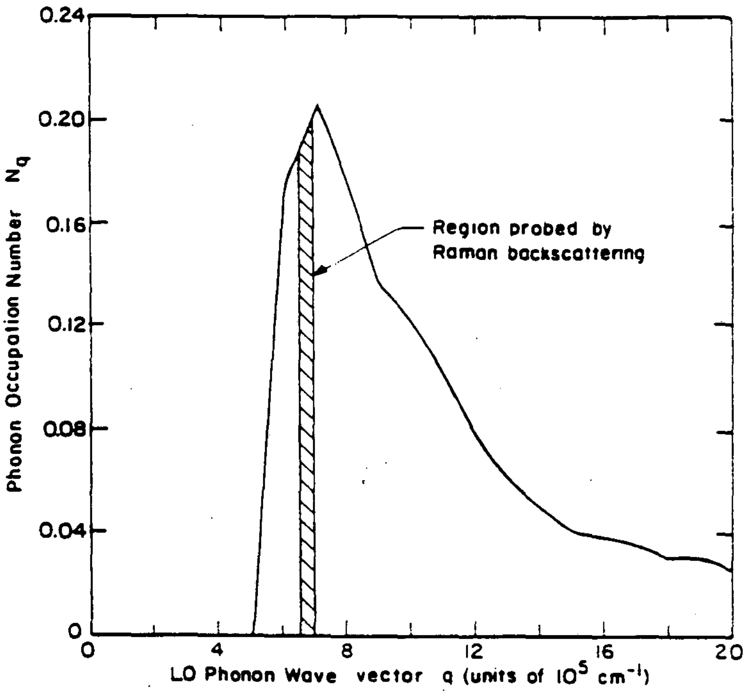

FIG. 3 Non-equilibrium LO phonon population generated by hot electrons in bulk GaAs. Reproduced from Ref.10.

For example there is a minimum value of Q of about $2\text{x}10^5$ cm^{-1} below which the populations drop to zero. The important feature of the curves in Fig. 2 is their different dependence on L. For the modes LO2 and IF1, the overall populations increase as L increases. For the mode IF2, the variation in population with L depends on Q. While for most values of Q the population decreases as L increases, for a narrow range of values of Q near the peak, the population of IF2 actually increases slightly with L. However, when integrated over all values of Q, the population of the mode IF2 decreases rapidly as L increases. Our results are consistent with the calculations of the electron-phonon coupling strengths by Mori and Ando.[6] These authors showed that for the IF2 mode the coupling strength decreases as the product QL is increased while the opposite is true for the IF1 mode.

2.2 Intersubband Scattering

For studying scattering of electrons from the n=2 to the n=1 subband by phonons, we consider QW with L between 15 and 25 nm. In this range of values of L, the separation between the two subbands is comparable to the confined LO phonon energies. Recently Yu and Kim[11] have proposed to use L to "tune" the intersubband separation across the confined LO phonon energy. The idea behind this proposal is that, in a Raman backscattering experiment, only LO3 phonons with $Q \lesssim 10^5$ cm^{-1} can be detected due to momentum conservation. Since Q_{min} in intersubband scattering is typically much larger than 10^5 cm^{-1}, one will not observe any NEOP in Raman scattering. The exceptional case is when the intersubband separation is resonant with the phonon energy. In this case LO3 with essentially zero Q can be emitted in intersubband scattering. As a result a sharp "resonance" in the NEOP population should occur as a function of L.

Using the same initial conditions as for intrasubband scattering, we obtained the NEOP distributions of the LO3 mode for various values of L shown in Fig. 4.

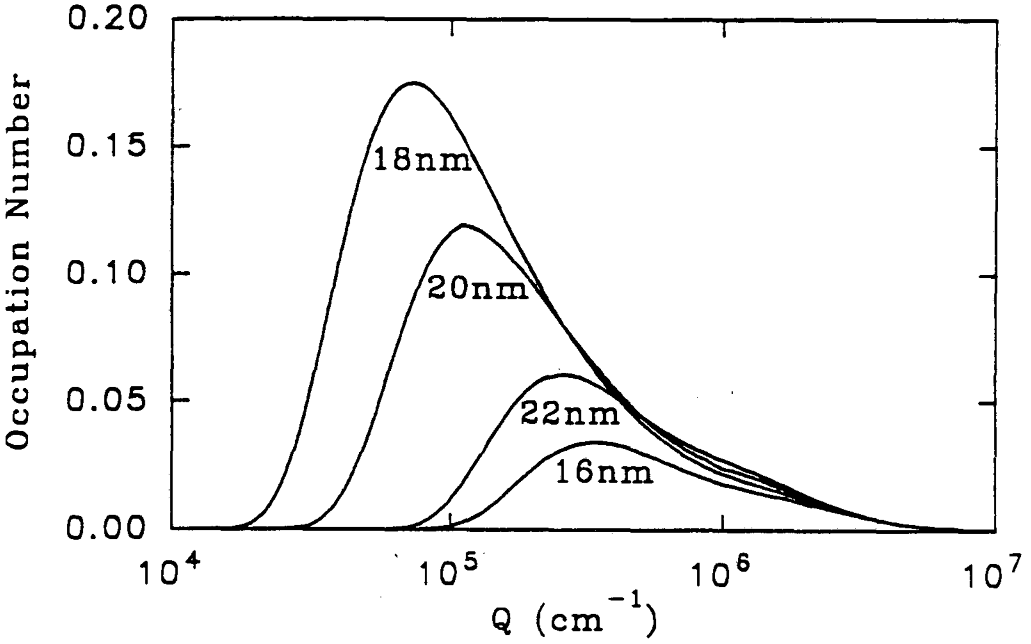

FIG. 4 A semi-log plot of the population of LO3 phonon generated by intersubband scattering as a function of Q for various L between 16 and 22 nm.

Unlike the case of intrasubband scattering, we find that Q_{min} depends strongly on L. In order to show this dependence of Q_{min} on L we have plotted Q on a logarithmic scale. Using these results we can calculate the NEOP population at a fixed Q equal to 7×10^4 cm^{-1} as a function of L. This is shown in Fig. 5. As predicted by Yu and Kim,[11] we find a peak at L=18.7 nm with a full-width at half-maximum of only 1.7 nm.

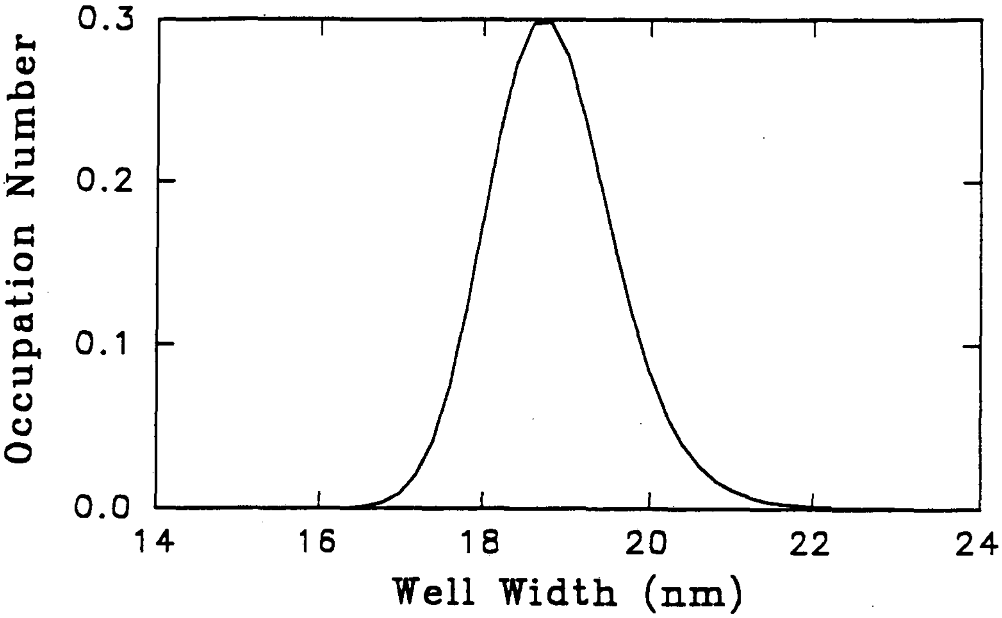

FIG. 5 A plot of the LO3 population for a fixed $Q=7\times10^{4}$ cm^{-1} versus L to show the "resonance" effect.

3. DISCUSSION

Experimental determination of the NEOP distributions calculated in the present paper will provide detailed information on the interaction between electrons and phonons in QW. The easiest way to measure non-equilibrium phonon distributions is via Raman scattering (RS), since the laser pulses used to excite the hot electrons can also be used to probe the phonons emitted by the hot electrons. Unfortunately RS cannot probe the phonon distribution over a large range of phonon wavevectors. In bulk GaAs samples, the wavevector of phonon observed in RS is limited by the wavevector of the photons due to wavevector conservation. For visible photons, the maximum phonon wavevector accessible by RS is typically less than 10^6 cm^{-1}. In QW only the component of wavevector parallel to the well (Q) is conserved. As a result it is necessary to irradiate the sample and collect the scattered radiation both at grazing angles to achieve the largest Q. Due to experimental constraints, the maximum Q accessible in RS from QW is less than 10^5 cm^{-1}. With this constraint, RS has only very limited use in studying NEOP in QW. As shown in Fig. 2, none of the phonons generated by intrasubband scattering in QW with L less than 60 nm can be observed by RS. For phonons generated by intersubband scattering, only QW with a very narrow range of L will have Q small enough to be observable by RS. The experimental observation of the "resonance" effect depicted in Fig. 5 would be a verification of the validity of our model calculation.

The above discussion assumes that wavevector is conserved in RS. It is well known that wavevector conservation in RS can be violated in the presence of defects.[14] Such breakdown in momentum conservation can result from elastic scattering between defects and electrons. In principle such defect-mediated RS involve a higher order perturbation theory and should be weaker than momentum-conserving Raman processes. However, it has been shown by Menendez and Cardona[15] and by Berg and Yu[16] that such defect-mediated process can dominate over momentum-conserving processes when the scattered photon energy is resonant with an energy gap or exciton. Thus one way to probe NEOP population in QW would be to use one picosecond laser to excite hot electrons high above the band gap and use another picosecond laser with wavelength resonant with the fundamental band gap to probe the NEOP emitted by the hot electrons. Such picosecond RS experiments have recently been reported by several groups[17,18] and the results presented here should help to interpret these experimental results.

4. CONCLUSIONS

We have presented a model calculation of the NEOP distributions generated by a hot electron gas in GaAs/AlAs QW as a function of the well width. We have shown that the NEOP populations generated by intrasubband and intersubband scattering depend differently on the well width. We have suggested experimental tests of the results of our model calculations.

5. ACKNOWLEDGEMENTS

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