Depolarization shift of the in-plane polarized interlevel resonance in a dense array of quantum dots

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We have investigated experimentally the midinfrared normal-incidence response of holes confined in an array of Ge/Si self-assembled quantum dots. The dots have a lateral size of about 15 nm and a density 3 $\times 10^{11}$ cm^{-2}. An in-plane polarized absorption in the 70–90 meV energy range is observed and attributed to the transition between the first two states in the dots. As the hole concentration in the dot ground state is increased, the absorption peak shifts to higher energies, its linewidth is reduced, and the line shape is changed from asymmetric to symmetric. We attribute all features to a depolarization-type effect similar to the case of two-dimensional systems. We believe that our results provide experimental evidence for the dynamic screening of an external field by in-plane polarized interlevel collective excitations.

The motion of the electrons in a two-dimensional (2D) system is quantized in the direction z perpendicular to the plane of the layer. The z-polarized infrared radiation can be absorbed by a 2D system causing interband transitions in the well. It is well known that in a dense 2D plasma, the collective intersubband charge-density excitations (resonant screening) lead to a density-dependent blueshift of the interband resonance away from its single-particle position[1] and to a resonance line narrowing for a nonparabolic system.[2] The renormalization of the intersubband absorption energy due to collective interactions is called the depolarization shift.[3]

In contrast to 2D systems, additional lateral confinement of carriers in quantum dots makes in-plane polarized transitions between the discrete levels possible. Recently, Metzner and Döhler[4] have pointed out that the dynamic many-particle interaction may also be effective between electrons oscillating in the lateral direction. The very promising candidates for the observation of in-plane polarized collective excitations are arrays of self-assembled quantum dots (SAQDs). However, for typical growth conditions, the sheet dot density ($<10^{10}$ cm^{-2}) is too low to observe the interdot correlation effects, which require the carrier density more than 10^{11} cm^{-2}. Previously we have studied hopping conduction in arrays of Ge/Si SAQDs, whose density was about 3 $\times 10^{11}$ cm^{-2}.[5] Such a high value has been achieved by reducing the Ge growth temperature and increasing the Ge growth rate during the molecular-beam epitaxy of the Ge/Si heterostructures. We have found that the temperature dependence of the lateral conductance as well as the typical hopping energy are determined by long-range interdot Coulomb interaction.[5] It is reasonable to expect that such a system could display the dynamic interaction effect as well.

The purpose of the present work is to examine directly the role of collective coupling for the in-plane polarized interlevel absorption of a dense array of Ge/Si SAQDs. Ge/Si(001) quantum dots exhibit a type-II band lineup. The large (~ 0.7 eV) valence-band offset characteristic of this heterojunction leads to an effective confinement of holes in the Ge region. The interlevel absorption studied here corresponds to the transition from the hole ground state to the first excited state in the dot; the states are associated with in-plane confinement and belong to the same z subband. The single-particle level spacing between these states has been previously found to be 70–80 meV.[6] Holes in the dots were generated by intense interband optical pumping. We find that as the pump intensity is increased, the absorption resonance moves to higher energies, the absorption line becomes narrower, and the line shape is changed from asymmetric to a Lorentzian form. To our knowledge, this is the first experimental evidence of the depolarization effect in arrays of SAQDs.

The sample was grown by molecular-beam epitaxy on a (001) oriented phosphorus-doped Si substrate with a resistivity of 5 Ω cm. It consists of 10 layers of Ge dots separated by 10-nm Si barriers. The whole structure was covered by a 20-nm Si cap layer. The Sb-doping level of the Si layers was $\approx 10^{16}$ cm^{-3}. Each Ge dot layer consists of a nominal Ge thickness of 10 ML (~ 14 Å) and was grown at 300 °C at a growth rate of 0.2 ML/s. The Si barriers were grown at 500 °C with temperature ramps before and after Ge growth. The deposition of Ge was monitored *in situ* by reflection high-energy electron diffraction (RHEED); the transition from a streaked to a spotty RHEED pattern, indicating the spontaneous formation of Stranski-Krastanov Ge islands, was observed after the deposition of about 5-ML-thick Ge. Structural characterizations of the Ge dots were performed by scanning tunneling microscopy[7] (STM) and also by cross-sectional transmission electron microscopy.[8] For the STM measurements, a reference sample with only one Ge dot layer without a Si cap was grown under the same growth condition. The SAQDs are found to be dense and small-sized. The average size of the dot base length is found around 15 nm, the height is ≈ 1.5 nm, and the dot uniformity is about $\pm 20\%$. The areal density of the dots is 3 $\times 10^{11}$ cm^{-2}, i.e., from one to three orders of magnitude higher than that reported previously in interlevel absorption experiments.[9]

In the dark, the dots contain no holes and the interlevel transitions are blocked. To provide holes for interlevel absorption, the sample was illuminated by a halogen lamp with
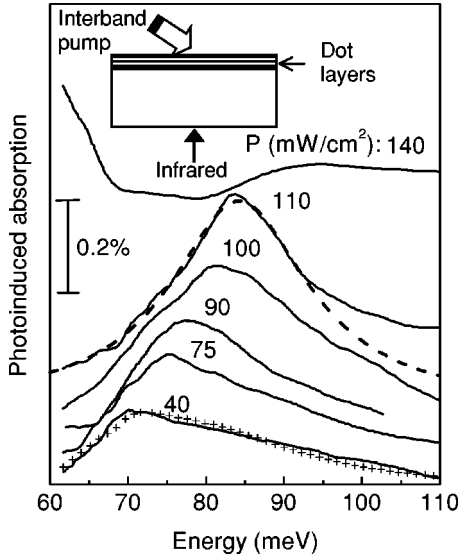
FIG. 1. The measured photoinduced absorption at various pump intensities. A constant offset is added for clarity. The crosses show the inhomogeneously broadened absorption calculated with Eqs. (1) and (2) with $\gamma = 1.25$ (20% dot size homogeneity). The dashed line depicts the result of fitting the high-intensity absorption line with a Lorentzian function (3). The inset shows schematically the experimental setup.

a 70-W output power modulated around 1.7 Hz for lock-in detection purposes. The pumping beam was filtered with a 0.35–0.6-μm bandpass filter and has a maximum intensity at 1.85 eV. The transmitted midinfrared radiation supplied by a globar source was measured using a bolometer with the lock-in technique. The photoinduced absorption spectra were taken in a single-pass normal incidence geometry at room temperature (Fig. 1) and in the pump intensities range of 40–140 mW/cm² (measured by a Si photodiode). The raw spectra were normalized by the reference spectrum recorded without interband optical pumping.

Let us estimate the effect of interband pumping on the dot occupation with holes. The hole density photogenerated directly in the Ge layers can be estimated as $n = G\tau_{Ge}$, where $G = \alpha P/h\nu$ is the generation rate, α the absorption probability, P the pump intensity, $h\nu$ the photon energy, and τ_{Ge} the interband recombination lifetime in Ge SAQDs. For $\alpha = 1.5 \times 10^{-4}$,[10] $P \approx 100$ mW/cm², $h\nu = 1.85$ eV, and $\tau_{Ge} = 3.9\ \mu$s,[11] the carrier density is $n = 2 \times 10^8$ cm⁻², which is about two orders of magnitude less than the dot density. Note, however, that when the sample is illuminated, electrons and holes are photogenerated in the whole structure (the penetration of the 0.6-μm wavelength light is approximately 2 μm in Si). The nonequilibrium holes diffuse from the Si bulk towards the Ge SAQDs and are accumulated in the dots. Since the hole diffusion length in Si [$10^2 - 10^3\ \mu$m (Ref. 12)] is much larger than the light penetration length, almost all photogenerated holes can reach and be captured by the SAQDs; then the injected hole density in the dots is given by a simple equation $n = P\tau_{Si}/h\nu$, where τ_{Si} is the carrier recombination lifetime in silicon. Measurements of τ_{Si} in a reference sample containing no Ge dots with time-resolved free-carrier absorption give $\tau_{Si} = 15\ \mu$s, in agreement with data reported in the literature.[13] Assuming P

$= 100$ mW/cm², $h\nu = 1.85$ eV, and $\tau_{Si} = 15\ \mu$s, we find $n = 5.1 \times 10^{12}$ cm⁻². For a 3×10^{11} cm⁻² SAQD density in each Ge layer, it yields a dot occupation number of about 2 and corresponds to approximately complete filling of the dot ground state (two holes per dot).

The photoinduced infrared absorption spectra measured at different pump intensities are reported in Fig. 1. An absorption peak is found at 70–90 meV, which is believed to come from the interlevel transition from the hole ground state to the first excited state in the dots [the $0 \rightarrow 2$ transition should appear at higher energies (at ≈ 140 meV) (Ref. 14)]. The free-carrier absorption associated with carriers that are not trapped in the dots is also observed at the highest pump intensity as an increasing absorption towards low energy. At low intensity, the peak is strongly asymmetric: the absorption increases rapidly on the low-energy side of the spectra and it gradually decreases on the high-energy side. Qualitatively, the asymmetric line shape can be understood from the following simple consideration. In the absence of collective effects, the interlevel transition is inhomogeneously broadened. The dominant broadening arises from fluctuations in the dot's size L, which give rise to a Gaussian distribution

$$D(L) = \frac{1}{\sqrt{2\pi}\sigma} \exp[-(L-L_0)^2/2\sigma^2], \qquad (1)$$

where L_0 is the average quantum-dot size. The level-spacing distribution $D(E)$ is related to $D(L)$ by $D(E) \propto D(L)(dE/dL)^{-1}$. Assuming that the interlevel spacing decreases with increasing the dot's size as $1/L^\gamma$, one obtains

$$D(E) \propto D(L)/E^{(1+\gamma)/\gamma}. \qquad (2)$$

To simulate the low-intensity absorption line, we fix the position of the absorption maximum, its intensity, and $\sigma/L_0 = 20\%$, then take γ as a fit parameter, and assume for simplicity the energy-independent oscillator strength.[15] The result for $\gamma = 1.25$ [Fig. 1 (crosses)] can be seen to reproduce the measured absorption spectrum very well.

Note that the photoinduced infrared absorption has to be proportional to the hole density in the ground state of the dots. Thus one expects the integrated absorption to follow a linear dependence on pump intensity, which is a signature of the linear absorption regime in the Si layer. We do observe such linear behavior at $P < 140$ mW/cm² in Fig. 2(a).

As the pump intensity increases, the absorption peak shifts to higher energies and becomes more symmetric, considerably reducing its linewidth [Figs. 2(b) and 2(c)]. Once P exceeds 110 mW/cm², the absorption linewidth tends to increase probably due to screening of dynamic interdot interaction by photogenerated free carriers. All these features are a signature of the collective phenomena,[2] here realized in a system of laterally confined states.[16]

It should be mentioned that in an electronic quantum dot with a parabolic confinement potential, the position of absorption peaks depends neither on the number of confined electrons nor on the presence of the static interaction between them because of the separability of the center-of-mass motion from the relative motion (generalized Kohn theorem).[17] However, a nonparabolic system or a quantum dot with holes can exhibit a deviation from the Kohn
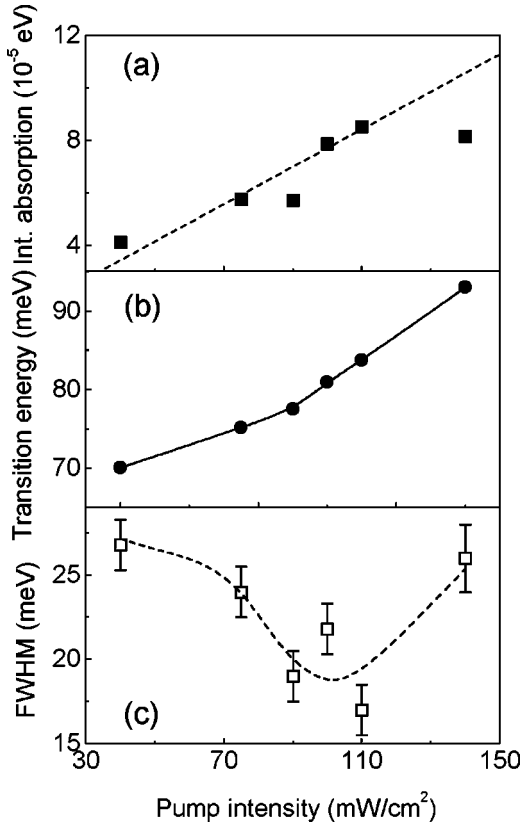
FIG. 2. Integrated absorption (a), resonance position (b), and the full width at half maximum (FWHM) (c) vs pump intensity.

theorem.[18] In this case, when the charge density in the ground state increases, the static Coulomb interaction would shift the ground-state energy towards the continuum and, hence, there should be a redshift in the transition energy, in contrast to our observation. We thus conclude that the observed energy shift is not due to charging the dots.

In the collective mode, the resonance should be homogeneously broadened[2] with the linewidth determined mainly by scattering mechanisms and not by fluctuations in the SAQD size. Therefore, the absorption can be expressed as

$$A = \frac{fNne^2\hbar}{2m_0\Gamma n_r c\epsilon_0} \frac{1}{1+[(E-E_0)/\Gamma]^2}, \qquad (3)$$

where f is the oscillator strength, N the number of dot layers, n the density of holes in the ground state of the dots, m_0 the free-electron mass, Γ the dephasing rate, n_r the index of refraction in the IR region of interest, ϵ_0 the vacuum permittivity, and c the speed of light. We fit the data at $P = 110 \, \text{mW/cm}^2$ to the functional form of Eq. (3) (dash line)

and extract a linewidth of $2\Gamma = 17 \, \text{meV}$ corresponding to an excited-state lifetime of $0.8 \times 10^{-13} \, \text{s}$, and an oscillator strength $f = 0.95$ that corresponds to an equivalent dipole length of 0.7 nm for an in-plane oriented interlevel transition with an energy of 70 meV.

For the theoretical analysis of the polarization selection rule and the dipole matrix element of the interlevel transition in the valence band of Ge/Si SAQDs, we have used the semi-empirical tight-binding method with the sp^3s^* atomic orbital basis. The strain distribution calculated using the valence force-field model has been taken into account.[19] The calculations were performed for a pyramid-shaped quantum-dot geometry. The full details of the calculations are beyond the scope of this paper and will be presented elsewhere. We would like here only to note that the transition between the ground and the first excited state is predicted to be in-plane polarized and characterized by an oscillator strength of 0.7 and a dipole length of 0.9 nm, in reasonable agreement with the experimental results.

Also from the magnitude of interlevel absorption (4 $\times 10^{-3}$), we can derive an experimental value of the absorption cross section for one layer of the dots. The absorption cross section for light polarized in the lateral direction is found to be $8 \times 10^{-16} \, \text{cm}^2$, which is comparable to the value achieved for an interlevel transition in the valence band of InAs/GaAs SAQDs ($1.6 \times 10^{-16} \, \text{cm}^2$).[20]

In summary, we have studied the interlevel photoinduced absorption of a dense array of Ge self-assembled quantum dots. As the carrier concentration in the dots increases, a blueshift of the transition from the hole ground state to the excited state, accompanied by narrowing the absorption peak, is observed. We argue that these observations represent, to our knowledge, the first experimental manifestation of a depolarization effect in arrays of interacting quantum dots. Unfortunately, there are no theoretical studies of the collective effects for lateral polarization so far, which prevents a comparison of the experimental data with theoretical results. Therefore, a complete and consistent theory is desirable. We hope that our result will stimulate theoretical investigations of the collective effect in systems with lateral carrier confinement.

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