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Quasi-Two-Dimensional Resonant Bound Polarons.

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Abstract. – A magneto-optical study has been made of the intra-donor $1s \rightarrow 2p^+$ transitions in δ-doped (planar-doped) GaAs-GaAlAs multi-quantum wells (MQW). A clear polaron pinning to the longitudinal-optical (LO) phonon of GaAs is observed. It is found that the magnitude of the resonant bound-electron LO-phonon interaction depends upon the location of the impurity in the MQW sequence. This reveals the remarkable possibility of monitoring quasi–two-dimensional resonant bound polarons in confined semiconducting structures. Our experiments also give evidence for «deeper» shallow donors in the GaAlAs barriers, which arise as a natural effect of the compensation of acceptor states.

Resonant Fröhlich polarons in weakly polar semiconductors have been extensively studied, since they are one of the most challenging examples of resonantly coupled fields in condensed-matter physics [1, 2]. The study of Fröhlich polarons has gained recently revitalized interest [3-9] in the context of quasi–two-dimensional (2D) electronic systems. Indeed, because of the relaxation of selection rule for momentum perpendicular to the interface (z-direction), polaronic effects are expected to the stronger than in bulk material [3]. However, the finite extension of the wave function in the z-direction and free-carrier screening are expected to reduce the interaction considerably, even below the bulk limit [4, 5]. The analysis of the experimental data (up to date restricted to the cyclotron resonance in degenerate systems [6-9]) has long been controversial and remains partly so today. Besides the difficulty of gaining reliable information in an energy range where the materials are opaque due to the reststrahlen band, the resonant behaviour of the dielectric function at the transverse optical (TO) phonon energy can lead to a misinterpretation of the results [9, 10]. However, the key role played by the free-carrier screening of the Fröhlich interaction has been unambiguously stressed [5]. Thus, there is need of experiments on

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resonant polarons in «insulating» systems, free of any screening. It is the aim of this letter to show that the magneto-optical study of the intra-donor transitions $1s \rightarrow 2p^+$ in selectively doped GaAs-$Ga_{1-x}Al_xAs$ ($x \simeq 0.3$) multi-quantum wells (MQW) provides us with a new opportunity to study quasi-2D resonant polarons in «insulating» systems, and to gain valuable information on their sensitivity to the reduced dimensionality.

We used GaAs-GaAlAs MQW grown by molecular beam epitaxy (MBE). The GaAs well (L_w) and GaAlAs barrier (L_b) thicknesses were chosen to be 100 Å. The results presented here have been obtained with a sample nominally doped on-centre in the wells: silicon dopant atoms were introduced in the MQW sequence ($N_d \simeq 10^{10}\,\text{cm}^{-2}$) using the δ-doping (or planar-doping) procedure in which growth is interrupted during doping [11]. Magneto-transmission (MT) and magneto-photoconductivity (MPC) measurements were carried out using an optically pumped far-infrared laser operating in the range $\lambda = (33 \div 70)\,\mu\text{m}$ and magnetic fields up to 20 T. Thus all our measurements are field-sweep measurements at fixed photon energy. For the MPC measurements four ohmic contacts were obtained by In diffusion: the spectra were recorded with a constant voltage applied to the sample (in the interface plane). Special care was taken to work in the ohmic region of the photoresponse of the sample.

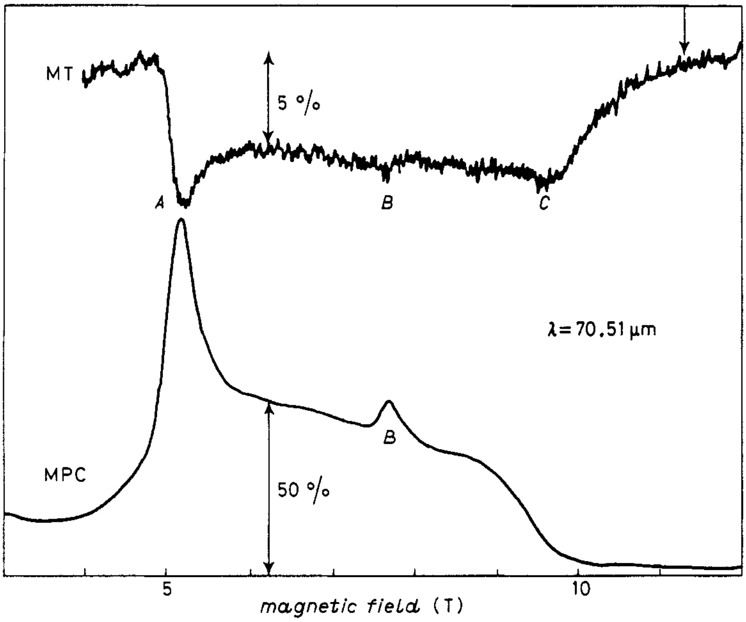

Fig. 1. – A comparison between MT and MPC spectra. $T = 4.2$ K for the MT spectrum and $T = 8$ K for the MPC spectrum (the MPC sensitivity is very high at this temperature). The arrow points out the field position of the 2D cyclotron resonance, which is observed only for $T > 12$ K when the donors are thermally ionized.

Typical MT and MPC spectra are shown in fig. 1. By comparison with the one-impurity theory [12, 13], the peaks A and C in the MT spectra are interpreted as due to the $1s \rightarrow 2p^+$ transitions of on-well-centre and on-barrier-centre donors, respectively. This dependence of the transition energies on the impurity location leads to the formation of a spatial one-impurity band with a very high density-of-states (DOS) [14] on-centre in the wells and in the barriers [12, 13]. This should in principle allow us to determine the real donor profile using an appropriate fitting procedure [15]. However, such an analysis provides us only with semi-quantitative information because of the additional peak B for which the one-impurity theory gives no explanation. This peak is commonly observed in doped MQW with narrow wells ($L_w < 200$ Å), but no interpretation has been found for it up to now [16]. It appears once dopants are introduced in the barriers [16]. We have carried out MT measurements in several uniformly and selectively doped samples (not shown) and peak B is indeed observed together with the expected peak C for barrier impurities. In fig. 2, we have plotted MPC spectra obtained with the magnetic field applied in the z-direction ($\theta = 0°$) and tilted ($\theta = 20°$) with respect to it. The results show that the whole structure—*including peak B*—is confinement related. A detailed discussion of peak B will be given elsewhere: for the purpose of this letter, we simply would like to summarize the conclusions we are led to draw from it. We think that it is due to «deeper» barrier impurities with a binding energy higher than expected as a consequence of the compensation of acceptor states. Acceptors are always present in the structure, as shown in independent luminescence experiments [17], and some donors in the barriers are ionized in order to compensate them. Thus, there is a finite probability for the electron weakly bound to the barrier donors to feel the attractive Coulombic potentials of these ionized donors. By analogy with bulk GaAs [18] this mechanism results in an increased binding energy, and thus in the peak B, while peak C is due to the remaining «isolated» neutral donors in the barriers.

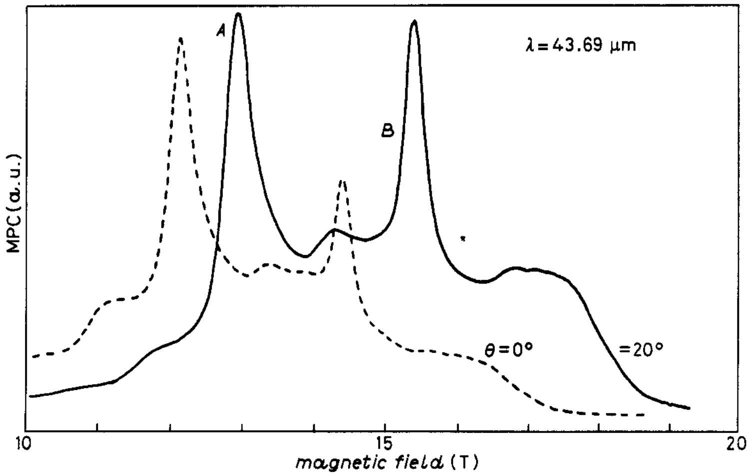

Fig. 2. – MPC spectra at $E = 28.38$ meV where the sample is opaque in MT experiments. The tilted field spectrum confirms that the whole structure is confinement related. The weak peaks at the low-field side of each $1s(z_i) \rightarrow 2p^+$ transition are the corresponding $1s(z_i) \rightarrow 3p^+$ transitions [12, 13]. The change in sensitivity between both spectra is due to a small change in temperature.

The MPC spectra in fig. 1 and 2 reveal the well-known sensitivity of the photoconductivity technique which allows us to reach the optical phonon energies with a very high signal-to-noise ratio, although the relative intensities of the peaks are not reproduced as usual [19] (peak C is almost not seen in the MPC spectra). The field positions of MPC peaks A and B are plotted in fig. 3 together with the bulk data of Sigg *et al.* [20]. While both intra-donor transition energies A and B vary almost linearly with the field for energies up to 25 meV a deviation from linearity is observed above this which is very pronounced as the phonon energies are approached. In this energy range great attention must be paid to the response function of the sample. As calculated in ref. [9, 10] the field

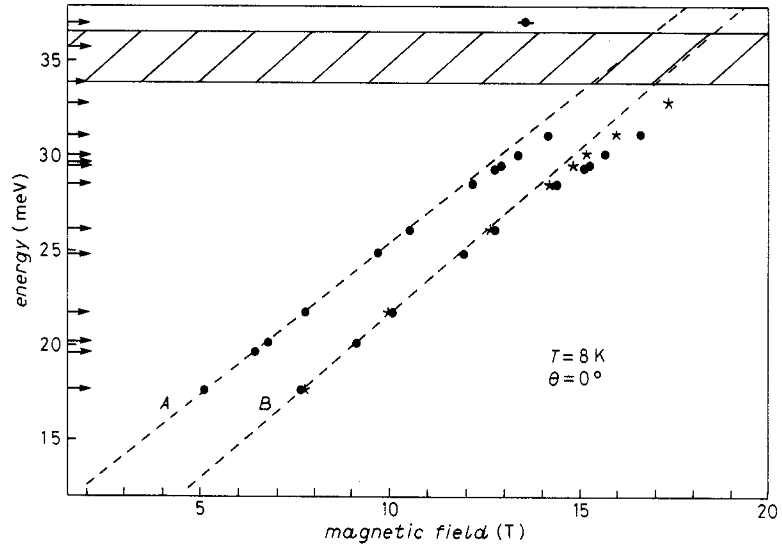

Fig. 3. – Energy dependence of the resonant field for the A and B (full points) $1s(z_i) \rightarrow 2p^+$ transitions, together with the bulk data (stars) of ref. [20]. The reststhralen band is visualized by the hatched zone. The arrows point out the energies of the laser lines we used. For some of them close to the phonon energies, no resonance could be detected.

position of the transmission minimum (in MT experiments) might not correspond to the field position of the quantum transition due to the resonant behaviour of the dielectric function at the TO-phonon energy, $\hbar\omega_{\text{TO}}$. In field-sweep MT measurements [8] and simulations [10] of the cyclotron resonance in GaInAs-based single heterojunctions, small kinks extending over a few wave numbers are revealed around $\hbar\omega_{\text{TO}}$ in graphs like that in fig. 3. Since our experimental data were obtained at least 3 meV below $\hbar\omega_{\text{TO}}$ they are not obscured by the function response and are reliable. Thus, a natural interpretation of fig. 3 is that polaron pinning of the $1s \rightarrow 2p^+$ transitions to the LO-phonon energy $\hbar\omega_{\text{LO}}$ (¹) of GaAs is observed. The experimental point above $\hbar\omega_{\text{LO}}$ give further support to our interpreation since its field position is consistent with the hybridization of the transition into two polaron branches, which is typical of a resonant polaron coupling [2].

It is worth comparing the magnitude of the resonant Fröhlich interaction in the MQW with that in the bulk. This cannot be made quantitatively because calculations of quasi-2D bound resonant polarons are not yet available. Moreover, it is likely that the band-nonparabolicity contributes significantly [22], making the discussion more complex. From cyclotron-resonance experiments and calculations in the bulk and in 2D, both polaron and nonparabolicity contributions are known to be comparable up to $E = 25$ meV. However, information on the relative strengths of resonant polarons can be gained by examining deviations of the transition energies from straight lines drawn with slopes equal to the experimental slopes at $E = 25$ meV where resonant polarons are not yet seen [20, 22]. Then, from fig. 3 it is obvious that resonant bound polarons in the bulk and in the well centres are very similar, but that the resonant polaron effect for the «deeper» barrier donors is stronger, typically twice. This finding is particularly interesting since it is confinement

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(¹) Optical phonons in GaAlAs have higher energies and high DOS confined phonons in GaAs have energies very close (within 3 cm^{-1}) to $\hbar\omega_{\text{LO}}$ for $L_{\text{w}} = 100$ Å [21].

related, as will be sketched now. The resonant polaronic correction to the energy of the $2p^+$ state can be evaluated within second-order Wigner-Brillouin perturbation theory. At the crossing point of the unperturbed levels, it reads

$$\Delta E(z_{\mathrm{i}}) = \frac{1}{\pi}\sqrt{\frac{\alpha}{2}}\left(\int_{-\infty}^{+\infty} \frac{\mathrm{d}^3 q}{q^2}\,|\langle 1s(z_{\mathrm{i}})|\exp[-i\boldsymbol{q}\cdot\boldsymbol{r}]\,|2p^+\rangle|^2\right)^{1/2}, \tag{1}$$

where z_{i} is the impurity location (the $2p^+$ state is roughly degenerate with z_{i} [12]), α is the Fröhlich constant ($\alpha = 0.07$ in GaAs), $\boldsymbol{q}$ is the phonon wave vector and $\boldsymbol{r}$ is the electron position operator. In eq. (1), polaron units [1, 2] were used. From eq. (1) it can immediately be seen that ΔE is z_{i}-dependent because of the z_{i}-dependence of the 1s state. Similarly, ΔE depends upon the MQW parameters (L_{w}, L_{b}) because the impurity properties depend upon all these parameters [12, 13]. In order to be more quantitative, let the wave functions be written as [23]

$$\langle \boldsymbol{r}|1s(z_{\mathrm{i}})\rangle = \chi(z)\,f_{1s}(z_{\mathrm{i}}, \rho)\,; \qquad \langle \boldsymbol{r}|2p^+\rangle = \chi(z)\,g_{2p^+}(\rho)\,, \tag{2}$$

where $\chi(z)$ is the envelope function of the ground electric subband in the GaAs well and $\rho = \sqrt{x^2+y^2}$. In eq. (2) it is assumed that the impurity states have their z motion forced by the confining potential. The radial envelope functions may be thought of either as hydrogeniclike or Landau-like trial wave-functions, depending upon the magnetic field strength [23]. We have checked that hydrogeniclike trial functions with the «effective Bohr radius» treated as a variational parameter are quite sufficient for on-well centre impurities at zero-field. Thus such a drastic motion separation as in eq. (2) should not change the physical meaning of the present approach. We have (eq. (3))

$$|\langle 1s(z_{\mathrm{i}})|\exp[-i\boldsymbol{q}\cdot\boldsymbol{r}]|2p^+\rangle|^2 = |\langle\chi|\exp[-iq_z z]|\chi\rangle|^2 \times |\langle f_{1s}|\exp[-i\boldsymbol{q}_{\parallel}\cdot\boldsymbol{\rho}]|g_{2p^+}\rangle|^2 \tag{3}$$

with $\boldsymbol{q}_{\parallel} = (q_x, q_y)$. The second term on the right side in eq. (3) depends strongly upon z_{i}. This can be further estimated using the dipole approximation

$$|\langle f_{1s}|\exp[-i\boldsymbol{q}_{\parallel}\cdot\boldsymbol{\rho}]|g_{2p^+}\rangle|^2 \simeq (q_x^2 + q_y^2 + 2q_x q_y)|\langle f_{1s}|x|g_{2p^+}\rangle|^2 . \tag{4}$$

It is remarkable that the oscillator strength for the dipole-excited $1s(z_{\mathrm{i}}) \to 2p^+$ transition appears in eq. (4). It has been calculated in ref. [13]: for interface impurities (which have a binding energy very close to that of our «deeper» barrier impurities) it is three times higher than for on-well-centre impurities. This results in a stronger resonant polaron effect and is in qualitative agreement with our experimental findings. In view of this, one should expect resonant polarons for isolated barrier impurities to be even stronger because the oscillator strength is much higher (ten times) than for on-well-centre donors. Although the model sketched above is very simplified, we think that it entails the remarkable feature that the properties of resonant polarons—like those of the bare impurities—can be monitored in MQW structures by the impurity location and also the MQW parameters.

In conclusion, quasi-2D resonant bound polarons have been observed for the first time. It has been shown that their strength can be leisurely monitored, which is a spectacular confinement effect.

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